Analysis of tensioned-web-over-slot die coating

A DISSERTATION
SUBMITTED TO THE FACULTY OF THE GRADUATE SCHOOL
OF THE UNIVERSITY OF MINNESOTA
BY

Jaewook Nam

IN PARTIAL FULFILLMENT OF THE REQUIREMENTS
FOR THE DEGREE OF
DOCTOR OF PHILOSOPHY

Marcio S. Carvalho, L. E. Scriven, Advisors

December 2009
Acknowledgments

First of all, I offer my sincerest gratitude to my advisor Marcio S. Carvalho, who support me throughout my thesis with his patience and knowledge. Even though he was not in Minneapolis, he leads and guides me to the right direction in my researches though weekly research meetings. Without his insights and advices, I could not able to finish my work on time.

I thank my parents, Youn-hee Nam and Ok-gi Byun, and my one and only brother, Jaeson, for their consistent helps during my graduate study from far away. I also thank my Korean friends in CEMS for their supports on my life in Minnesota.

I am indebted to many people who helped me during my study, including Dr. Juan de Santos and Dr. Takeaki Tsuda for their guides on my early computational research, Engineer Wieslaw Suszynsky for his helps in flow visualization experiments, Dr. Scott A. Walker at Imation for his fruitful comments and helps on tensioned-web coating process. Prof. Daniel D. Joseph for his helps on two-layer stability analysis, Shuzo Fujigami for his useful comments on my researches, Profs. Satish Kumar and Lorraine Francis for their helps on my researches.

I would like to thank my coating flow research colleagues in the Coating Process Fundamentals Program at the University of Minnesota, including Dan O’Neal, Benson Tsai, Shawn Dodds and Damien Brewer. And I specially thank to Kristanto Tjiptowidjojo and Alex J. Lee who always stimulate me with their fruitful discussions. I also appreciate helps from Japanese industrial fellows, including Makoto Komatsubara, Hiroshi Yoshiha, Hiroaki Kobayashi, Tomohiro Matsuda, and Yoshifumi Morita.

Finally, I dedicate this thesis to my advisor the late L. E. “Skip” Scriven. His continual insistence on perfection in my researches led me throughout the graduate study. He deeply involved in early part of my works but unfortunately he could not see their completion. I’d like to finish with my graduate study and start my new career with his favorite word.

“Excelsior!”
Abstract

In tensioned-web-over-slot die (TWOSD) coating, web is sustained only by the tension of the web wrapped around the coating die. The distance between the die and web is set by the interaction between the hydrodynamic force of coating liquid and the normal stress resultant from the curved tensioned web, in what is called elasto-hydrodynamic interaction.

In order to analyze this particular the coating flow, several tools are developed and tested on other coating flows relatively simpler than TWOSD flow. The theoretical conditions for the onset of vortex that degrades product qualities are proposed and the critical vortex birth trajectories inside the parameter space are tracked automatically by a direct tracking method. To detect a defect-causing flow feature in multi-layer coating, mid-gap invasion, the position of an interlayer separation point was tracked by theoretical model. The results are verified by flow visualization experiment for two-layer fixed-gap slot coating. Also linear stability analysis was performed, in order to detect unstable interlayer that signals coating defects in the two-layer slot coating.

The purpose of the research is to understand the complicated flow characteristic inside the coating bead by solving the two-dimensional Navier-Stokes theory using finite element method and visualizing the coating bead flow on a lab-scale TWOSD coater. Using the tools described above, the flow features that leads to coating defects, such as, bead breakup, weeping, mid-gap invasion and feed slot vortex, are identified and mathematical forms of the onset condition for the features are presented. The onset conditions are combined into the direct tracking method that was used to construct the vortex-free operating window for the given die lip configurations. Furthermore, the tracking method can be used to shows the effect of die lip design on the critical parameters for the onset of the flow features.
Contents

Acknowledgments i

Abstract ii

Table of Contents iii

List of Tables x

List of Figures xii

1 Introduction 1

1.1 Background ......................................................... 1

1.2 Survey of tensioned-web coating methods ......................... 5

1.2.1 Tensioned-web under a puddled knife ....................... 6

1.2.2 Tensioned-web-over-roll coating ............................. 8

1.2.3 Tensioned-web-over-microgravure-roll coating ............... 11

1.2.4 Tensioned-web-over-slot die coating ....................... 13

1.2.5 Two-layer tensioned-web-over slot die coating .......... 17
## CONTENTS

3.3.2 Half-submerged-forward roll coating ........................................... 79

3.4 Final Remarks ................................................................. 87

4 Mid-gap invasion in two-layer slot coating .............................. 89

  4.1 Introduction ................................................................................. 89

  4.2 Mid-gap invasion: flow visualization ........................................ 92

  4.2.1 Experimental set-up ................................................................. 93

  4.2.2 Onset of mid-gap invasion ....................................................... 94

  4.3 Mid-gap invasion: Navier–Stokes theory .................................... 98

  4.3.1 Governing equation and boundary conditions ....................... 98

  4.3.2 Solution of the Navier–Stokes system by G/FEM ................. 104

  4.3.3 Designing a mesh to track the interlayer separation point ....... 105

  4.3.4 Results from numerical model ............................................... 109

  4.4 Mid-gap invasion: a simple model and 1/3 rule ...................... 116

  4.4.1 Simple flow representation near mid lip ............................ 117

  4.4.2 1/3 Rule, a simple criterion for onset of mid-gap invasion ... 119

  4.5 Conclusions .......................................................................... 120

5 Linear stability analysis of two-layer fixed-gap slot coating .......... 122

  5.1 Introduction .............................................................................. 122

  5.2 Linear stability analysis of viscous coating flow ....................... 125

  5.2.1 Formulation of equations ..................................................... 125
# CONTENTS

5.2.2 Discretization by G/FEM ........................................... 129

5.2.3 Filtering eigenvalues at infinity ................................. 133

5.2.4 Flow rate ratio continuation ..................................... 138

5.3 Results & Discussion .................................................. 139

5.3.1 Construction of the neutral curve ............................... 141

5.3.2 Finding stable flow rate ratio range ............................ 144

5.3.3 Critical flow rate ratio condition for mid-gap invasion ... 146

5.3.4 Finding desirable viscosity ratio range ........................ 148

5.4 Conclusion ............................................................. 150

6 Computational analysis of single-layer TWOSD coating flow 153

6.1 Introduction ............................................................ 153

6.2 Elastohydrodynamic model and solution method ............. 156

6.2.1 Governing equations and boundary conditions .......... 156

6.2.2 G/FEM solution of TWOSD flow ............................... 163

6.2.3 Direct tracking of flow feature ................................ 167

6.2.4 Vortex-free operating window construction ................ 171

6.2.5 Die lip configurations ............................................ 174

6.3 Results & Discussion .................................................. 178

6.3.1 Effect of die lip geometry on operating window .......... 178

6.3.2 Feed slot vortex birth condition ............................... 187

6.3.3 Minimum wet thickness ......................................... 189
CONTENTS

6.4 Final remarks .................................................. 196

7 Flow visualization of single-layer TWOSD coating 198

7.1 Introduction ................................................... 198

7.2 Flow visualization — setup .................................. 200

7.3 Flow visualization results .................................. 204

7.4 Comparison: computational model vs. experiment .......... 210

7.5 Conclusion ..................................................... 212

8 Computational analysis of two-layer TWOSD coating 214

8.1 Introduction ................................................... 214

8.2 Elastohydrodynamic model and solution method .......... 219

8.2.1 Governing equations and boundary conditions ............ 219

8.2.2 Solution of the Navier–Stokes / shell system by G/FEM ... 228

8.2.3 Limit flow states in two-layer TWOSD coating flow .......... 229

8.2.4 Construction of vortex-free operating window ............ 231

8.3 Result & Discussion ......................................... 234

8.3.1 Minimum thickness in two-layer TWOSD coating .......... 234

8.3.2 Mid-gap invasion in two-layer tensioned-web-over-slot die coating 236

8.3.3 Effect of operating conditions on vortex-free operating window 238

8.3.4 Effect of die lip geometry on operating window ............ 252

8.4 Final remarks ................................................ 277
<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>9  Epilogue</td>
<td>281</td>
</tr>
<tr>
<td>Bibliography</td>
<td>285</td>
</tr>
<tr>
<td>A  Rounded corner mesh generation</td>
<td>295</td>
</tr>
<tr>
<td>A.1 Smooth node distribution along rounded corner of die lip</td>
<td>295</td>
</tr>
<tr>
<td>A.1.1 Patching straight line and arc of circle</td>
<td>295</td>
</tr>
<tr>
<td>A.1.2 Node distribution</td>
<td>297</td>
</tr>
<tr>
<td>A.1.3 Coupling with elliptic mesh generation system</td>
<td>306</td>
</tr>
<tr>
<td>A.2 Example: downstream slot coating — Low flow limit</td>
<td>310</td>
</tr>
<tr>
<td>A.2.1 Results for downstream slot coating with rounded corner</td>
<td>314</td>
</tr>
<tr>
<td>B  Inter-regional mesh boundary condition for structured mesh</td>
<td>320</td>
</tr>
<tr>
<td>B.1 Mesh boundary conditions for region interfaces</td>
<td>320</td>
</tr>
<tr>
<td>B.1.1 Example of pentagon lid-driven cavities</td>
<td>321</td>
</tr>
<tr>
<td>B.1.2 Boundary condition for inter-regional boundary</td>
<td>323</td>
</tr>
<tr>
<td>B.1.3 Residual equations for inter-regional mesh boundary condition</td>
<td>330</td>
</tr>
<tr>
<td>B.1.4 Corner condition at internal five region-confluence</td>
<td>330</td>
</tr>
<tr>
<td>C  Filtering eigenvalues at infinity for two-layer plane flow</td>
<td>332</td>
</tr>
<tr>
<td>C.1 Introduction</td>
<td>332</td>
</tr>
<tr>
<td>C.2 Filtering eigenvalues at infinity</td>
<td>334</td>
</tr>
<tr>
<td>C.2.1 The algorithm</td>
<td>335</td>
</tr>
</tbody>
</table>
CONTENTS

C.2.2 Recover original generalized eigenvector . . . . . . . . . . . . . . . . . . . . . . 341
C.2.3 An example: the two-layer flow system with four elements . . . . . . . . 342
C.2.4 Compare with full QZ method . . . . . . . . . . . . . . . . . . . . . . . . . . . 343
C.2.5 Effect of elements on eigenvalue . . . . . . . . . . . . . . . . . . . . . . . . . 347
C.2.6 Compare with numerical results . . . . . . . . . . . . . . . . . . . . . . . . . 349
C.3 Final remarks . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 351
## List of Tables

<table>
<thead>
<tr>
<th>Table</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.1</td>
<td>Summary of vortex birth</td>
<td>54</td>
</tr>
<tr>
<td>3.1</td>
<td>Parameters used in single-layer slot coating</td>
<td>73</td>
</tr>
<tr>
<td>3.2</td>
<td>Parameters used in half-submerged forward roll coating</td>
<td>81</td>
</tr>
<tr>
<td>4.1</td>
<td>Parameters used in two-layer dual slot coating</td>
<td>108</td>
</tr>
<tr>
<td>5.1</td>
<td>Dimensionless variables used in stability analysis</td>
<td>126</td>
</tr>
<tr>
<td>5.2</td>
<td>Mass and Jacobian entries</td>
<td>134</td>
</tr>
<tr>
<td>5.3</td>
<td>Mass and Jacobian matrix entries for boundary conditions</td>
<td>135</td>
</tr>
<tr>
<td>5.4</td>
<td>Base case physical parameters</td>
<td>141</td>
</tr>
<tr>
<td>6.1</td>
<td>Operating and geometric parameters for base case</td>
<td>160</td>
</tr>
<tr>
<td>6.2</td>
<td>Geometric parameters for die lip configurations</td>
<td>176</td>
</tr>
<tr>
<td>7.1</td>
<td>Operating conditions and parameters for flow visualization</td>
<td>204</td>
</tr>
<tr>
<td>8.1</td>
<td>Operating and geometric parameters for base case die lip configuration</td>
<td>223</td>
</tr>
<tr>
<td>8.2</td>
<td>Dimensionless number used in two-layer TWOSD study</td>
<td>224</td>
</tr>
</tbody>
</table>
# LIST OF TABLES

<table>
<thead>
<tr>
<th>Table Number</th>
<th>Table Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>8.3</td>
<td>Geometric parameters for die lip configurations</td>
<td>254</td>
</tr>
<tr>
<td>A.1</td>
<td>Operating parameter for downstream slot coating flow</td>
<td>311</td>
</tr>
<tr>
<td>B.1</td>
<td>Residual equations for inter-regional node</td>
<td>331</td>
</tr>
<tr>
<td>C.1</td>
<td>Dimensionless variables used in stability analysis</td>
<td>335</td>
</tr>
<tr>
<td>C.2</td>
<td>Comparing with results from QZ method</td>
<td>345</td>
</tr>
<tr>
<td>C.3</td>
<td>Comparison of most dangerous grow rates for different element numbers</td>
<td>347</td>
</tr>
<tr>
<td>C.4</td>
<td>Comparing with theoretical results</td>
<td>350</td>
</tr>
<tr>
<td>C.5</td>
<td>Comparing with results from spectral tau method</td>
<td>351</td>
</tr>
</tbody>
</table>
List of Figures

1.1 Various coating methods .................................................. 2
1.2 Typical tensioned-web-over-slot die coating .......................... 4
1.3 Various knife contours .................................................. 6
1.4 Tensioned-web under puddled knife coating system .................. 6
1.5 Simple spreader model for unyielding web and flexible web ....... 7
1.6 Tensioned-web-over-roll coating ........................................ 9
1.7 Tensioned-web-over-roll microgravure coating ....................... 12
1.8 Tensioned-web-over-slot-die coating .................................. 14
1.9 Single-layer tensioned-web-over-slot die coating patents .......... 16
1.10 Two-layer tensioned-web-over-slot die coating patents .......... 18
1.11 Slot coating window example ....................................... 23
1.12 Defect-causing flow features for TWOSD coating flow .......... 24

2.1 Pathline near a stagnation point inside flow ....................... 37
2.2 Evolution of eigenvectors at a stagnation point ..................... 38
**LIST OF FIGURES**

<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.3</td>
<td>A vortex birth at free surface</td>
<td>44</td>
</tr>
<tr>
<td>2.4</td>
<td>Vortex birth at a stationary solid</td>
<td>48</td>
</tr>
<tr>
<td>2.5</td>
<td>Onset of vortex birth at stationary solid (non-simple degenerate case)</td>
<td>50</td>
</tr>
<tr>
<td>2.6</td>
<td>Saddle point birth from a solid wall</td>
<td>51</td>
</tr>
<tr>
<td>3.1</td>
<td>Vortex in coating flows</td>
<td>56</td>
</tr>
<tr>
<td>3.2</td>
<td>Schematic procedure of direct tracking algorithm</td>
<td>69</td>
</tr>
<tr>
<td>3.3</td>
<td>Single-layer slot coating: boundary condition and mesh</td>
<td>72</td>
</tr>
<tr>
<td>3.4</td>
<td>Possible vortices in slot coating flow</td>
<td>74</td>
</tr>
<tr>
<td>3.5</td>
<td>Cusp point tracking results</td>
<td>76</td>
</tr>
<tr>
<td>3.6</td>
<td>Die lip vortex birth tracking results</td>
<td>78</td>
</tr>
<tr>
<td>3.7</td>
<td>Half-submerged-forward roll coating : boundary condition and mesh</td>
<td>80</td>
</tr>
<tr>
<td>3.8</td>
<td>Possible vortices in half-submerged forward roll coating flow</td>
<td>82</td>
</tr>
<tr>
<td>3.9</td>
<td>Symmetric vortex birth near film-splitting zone tracking results</td>
<td>84</td>
</tr>
<tr>
<td>3.10</td>
<td>Asymmetric vortex birth near film-splitting zone tracking results</td>
<td>85</td>
</tr>
<tr>
<td>4.1</td>
<td>Two-layer dual slot coating schematic diagram</td>
<td>90</td>
</tr>
<tr>
<td>4.2</td>
<td>Interlayer configuration for before and after mid-gap invasion</td>
<td>91</td>
</tr>
<tr>
<td>4.3</td>
<td>Experimental dual slot coating apparatus</td>
<td>93</td>
</tr>
<tr>
<td>4.4</td>
<td>Flow visualization of mid-gap invasion</td>
<td>95</td>
</tr>
<tr>
<td>4.5</td>
<td>Critical bottom-layer thickness at mid-gap invasion</td>
<td>97</td>
</tr>
<tr>
<td>4.6</td>
<td>Flow near interlayer separation point</td>
<td>101</td>
</tr>
</tbody>
</table>
LIST OF FIGURES

4.7  Boundary conditions for dual slot coating ............................................. 103
4.8  Two mesh topologies for the mid-gap invasion ....................................... 106
4.9  Interlayer separation point location at viscosity ratio $m = 1$ ....................... 110
4.10 Interlayer separation point location for different viscosity ratio .................. 111
4.11 Interlayer separation point location at viscosity ratio $m = 0.2$ ................. 113
4.12 Interlayer separation point location at viscosity ratio $m = 5$ ................. 115
4.13 Two mechanisms of mid-gap invasion .................................................. 117
4.14 Evolution of the rectilinear velocity profile under the mid lip .................... 118
5.1  Two-layer slot coating flow ................................................................. 123
5.2  Base flow configuration for the downstream coating gap flow ................. 125
5.3  The sequence of entries in residual vector ............................................. 132
5.4  Results from stability analysis of two-layer slot coating flow .................... 140
5.5  Most dangerous growth rate versus wavenumber plot ............................ 142
5.6  Neutral curve for density ratio $r = 1$ and viscosity ratio $m = 2$ ............. 143
5.7  Neutral curve for density ratio $r = 1$ and viscosity ratio $m = 2$ ............. 145
5.8  Neutral curve for density ratio $r = 1$ and viscosity ratio $m = 0.5$ .......... 147
5.9  Neutral curve for density ratio $r = 1$, flow rate ratio $f = 0.04$ ............. 149
5.10 Neutral curve for density ratio $r = 1$ and total wet thickness $h_{w,t}^* = 0.40$ 151
6.1  A schematic diagram of tensioned-web-over-slot die coating system .......... 154
6.2  Boundary conditions for single-layer TWOSD coating flow .................... 157
LIST OF FIGURES

6.3 Parameters for single-layer TWOSD coating flow .......... 161
6.4 Change of flow states as wet thickness increases .......... 168
6.5 Direct tracking algorithm ........................................ 172
6.6 Construction of vortex-free window for single-layer TWOSD coating flow ........ 173
6.7 Die lip configurations considered in this study .......... 175
6.8 Effect of lip length on vortex-free operating window .......... 177
6.9 Pressure profile along web for different lip length .......... 179
6.10 Pressure profile along web for different lip length at high speed .......... 180
6.11 Pressure profile at vortex birth for different lip length .......... 181
6.12 Effect of lip curvature on vortex-free operating windows .......... 182
6.13 Pressure profile for three different die lip curvatures .......... 183
6.14 Effect of apex point on vortex-free operating window .......... 184
6.15 Pressure profile along web for different apex point location .......... 185
6.16 Effect of straight die lip on vortex-free operating window .......... 186
6.17 Pressure profile along web for different straight lip length .......... 187
6.18 Pressure profile along feed slot wall at vortex birth .......... 188
6.19 Critical wet thickness for feed slot vortex birth .......... 190
6.20 Effect of inertia on feed slot vortex birth .......... 191
6.21 Minimum wet thickness with respect to geometric parameters .......... 193
6.22 Minimum wet thickness correlation .......... 194
6.23 Curve-fitting of minimum thickness correlation parameter .......... 195
### LIST OF FIGURES

<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.1</td>
<td>Defect causing flow features in TWOSD coating</td>
<td>199</td>
</tr>
<tr>
<td>7.2</td>
<td>Tensioned-web-over-slot die coating apparatus setup</td>
<td>201</td>
</tr>
<tr>
<td>7.3</td>
<td>The coating die used for the flow visualization</td>
<td>203</td>
</tr>
<tr>
<td>7.4</td>
<td>Camera setup and visualization area in coating bead</td>
<td>204</td>
</tr>
<tr>
<td>7.5</td>
<td>Flow visualization of ribbing instability</td>
<td>205</td>
</tr>
<tr>
<td>7.6</td>
<td>Flow visualization of bead breakup</td>
<td>207</td>
</tr>
<tr>
<td>7.7</td>
<td>Flow visualization of weeping</td>
<td>208</td>
</tr>
<tr>
<td>7.8</td>
<td>Flow visualization of feed slot vortex</td>
<td>209</td>
</tr>
<tr>
<td>7.9</td>
<td>Comparison between computational predictions and visualization results</td>
<td>211</td>
</tr>
<tr>
<td>8.1</td>
<td>Two types of two-layer TWOSD coating</td>
<td>215</td>
</tr>
<tr>
<td>8.2</td>
<td>Dual slot TWOSD coating model boundary conditions and parameters</td>
<td>220</td>
</tr>
<tr>
<td>8.3</td>
<td>Mesh of two-layer TWOSD coating flow model</td>
<td>227</td>
</tr>
<tr>
<td>8.4</td>
<td>Change of flow states as wet thickness changes</td>
<td>230</td>
</tr>
<tr>
<td>8.5</td>
<td>Coating window construction for two-layer TWOSD coating flow</td>
<td>233</td>
</tr>
<tr>
<td>8.6</td>
<td>Finding minimum top-layer wet thickness</td>
<td>235</td>
</tr>
<tr>
<td>8.7</td>
<td>Mid-gap invasion analysis for two-layer TWOSD</td>
<td>237</td>
</tr>
<tr>
<td>8.8</td>
<td>Pressure profiles for before and after mid-gap invasion</td>
<td>239</td>
</tr>
<tr>
<td>8.9</td>
<td>Effect of viscosity ratio on vortex-free operating windows</td>
<td>241</td>
</tr>
<tr>
<td>8.10</td>
<td>Effect of viscosity ratio on four critical flow states</td>
<td>242</td>
</tr>
<tr>
<td>8.11</td>
<td>Change of pressure profile near bead breakup states</td>
<td>243</td>
</tr>
<tr>
<td>8.12</td>
<td>Critical pressure profile near feed slot vortex birth</td>
<td>244</td>
</tr>
</tbody>
</table>
LIST OF FIGURES

8.13 Effect of tension on vortex-free operating windows .................................. 246
8.14 Effect of tension on four critical flow states ............................................. 247
8.15 Change of pressure profiles and web curvatures near mid-gap invasion .... 248
8.16 Effect of web speed on vortex-free operating windows ............................ 250
8.17 Effect of web speed on four critical flow states ........................................ 251
8.18 Re-plot of vortex-birth tracking results for web speed ............................. 252
8.19 Die lip configurations for two-layer TWOSD coating flow ...................... 253
8.20 Downstream lip radius change scheme .................................................... 255
8.21 Effect of downstream lip radius on vortex-free operating window ............. 257
8.22 Effect of downstream lip radius on four critical flow states ...................... 258
8.23 Change of pressure profile near mid-gap invasion .................................. 259
8.24 Change of pressure profile for upstream feed slot vortex ........................ 260
8.25 Mid lip radius change scheme ............................................................... 261
8.26 Effect of mid lip radius on vortex-free operating window ....................... 263
8.27 Effect of mid lip radius on four critical flow states .................................. 264
8.28 Change of critical pressure profiles near bead breakup ............................ 265
8.29 Change of critical pressure profile for upstream feed slot vortex ............. 266
8.30 Underbite downstream die lip change scheme ........................................ 267
8.31 Effect of mid lip radius on four critical flow states .................................. 268
8.32 Effect of underbite downstream die lip on vortex-free operating windows ... 269
8.33 Change of critical pressure profile near mid-gap invasion ....................... 270

xvii
<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>8.34</td>
<td>Mid lip apex point change scheme</td>
<td>273</td>
</tr>
<tr>
<td>8.35</td>
<td>Effect of mid lip apex point on vortex-free operating windows</td>
<td>274</td>
</tr>
<tr>
<td>8.36</td>
<td>Effect of mid lip apex point on four critical flow states</td>
<td>275</td>
</tr>
<tr>
<td>8.37</td>
<td>Change of critical pressure near mid-gap invasion</td>
<td>276</td>
</tr>
<tr>
<td>8.38</td>
<td>Change of critical pressure profile near bead breakup</td>
<td>278</td>
</tr>
<tr>
<td>A.1</td>
<td>Example of patched geometry</td>
<td>296</td>
</tr>
<tr>
<td>A.2</td>
<td>Dual grid strategy</td>
<td>301</td>
</tr>
<tr>
<td>A.3</td>
<td>Weighting function smoothing</td>
<td>302</td>
</tr>
<tr>
<td>A.4</td>
<td>Weighting function integration scheme</td>
<td>303</td>
</tr>
<tr>
<td>A.5</td>
<td>Geometry tracking</td>
<td>306</td>
</tr>
<tr>
<td>A.6</td>
<td>Evaluate arclength along rounded corner</td>
<td>308</td>
</tr>
<tr>
<td>A.7</td>
<td>Coupling patched geometry to G/FEM system</td>
<td>309</td>
</tr>
<tr>
<td>A.8</td>
<td>Schematic representation of downstream of the slot coating system</td>
<td>312</td>
</tr>
<tr>
<td>A.9</td>
<td>Slot coating mesh with different static contact angles</td>
<td>314</td>
</tr>
<tr>
<td>A.10</td>
<td>Mesh plots for the static contact line movement</td>
<td>316</td>
</tr>
<tr>
<td>A.11</td>
<td>Streamline plots for flow near the static contact line</td>
<td>317</td>
</tr>
<tr>
<td>A.12</td>
<td>Measuring the location of the static contact line</td>
<td>318</td>
</tr>
<tr>
<td>A.13</td>
<td>Static contact line arclength position</td>
<td>319</td>
</tr>
<tr>
<td>B.1</td>
<td>Pentagon lid-driven cavity example</td>
<td>322</td>
</tr>
<tr>
<td>B.2</td>
<td>Two types of subdomain connections</td>
<td>325</td>
</tr>
</tbody>
</table>
LIST OF FIGURES

B.3 Pentagon lid-driven cavity mesh ............................................. 329
B.4 Subdomain boundary numbering scheme ................................. 330
B.5 Comparison between two corner conditions ............................ 331
C.1 Degree of freedom numbering scheme for the stability problem .... 342
C.2 Non-zero entries in matrix related to first matrix transformation .... 343
C.3 Non-zero entries in matrix related to second matrix transformation ... 344
C.4 Comparison between three eigenvalue solving methods ............... 346
C.5 Ten most dangerous leading eigenvalues for different methods ...... 347
C.6 Modulus of disturbances in velocity and pressure ..................... 348
C.7 Ten most dangerous eigenvalues for different numbers of elements . 349
Chapter 1

Introduction

1.1 Background

In a coating operation, liquid is applied continuously to a moving substrate in order to produce a uniform liquid layer (Scriven, 2005b). The moving substrate, also called web, is usually flexible. Depending on the final product, the web can be made of different materials, for example:

- paper and paperboard (adhesive, impregnation)
- cellulose and polymer films (magnetic storage media, optical films, photographic)
- textile fibers and fabrics (application of finished)
- metal foils (polymer coating)

Numerous methods have been developed for depositing a liquid layer on a substrate. The wet thicknesses of the deposited layers, i.e. “thicknesses as coated”, range from $10 \mu m$ or somewhat less up to $300 \mu m$ and more, depending on the applications (Sartor, 1990). One simple common approach is to apply a large volume of coating liquid and then meter off the excess to obtain the desired uniform thickness and, in some
cases, oriented the micro-structure of the coated layer. This is called post-metering coating and occurs in blade, membrane, rigid knife and rod coating methods. The alternative is to apply just the desired amount of liquid as a uniform layer directly onto the substrate and remove none. This is called pre-metering coating and occurs in slot, slide and curtain coating method, which are the principal methods of continous precision coating. Figure 1.1 sketches some of these methods.

**Post-metered coating method**

(a) Rigid knife coating.  
(b) Flexible blade coating.  
(c) Rod, bar coating.

**Pre-metered coating methods**

(d) slide fed curtain coating.  
(e) Slide coating.  
(f) Slot die coating.

Figure 1.1: Schematic of various coating methods (From Scriven 2005b).

In rigid knife or rod coating, the lowest thickness of uniform coating is bounded by the mechanical limits on the uniformity of the clearance between the substrate, whose thickness may vary and the backup roll, with its run-out caused by out-of-roundness of
the roll surfaces, shaft mounting, and bearings. Flexible blade coating and membrane
coating can maintain a smaller gap clearance than is feasible with a rigid gap because
their metering action does not come from a fixed dimension but from the elasto-
hydrodynamic interaction between a deformable element and the coating liquid itself.
In post-metered coating methods, which control the wet thickness by metering off
the excess liquid, precise thickness control is difficult to achieve compared with pre-
metered coating method, at which the thickness is directly controlled by the flow rate,
usually issued from the feed slot of a coating die (Scriven, 2005a).

Slot, slide, and curtain coating are pre-metered methods by which more than one
liquid layer can be applied together, one atop the other, on a moving substrate. Slot
coating is practically limited to two layers due to machining of the die. It is extremely
difficult to assemble and build die blocks of more than three feed slots within 1 ∼ 2 mm,
(i.e. the typical length of a die lip surface. Historically, slide and curtain coating
methods were the method of choice for simultaneous multilayer coating, because of
the simplicity in the set-up, which consist of stacking-up of multiple, but the almost
identical, die blocks. The methods were developed in the photographic industry
and were later used for medical imaging, graphic arts, inkjet receptive, and other
products. However, as pointed out by Yih (1963), liquid flow down an inclined plane
is susceptible for the flow instability. Therefore, desirable die lip designs and ranges
of operating conditions are limited.

In slot coating, the gap height, the clearance between the substrate and the die
lip surface, is kept small so that the coating flow is well confined between the sub-
strate and the surfaces of the die lips. This feature keeps the area of the free sur-
faces smaller than other coating flows: it improves the resistance to external dis-
urbances. In general, web speeds for the slot coating range from 50 ∼ 100 ft/min
(0.25 ∼ 0.5 m/s) to 500 ft/min (2.5 m/s) (Sartor, 1990) and even substantially higher,
up to ∼ 10 m/s (Gates, 1999). The maximum speed is a strong function of the desired
thickness, liquid properties and coating gap.

Slot coating is also categorized as a high speed coating method, though the speeds
are not as high as has been achieved with slide and, especially, curtain coating. But
there is a variant of slot coating in which speeds in the same range of curtain coating
1.1. BACKGROUND

have been reached.

![Diagram of tensioned-web-over-slot die coating](image)

**Figure 1.2:** Typical tensioned-web-over-slot die coating (From Chino et al., 1988)

The tensioned-web slot coating — also called tensioned-web-over-slot-die (TWOSD) coating and sometimes free-span slot coating — is an elastohydrodynamic coating system. It exploits the interaction between the normal stress resultant from curved flexible web under tension and pressure inside the coating liquid. The typical configuration of TWOSD coating method presented in patents (e.g. Chino et al., 1988) as shown in Fig. 1.2. It is a pre-metered coating method, like slot coating, and also exploits the elastohydrodynamic interaction to maintain an extremely small gap height between the die lip surfaces and the moving web, like the flexible blade coating. In slot coating, the minimum wet thickness at a given die configuration is proportional to the gap height. Therefore the TWOSD coating can deliver ultra-thin layer with a precise wet thickness control.

This is the main reason of a success of the TWOSD coating in manufacturing magnetic media, that requires depositing particulate suspensions (dual-layer) film on a polymeric substrate moving at high speed: it is also used in other high speed applications and it is a strong candidate for a ultra-thin and nano-structured coating. The basic equipment is relatively straightforward to build, although the demands of accurate web movement and tension control in both the machine and cross-web directions can be severe. However, its ability to produce thin and uniform wet coatings at
high speed is remarkable. Most patents on tensioned-web coating, as Tomaru (1995), Shibata et al. (1996), and Shibata et al. (1987), claim their own die lip design principles. They also emphasized on the improved performance of the properly designed TWOSD coating system compared with the conventional slot coating. However, their qualities — thickness and surface uniformities — depend not only on the equipment design but also the properties of liquid and the process conditions. Hence there is a great need of fundamental understanding of this process, which requires a scientific approach to the TWOSD coating method. This is the main goal of this thesis.

1.2 Survey of tensioned-web coating methods

Tensioned-web coating is an elastohydrodynamic coating system where pressure and viscous stresses in the coating liquid are balanced by elastic stresses on the flexible web adjacent to the flow (Scriven 2005a). Elastohydrodynamic coating systems include flexible blades, membrane coaters, squeeze roll, gravure coaters, tensioned-web roll and slot coaters. Pranckh and Coyle (1997) described and analyze some of the most common elastohydrodynamic mechanism in coating systems.

The common feature of tensioned-web coating is that traction exerted by the liquid deforms a flexible substrate under tension. This deformation affects the flow between the web and the solid surface of roll or die. This feature helps to maintain a very small gap clearance with relatively small cost, because expensive precision machining of rolls, bearing and die mounting is not demanding in this system.

Therefore, it is worthwhile to examine the history of tensioned-web coating. Useful information about each method, especially the role of the elastohydrodynamic interaction on each coating flow is the focus of this survey.

In general, there are four major classes in the tensioned-web coating:

- Tensioned-web under a puddled knife.
- Tensioned-web over roll,
1.2. SURVEY OF TENSIONED-WEB COATING METHODS

![Figure 1.3: Various Knife contours (From Weiss 1977).](image)

- Tensioned-web over microgravure rod or roll,
- Tensioned-web over slot die.

1.2.1 Tensioned-web under a puddled knife

Tensioned-web under a puddled knife is the most simple case of tensioned-web coating system. Coating liquid is fed to the web directly behind the knife or rod. The web, either is a free span or supported by a blanket, moves at speeds of $10 \sim 300$ fpm (Booth 1970) and the excessive coating liquid is metered by a knife installed above the web. Figure 1.3 shows different types of knife used in industry. The basic system compositions are shown in Figure 1.4.

Usually, these systems employ a batch or intermittent feed to the puddle, which is generally confined laterally by some sort of end dams as in puddle coating (Shepherd 1970).
1.2. *SURVEY OF TENSIONED-WEB COATING METHODS*

![Diagram of spreader models](image)

(a) Uniform gap spreader  
(b) Membrane spreader

**Figure 1.5:** Simple spreader model for unyielding web and flexible web.

Liquid in the puddle necessarily recirculates to form a rolling bank behind the knife or bar. The puddle changes continuously as it is fed and depleted. Therefore, the method is not suitable to coat uniform liquid layer precisely.

The method can be analyzed by examining characteristics of knife or bar coating. The uniform gap spreader, which is shown in Fig. 1.5(a), provides a first but unrealistic approximation. A more realistic analysis can be done considering the bar or knife rigidly mounted closed to flexible web as in Figure 1.5(b).

Usually, the web is appreciably compliant, i.e. flexible. It may also extend lengthwise, contract crosswise, or compress thickness-wise, but those deformation modes are not nearly as unimportant as the bending. Therefore the simplest model for tensioned-web-under-puddled knife is the simple spreader model with membrane web modeled as a membrane which has no bending resistance. The gap depends on the elasto-hydrodynamic interaction between the curvature of the membrane and the pressure that the lubrication-type flow generates. This means the flow and the coating thickness are determined by the gap, which in turn is controlled by the wrap angle and tension on the web. Shear-sensitive behavior of the liquid is common can be incorporated in the model.

The knife and bar over tensioned web are also sometimes used for smoothing — no liquid removal, only redistribution, i.e. no change in the mean coating thickness. See Scriven (2005a) for details about smoothing and metering functions in coating process. This method is frequently used for highly viscous (but usually small amount of) liquid for relatively thin coating that does not require high precision, such as green
1.2. SURVEY OF TENSIONED-WEB COATING METHODS

1.2.2 Tensioned-web-over-roll coating

Tensioned-web-over-roll coating is relatively simple in a mechanical point of view and suffice for many coating application. This coating method is also called “kiss-roll” coating which use a single or multiple roller system to deposite a coating on the substrate without the use of impression or backup roller that creates a coating nip in regular multiple roll coating (Weiss 1977). The angle at which the web travels to and from the applicator is called wrap angle. Wrap angle is determined by the position of idler rolls. It is most often used to coat and impregnate porous, permeable webs with controlled amount of liquid. Since there is no back-up roll, liquid reaching the other side of web does not cause problems like in regular roll coating, for example fouling the gap between the roll surface and the web.

Continuous web can be coated by passing it over a turning roll that carries the liquid and transfers part of it to the web. The web must be in tension and must wrap the roll through at least a small angle, in order to provide the elastic boundary for the liquid flow and to impose load in the coating bead. Liquid can be dipped from a pool in a pan onto the surface of the roll, or applied by a blade-sealed distribution chamber, or even sprayed, as shown in Fig. 1.6.

The transfer can be by a “film split” between co-moving web and roll surface or by a “reverse wipe” between counter-moving web and roll surface. The tolerances required for roll surface, bearings, and mounting are generally much less than when it compared with regular roll coating methods.

Typical diameter of roll is from 30 in, in paper industry (Booth 1993), to 2 ~ 8 in in laboratory size roll coating, the range of radii can go down to rods which are few centimeters or even mm in diameter. The roll size is partly determined by the flexural rigidity $F = E I$, which is defined as the force required to bend a rigid structure to a unit curvature, where $E$ is Yound modulus and $I$ is moment of inertia (S.P. Timoshenko and Goodier 1973). The moment of inertia of cylindrical geometry with mass $M$,
1.2. SURVEY OF TENSIONED-WEB COATING METHODS

(a) Pan fed, direct with reverse transfer, i.e. wipe transfer

(b) Pan fed, direct with forward transfer, i.e. film split transfer

(c) Covered pan fed, “fountain feed”, two roll with reverse transfer

(d) Fed by blade-sealed pressurized distribution chamber, with forward transfer

Figure 1.6: Tensioned-web-over-roll coating.
radius $R$, and Length $L$ is $\frac{1}{4MR^2} + \frac{1}{3ML^2}$. As the length of the roll increase, larger diameter are required to prevent bending.

Unlike the tensioned-web under knife coating scheme described in Section 1.2.1, liquid is continuously fed by pipe or carried to the coater from the temperature-controlled storage. Sometimes the in-line preparation step is required to get rid of impurities by filtering. Usually feed methods are not critical to coating conditions; simple pumping and gravity feed are used commonly.

Frequently the distribution function is performed by a pan, as in Fig. 1.6(a). However, in this setup, it is easy to entrain bubbles in the liquid. Furthermore, the setup is also susceptible to contamination, evaporation and long residence time. To overcome these drawbacks of pan, a feeding covered pan, a sealed “fountain feed” and a distribution chamber were invented, as in Fig. 1.6(b). The covered pan is adopted for reducing the free liquid surface and the size of pan. Sometimes, a flexible blade is adopted as a metering device.

A distribution chamber of slot or die coater reduces the liquid free surface and the gravity leveling function, thus this is usually adopted with volatile or reactive coating liquids, as in Fig. 1.6(d). However, maintaining the uniform pressure difference across the cross-web direction for successful distributing requires properly designed slot die.

In a multiple roll system, one or both rolls may have compliant cover to prevent possible problems from a roll run-out and a roll clashing, and to suppress flow instability (Carvalho, 1996). The uniformity of the metering function depends on the uniformity of the pressure across the inflow side of the gap and uniformity of liquid mobility in the gap. For successful metering, the machining tolerance in bearing and mounting of a pair of rolls and the drive system, which cause metering rate variation due to the roll speed variation, are critical. Also the viscosity of the solution needs to be considered, if it strongly depends on temperature.

The application zone which is also known as the coating bead, is in general classified in two types: film split, where the web moves in the same direction as the roll, and “reverse wipe”, where the web moves in the opposite direction to the roll. These can be explained using pressurized forward-roll coating and pressurized reverse-roll
coating which are examined by [Benjamin (1994)]. The system with film split is almost similar to the forward-roll coating, but pressure of the coating bead is higher than in roll coating because of the elastohydrodynamic interaction. The same logic can be applied to reverse direction web with reverse-roll coating system. In terms of tensioned-web-over-roll coating, foil bearing can be considered as a special case, which has a stationary roll and moving web.

The variant of tensioned-web-over-roll coating are tensioned-web-over knurled roll and tensioned-web-over gravure cell. When the roll diameter is small, i.e. of the order of cm, these variants are called “microgravure coating”.

### 1.2.3 Tensioned-web-over-microgravure-roll coating

The coating bead of tensioned-web-over-microgravure-roll or rod coating is different from other tensioned-web coating. The coating liquid is provided from the grooves or cells not from a smooth surface or slot.

The continuous web can be coated by passing it over a turning rod that has its surface engraved or etched with a pattern of grooves or cells that enable thin coating to be created more readily than with smooth roll or rod. Tensioned-web-over microgravure-roll or rod coating is base on knurled roll, which has grooves, and gravure coating, which are self-metering coating processes. It means that the metering function depends strongly on the volume factor, the ratio of the volume of liquid in the cell on the knurled roll to surface area, i.e. the liquid per unit superficial area. This depends in turn on how the grooves or cells are refilled with liquid in each revolution of the rod. The other factor that affects the metering function is the speed difference between the web and the rod surface in the forward “film split” type system.

Additional measures, that can help improve smoothness, is to endow the patterned rod with subsidiary grooves that connect the cells. If the coating solution cannot level fast enough to the degree needed after it has been transferred to the web, the so called ribbing instability, which is the amplified interaction between frequency of the induced patterns, can occur. Smoothing is redistributing of the coated liquid that was non-uniformly deposited on the roll in order to make a uniform layer without
removing any liquid. A diagram of this type of system is shown Figure 1.7 which indicates how the action in the coating bead can be easily visualized if the web is transparent (From Hanumanthu, 1996).

As shown Fig. 1.7, a doctor blade is usually used in gravure coating. The purpose of the doctor blade is to remove the liquid from the unengraved portions of the engraved roll, i.e. it leaves only a thin layer of liquid on the land of a pattern or cell.

Typical size of the roll in microgravure coating is about $1 \sim 3\text{cm}$ (Hanumanthu, 1996). Because of the small dimension, microgravure and micro-knurled cylinders are easier to handle than the heavier, large-diameter knurled and gravure rolls. As pointed out in roll coating case, flexural rigidity of cylindrical geometry show that longer rods have to be stiffer. Rods are generally supported by some sort of rod holder, in which they are lubricated by the coating liquid itself.

The quality of the final coating is strongly influenced by the coating bead. This relationship is represented as an operating window which maps the coating quality
1.2. SURVEY OF TENSIONED-WEB COATING METHODS

as a function of the operating parameters: the speed ratio between the rod and the web, liquid viscosity, surface tension, etc (Hanumanthu, 1996). The operating window indicates that at some conditions coating defect are present. For example, down-web barring and a wavy mark in the cross-web direction are observed when the speed of web is high enough that air fingers grow from the meniscus, breaking the bead.

1.2.4 Tensioned-web-over-slot die coating

In tensioned-web-over-slot die (TWOSD) coating, the web is coated with a pre-metered layer of liquid by passing over a slot die from which liquid issues at a controlled rate per unit width of web.

The origin of the method is not clear, but the method was developed for applying thin, uniform layers of shear-thinning magnetic suspensions to a thin flexible web at comparatively high speed circa 1979. In that year, a patent issued by Pipkin and Schaefer (1979) described a slot coating system having the die pushed against a tensioned web without backup roll to produce magnetic storage media, tapes and floppy disks, as sketched in Fig. 1.8.

The absence of the back up roll is the main difference between TWOSD coating and the conventional fixed-gap one. The basic mechanism of this system exploits the balancing between the normal component of the resultant of the longitudinal tension in the curved web passing over the liquid issuing from the die slot and the pressure force of the liquid arose from the lubrication flow between the die lips and the moving web. Therefore, the elastohydrodynamic interaction controls the gap height, the distance between the moving web and the die lips.

Due to the interaction, the method can maintain an extremely thin gap height down, to the order of a micron, without the risk of clashing rigid surfaces. Given that the minimum wet thickness in slot coating method is proportional to the gap height, the method is suitable to produce thinner then 10µm compared with the conventional slot coating method when the web speed is 100m/min or more (Tanaka and Noda, 1984). This coating method proved to be better alternative to roll coating and gravure coating that were common in manufacturing of flexible magnetic media at that time.
1.2. SURVEY OF TENSIONED-WEB COATING METHODS

Figure 1.8: Tensioned-web-over-slot-die coating. Note that an oscillator motor is connected to the oscillating arm and is arranged to provide an oscillating movement of the arm to move the head back and forth across the tape. (From Pipkin and Schaefer 1979)
1.2. SURVEY OF TENSIONED-WEB COATING METHODS

As shown in Fig. 1.8, two idler rolls guide the moving web, which is under tension in such a way to control the approach and departure angles of the moving web with respect to the die. One of the benefits of the tensioned-web coating system is the inherent flexibility. By varying parameters such as web tension approach and departure angles of the web, it is possible to control the pressure profile without the need of a vacuum chamber that is an essential component of conventional slot coating when coating thinner than $1/3$ of the gap height.

Not only the operating conditions, but also the die lip design is critical to control the elastohydrodynamic interaction of the coating flow. Subsequently, a variety of patents on the method were published. Early patents use straight die lip design like the fixed-gap slot coating method, as shown in Fig. 1.9(a). Most of the available scientific research in tensioned-web coating use lip design based on this patent (Feng, 1998; Lin et al., 2008; Park, 2008). As Park (2008) pointed out, the die lip salient point, B in Fig 1.9(a), is the important design parameter to control the pressure peak inside the coating bead, that can affect the range of operating conditions. However, too sharp die lip may scratch and tear the web. In order to prevent these defects, most of the subsequent die lip designs were based on a lip geometry with a prescribed radius of curvatures, as shown in 1.9(b). Furthermore, the lip radius can be directly related to the curvature of the web that adjust the normal stress resultant from the web, especially for small flow rates. Since the pressure profile is a function of the lip curvature, the patents claimed the proper range of the lip radius based on the operating conditions.

Another important issue discussed in the patent literature is the undesired presence of recirculations under the die lip and inside the feed slot (Takahashi and Shibata, 1995). The recirculations, or micro-vortices, are dangerous in coating flows. They tend to centrifuge denser particles, to desorb dissolved gas, to collect and discharge bubbles, to hold formulations long enough for unwanted flocculation or polymerization, and to become nodular along their length and thereby detract from cross-wise coating uniformity. Particle trapped near the feed slot exit may lead to formation of streak lines along the flow direction. The patent claimed that the vortex near the feed slot decreases its size or vanished as the apex point of the downstream die lip is shifted from the corner of the feed slot exit to the middle of the downstream die lip.
1.2. **SURVEY OF TENSIONED-WEB COATING METHODS**

(a) Straight die lip (Tanaka and Noda, 1984)

(b) Rounded die lip (Shibata and Takahashi, 1994)

(c) Offset downstream die lip by shifting the lip apex point (Takahashi and Shibata, 1995)

(d) Downstream die lip with different lip curvatures (Okuno and Kawabe, 1994)

**Figure 1.9:** Die shapes of tensioned-web slot die for single-layer coating shown in patents.
1.2. SURVEY OF TENSIONED-WEB COATING METHODS

Another type of die lip designs claimed in different patents (Okuno and Kawabe, 1994; Shibata and Chino, 1993) was developed to better control the pressure profile. It uses multiple die lip curvatures for the downstream die lip (Fig. 1.9(d)). Okuno and Kawabe (1994) claimed that this composite die design can achieve the desirable gap height profile along the die lip for successful coating.

Several patents claimed different aspects of die-lip design, especially its shape, but also surface hardness of the lips, special treatment of the corners and outer edges of the lips, ranges of viscosity of coating liquid and so on. But most of them are based on experimental experience and ad hoc reasoning not fundamental or scientific evidence, well controlled scientific experiments and basic physical principles.

1.2.5 Two-layer tensioned-web-over slot die coating

There are several techniques for applying multiple layer of liquids using TWOSD coating method. The most efficient way to manufacture multi-layered products is to apply all the layers at once before they are solidified. The scientific literature about the dual slot TWOSD coating method is scarce, but there are several patents about it. Chino et al. (1989) emphasized the importance of downstream and mid die lip radii. Based on a specific die lip design (Fig. 1.10(a)), they disclosed the guideline for the choice of lip radii based on the flow rate, in order to prevent streak line along the flow direction. They also warned that too large downstream die lip radius \( R_2 \) can cause the meniscus to invade into the downstream die lip and too small \( R_2 \) can cause die shoulder wetting, that may cause non-uniform meniscus in the cross-web direction.

Takahashi and Shibata (1995), like in single-layer TWOSD coating, used the apex point to prevent or suppress micro vortex inside feed slot.

Tomaru (1995) emphasized that the process can be optimized by controlling drag forces \( P_1, P_2 \) and \( P_3 \) that comes from liquid, and normal stress resultant \( R_1 \) and \( R_3 \) (Fig. 1.10(c)). The forces are adjusted by the position of idlers and location of die lip surfaces. Based on a membrane approximation of the tensioned web (Flügge 1973), he computed drag forces as a function of operating conditions.
1.2. **SURVEY OF TENSIONED-WEB COATING METHODS**

(a) Two different die lip radii for downstream and mid die lips \(^{(Chino et al., 1989)}\)

(b) Mid lip apex point close to downstream die lip \(^{(Takahashi and Shibata, 1995)}\)

(c) Die lip configurations and idlers positions to control drag forces \(P_1\), \(P_2\), and \(P_3\) \(^{(Tomaru, 1995)}\)

(d) Overbite upstream die lip with short mid die lip length \(^{(Tomaru et al., 1997)}\)

(e) Using upstream die lip as smoothing out the pre-wetted liquid film \(^{(Kistler et al., 2000)}\)

**Figure 1.10:** Die shapes of tensioned-web slot die for dual slot coating shown in patents.
In another patent, Tomaru et al. (1997) disclosed the method of using a pre-wetted web with upstream die lip overbite configuration, i.e. pushing the upstream die lip further toward the web than the other lips in order to scrape the excessive pre-wetted layer (Fig. 1.10(d)). According to them, the “liquid-sealing” of the upstream die lip using pre-wetted layer can prevent defects that come from the upstream meniscus. They also proposed a short-length mid lip \( L_3 \) in order to prevent large movement of the interlayer separation point that causes waviness of the interlayer.

When the pre-wetted layer has a functional proposed, three-layer coating is also possible as sketched in Fig 1.10(e). They claimed that the pre-wetted layer coated by gravure coating, that has patterns from the grooved roll, can be smoothed by the upstream die lip of the coater. For successful smoothing out the surface of the pre-wetted layer, they proposed a correlation of the desirable upstream die lip radius \( R_u \) to operating conditions:

\[
R_u = \frac{2h_{t,w}}{K(6\eta U_w/T)^{2/3}}
\]

(1.1)

where \( h_{t,w} \) is the total wet thickness and \( K \) is a constant from 0.1 to 0.5. \( U_w, \eta, \) and \( T \) are web speed, viscosity measured at shear rate \( 10,000 \text{sec}^{-1} \), and web tension, respectively. Note that the equation comes from the similarity between the upstream coating bead and air foil bearing system: it is based on the equation for the lubricating thickness of a liquid bearing between a moving flexible foil and a curved surface (Blok and van Rossum, 1961). Also their die lip design (Fig. 1.10(e)) had the apex point located in the middle of the mid lip surface, which is rarely observed in other patents, even though the reasoning for this choice of apex point location is not clear.

1.3 Scientific researches on flows in tensioned-web coating

Tensioned-web over roll coating, also called kiss coating, is based on the mechanism of foil bearing (Pranckh, 1989). The basic characteristics of both systems can be captured by the lubrication flow inside a channel that has, at least one side, a flexible element.
Early researches on tensioned-web coating flow were similar to the analysis used in air foil bearing (Eshel and Elrod 1965). Pearson (1985) investigated the stability of the liquid flow in tensioned-web roll coating in forward mode. The flow model assumed that the substrate as perfectly flexible, i.e. membrane approximation (Flügge 1973). The flow inside the channel are described by the Reynolds equation. He found an indication of the effect of disturbances of the liquid flow, but the results have to be considered with caution because of ad hoc pressure boundary condition at the exit (Pranckh 1989).

In the Coating Process Fundamentals Program at the University of Minnesota, different coating flows have been analyzed from a standpoint of fundamental fluid mechanics and interfacial phenomena. Among the different methods, the elastohydrodynamic interaction in tensioned-web coating flows has been also studied. Shibata and Scriven (1986) analyzed single-layer TWOSD coating by solving the Navier-Stokes system by means of Galerkin’s finite element method (G/FEM). The web was described by membrane theory and the tension variation along the length of the substrate was neglected. These approximations are valid only at a limited parameter range, because it cannot capture the accurate liquid/web interaction when the curvature of web changes rapidly, that occurs when complex die lip geometry is used. Tomaru and Scriven (1998) expanded Shibata’s theory to dual-slot TWOSD coating flow. After them, Feng (1998) published an analysis about the single-layer TWOSD coating flow using a similar method. Carvalho (2003) proposed a tensioned-web roll coating flow model by coupling two-dimensional Navier–Stokes theory and thin inextensible shell theory with linear elastic models (Flügge 1973). The first thesis in Minnesota about TWOSD was written by Lee (2001). He performed experiments in a bench-scale tensioned-web slot coating system with the help of Engineer Wieslaw Suszynski.

Recently, Park (2008) has succeed in explaining the essential flow characteristics in TWOSD coating in terms of different configuration of foil bearing. He recognize that TWOSD coating flow can be expressed as the combination of multiple foil bearings in series. Furthermore, he developed an elasto-visco-capillary model to describe single-layer TWOSD coating flow. According to his results, the die geometry has a strong effect on the range of desirable operating parameters (coating window). However, this $1 - D$ model is not able to describe the presence of micro recirculation in the coating.
bead, that may lead to several coating effects.

He also performed computational studies on two-layer TWOSD coating using two
different models: a 1-D elasto-visco-capillary model and a 1-D/2-D hybrid model.
Both models assume that the moving web behaves like a thin membrane. For elasto-
visco-capillary model, he used lubrication approximation to describe the liquid flow
and the downstream meniscus is treated by the equation proposed by Landau and
Levich (1942). Using the model, he showed the change of location of the upstream
meniscus and the interlayer separation point as a function of operating conditions and
die lip curvatures. However, the flow near the interlayer separation point is highly
two-dimensional, because the flow separates at a point from the die lip. Therefore the
model may not be suitable for predicting the interlayer separation point location. In
the 1-D/2-D hybrid model, he used two-dimensional Navier-Stokes equation for the
flow near the mid lip and both upstream and downstream of the mid lip were described
by the same equations used in the elasto-visco-capillary model. Using the model, he
focused on the effect of operating conditions on the size of the downstream feed slot
vortex. But he did not emphasize on the interlayer separation point movement that
may lead to coating defects and coating windows for the two-layer coating method.

Lin et al. (2008) took a different approach from Scriven and co-workers. They used
lubrication approximation to describe the liquid flow and a simplified membrane
theory for the web. The web configuration is only computed at the exit of the feed
slot by balancing the normal force of the curved web to the liquid traction. Under
the die lips, the web location is approximated simply by using geometric arguments.
The downstream meniscus location is based on the Landau-Levich equation, which
is only valid at low capillary number. Even with all the approximations made on
the development of the model, the predictions show the same trends observed in the
experiments by Lin et al. (2007).
1.4 Defect-causing flow features in tensioned-web-over-slot die coating flow

As in other coating processes, the range of uniform coating thickness possible for TWOSD coating is limited. The region in the space of operating parameters of a coating process where the delivered liquid layer is adequately uniform is usually referred as the coating window of the process.

Historically, the fixed-gap single-layer slot coating is one of the most researched coating process. Ruschak (1976) proposed the first primitive coating window, or called the feasibility window, based on a simplification that viscous forces are neglected, except in the draw-down region. The window showed the range of vacuum pressure at a given wet thickness. Higgins and Scriven (1980) extended Ruschak’s analysis of the coating bead by adding drag by the web and die lips. The model is called the visco-capillary model for the slot coating. Sartor (1990) showed that the visco-capillary model gives very accurate predictions on the coating window, by comparing with experiment data. He also analyzed the coating flow by solving the two-dimensional Navier–Stokes equations by means of Galerkin finite element method. From the computational model and flow visualization, he also found that the relationship between the feed slot height and the critical wet thickness for the vortex birth. The effect of die lip design on the coating window was extensively studied by Gates (1999).

In the fixed gap slot coating, the coating window is bounded by different modes of failures, as discussed by Romero et al. (2004). Figure 1.11 shows a typical coating window for the slot coating process bounded by three mode of failure:

1. Too great vacuum at the upstream meniscus causes liquid to be drawn along the die surface into the vacuum chamber: Weeping.

2. Too little vacuum at the upstream meniscus cannot provide enough pressure gradient to match the viscous drag force, so that the meniscus shift toward the feed slot that lead to break up of coating beads: Bead breakup.

3. At given web speed, too low a flow rate per unit width from the slot causes the
1.4. **DEFECT-CAUSING FLOW FEATURES IN TWOSD COATING FLOW**

![Coating Window Diagram](image)

**Figure 1.11:** Coating window of a fixed-gap slot coating process in the plane of vacuum $P_{vac}$ vs. gap-to-thickness ratio $H_0/t$, at a fixed capillary number $Ca = \mu U_w/\sigma$, where $H_0$, $t$, $\mu$, $U_w$, and $\sigma$ are gap height, wet thickness, viscosity, web speed and surface tension, respectively. The boundaries of the window are set by different bead breakup mechanisms. $t_{min}$ represents the minimum film thickness that can be deposited onto the substrate at a given capillary number. (from Romero et al. 2004)

In tensioned-web coating flow, the normal component of the web tension is mostly balanced by the pressure force inside the bead. Even though the modes of failures or the defect-causing flow features are very similar to the conventional fixed-gap slot coating, the critical operating conditions for the onset of flow features are radically different from the conventional one.

Figure 1.12 shows the defect-causing flow features in TWOSD coating flow. When wrap angles of the moving web are not properly adjusted, especially too large downstream wrap angle, the downstream static contact line will wet the die shoulder leading to a non-uniform wavy downstream meniscus. The opposite case, too small
1.4. DEFECT-CAUSING FLOW FEATURES IN TWOSD COATING FLOW

downstream wrap angle, will create a diverging channel between the die lip and the moving web near the meniscus that may cause a large adverse pressure gradient. This pressure gradient can cause a ribbing instability that destroys cross-web coating uniformity.

The minimum possible wet thickness can be related to the invasion of the upstream meniscus towards the feed slot, that occurs if the pressure difference under the upstream die lip is not strong enough to balance the viscous drag force. The coating bead drastically rearranges into a three-dimensional form that delivers separate rivulets to the substrate. The event will lead to breakup of the bead that causes dry lanes: it is called bead breakup.

When the flow rate is too high, the high pressure difference pushes the meniscus beyond the upstream die lip corner, leading to a non-uniform meniscus that causes a variation across the coating width. It also results in loss of liquid from the bead,
which destroys the pre-metered action: this defect is called *weeping* or *dripping*. The similar flow features — leaking instability and nodulose meniscus — in the fixed-gap slot coating flow are discussed by Gates (1999) in detail.

Not only the flow instabilities that occur near the meniscus, but also internal flow features, can degrade the coating quality. The microscopic vortex, gyre, or recirculation inside the feed slot is one of them. It typically extends across the entire coating bead, as documented by Sartor (1990). It tends to centrifuge denser particles, to desorb dissolved gas, to collect and discharge bubbles, to hold formulations long enough for unwanted flocculation or polymerization.

Up to the author’s knowledge, scientific literature on coating windows for the TWOSD coating method is scarce, especially compared with the conventional fixed-gap slot coating. Most of them focused on the change of flow states, including web profiles and pressure profiles, as a function of the process operating conditions or die lip design change. Some, for example Lin et al. (2008), focused on bead breakup phenomena because the minimum wet thickness is one of the important operating limit of the process. Only Lee (2001) and Park (2008) reported weeping and bead breakup limit curves in the coating window in the plane of wet thickness and web speed for single-layer TWOSD coating. However, the number of data points on the limit curves are usually limited due to time for computation or experiment. In this work, coating windows for both single-layer and two-layer TWOSD coatings are presented for various die lip design. Furthermore, the windows are constructed with an computationally efficient manner so that the data points along each limit curves are enough to represent precise boundaries of the windows.

### 1.5 Final remarks

This thesis, like Park (2008)’s before it, treats tensioned-web-over-slot die (TWOSD) coating, both single layer and two layer. The purpose of the research is to understand the complicated flow characteristic inside the coating bead by solving the two-dimensional Navier-Stokes theory using finite element method and visualizing the coating bead flow on a lab-scale TWOSD coater. Through the analysis of TWOSD
coating flow, the ultimate goal is to construct the coating windows for the TWOSD coating process and determine the effect of die lip and operating conditions.

In order to analyze this particular the coating flow, several tools are developed and tested on other coating flows relatively simpler than TWOSD flow. As pointed out previously, microvortices inside flow can degrade product quality or cause coating defects. For the creation of accurate coating window theoretically, the onset of vortex birth must be detected systematically, without cumbersome checking the existence of vortex by post-processing of the solution of Navier–Stokes equations. Here, we propose the vortex birth condition in two-dimensional steady flow based on application of standard bifurcation theory on a pathline of fluid particle. The critical vortex birth trajectories inside the parameter space are tracked automatically using a direct tracking method, proposed in this study.

Another important flow feature, that is especially critical to two-layer coating, is related to position of an interlayer separation point: mid-gap invasion. A theoretical model for tracking interlayer separation point is developed and tested in a simpler system than the two-layer TWOSD coating: two-layer fixed-gap slot coating flow. Die lip corners are treated as a rounded corner with prescribed radius in order to allow the separation point to move along the corners. A new mesh generation scheme is proposed for maintaining smooth but highly concentrated meshes near the high curvature regions of the boundary. The results from flow visualization in a fixed-gap slot coating supported the predictions from the theoretical model.

The uniformity of the interlayer in both fixed-gap and tensioned-web dual slot coating plays a crucial role on the final product quality. Since unstable interlayer is one of the indications to wavy interlayer, we performed linear stability analysis to predict the neutral-stability curves that define the region of stable flow as a function of flow rate ratio and viscosity ratio. A new eigenvalue solver based on Valerio et al. (2007) are proposed and it allows to perform the analysis over a wide range of the operating conditions within reasonable time.

For the fundamental understanding of the flow in TWOSD coating, an accurate estimation of elastohydrodynamic interaction is crucial. Previous computational model for the TWOSD coating flow described the moving flexible web as a thin membrane
1.5. FINAL REMARKS

that neglects the tension variation along the length of the web. However this membrane approximations cannot capture the accurate liquid/web interaction when the curvature of web changes rapidly, that occurs when complex die lip geometry is used.

Using the tools described above, single-layer TWOSD coating flows are analyzed. The flow features that leads to coating defects, such as, bead breakup, weeping, and feed slot vortex, are identified and mathematical forms of the onset condition for the features are presented. The onset conditions are combined into direct tracking method that was used to construct the vortex-free operating window for the given die lip configurations. Furthermore, the tracking method can be used to shows the effect of die lip design on the critical parameters for the onset of the flow features.

Similarly, two-layer TWOSD coating flows are analyzed as well. The interlayer was approximated as a sharp interface as in fixed-gap slot coating. As in the single-layer coating, direct tracking method combined with the onset of flow features that lead to coating defects or degrade qualities is used to construct the vortex-free operating window. Effect of operating conditions and die lip design are examined in detail.

With complete analyses of TWOSD coating flow, desirable die lip design and ranges of operating conditions for both single- and two-layer TWOSD coating methods are presented. The methods proposed in this thesis are not confined to TWOSD coating flow, but can be used in the analysis other flow-related processes or systems. Vortex birth analysis and direct tracking method can be used to find vortex-free operating conditions or design parameters. Also the interlayer model combined with rounded corners can be used for multi-phase flow systems where interlayer separation point is critical for operability limits.
Chapter 2

Identifying the birth of vortices in flows

2.1 Introduction

A vortex in a flow is typically described as eddy, recirculation, and swirl. From gigantic hurricanes on earth to micro-size gyres in coating flow, they share the same characteristic: the rotating motion of material particles around a common center [Lugt 1983]. Most vortices show a three dimensional spiral structure. Pathlines are not perpendicular to the axis of rotation but have a component parallel to it. However, in two-dimensional flows the pathlines of vortices are perpendicular to the axis of rotation and the structure is two dimensional.

A vortex inside a flow can be visualized by pathlines, streamlines or streaklines. Experimentally, this is done by using flow markers, such as dust, ink, and smoke. To identify a minuscule vortex, one may need a special tracer, such as hydrogen bubble or dye. In theory, numerically or analytically computed streamlines or pathlines illustrate the presence of a vortex by generating multiple-closed loops around a common center. However, streamlines and pathlines are not invariant under a change of frame of reference. For example, a moving sphere in a stationary fluid has closed streamlines varying with time in a laboratory reference; however when the observer moves
2.1. INTRODUCTION

with the sphere, the flow becomes time-independent and the closed loop streamlines disappear (Lugt 1983). This example shows that closed streamlines do not guarantee the existence of a vortex: without supporting flow theory and a proper frame of reference, a vortex can be invisible.

The study of the existence of a vortex in a flow is usually based on flow topology analysis (Bakker 1991). Early work on vortex formation was focused on the separation and attachment of a streamline from a solid object in laminar flow (Davey 1961; Lighthill 1964). The streamline that connects the separation and attachment points encloses a flow recirculation.

At the vortex center, the stream function reaches a maximum or minimum and the velocity is zero. This critical point or singular point concept was used to analyze the three dimensional flow near solid objects and to find stagnation and separation points (Tobak and Peake 1982; Perry and Chong 1987).

The transition from parallel streamlines or pathlines to complex ones, such as closed-loop pattern, carries the analysis to flow topology naturally. Bifurcation theory brings the concept of a degenerate point, which represents the collapse of two or more stagnation points into a single point, to establish the local flow kinematics at the conditions of “metamorphosis” of the flow pattern. Typically flow bifurcation theory assumes steady-state and incompressible flows, consequently streamlines, pathlines, and streaklines are identical (Prandtl and Tietjen 1934).

The analysis of degenerate stagnation points near a solid wall in a two-dimensional incompressible viscous flow were studied in detail by Bakker (1991). Characteristic flow patterns were examined by independently varying the Taylor series coefficients of the velocity field expansion. Lugt (1987) analyzed the presence of separation points in viscous free surface flows also using Taylor series expansion of the velocity field. The conditions at which degenerate stagnation points appear at liquid-gas interface were analyzed by Brøns (1994). The presence of simple degenerate stagnation points inside a flow, away from boundaries, were considered by Brøns and Hartnack (1999). Hartnack (1999) and Gürcan et al. (2005) revisited Bakker (1991) work and explored simple and non-simple degeneracies of stagnation points. Multiple vortex / saddle patterns and their evolution inside a shear dominated flow were considered by Gaskell...
2.1. **INTRODUCTION**

Most of these analysis is focused on the new flow structures that occur when the flow field is varied through a degenerate configuration. The kinematics near a critical point is described by Taylor series expansion of the stream function. The coefficients of the expansion are considered as bifurcation parameters. The resulting streamline pattern can only be determined by considering the higher-order terms of the Taylor series. However, the signal of degeneracy of a stagnation point and consequently of vortex birth can be detected by considering only the linear term.

Here we describe the flow kinematics around stagnation points by using Taylor series expansion of the velocity field to find the conditions of vortex birth for any type of two-dimensional incompressible flows with a comprehensive view. The local conditions for vortex birth can be defined by only considering the linear term in the Taylor series. Also using eigenanalysis of the linear term, the vortex birth process can be depicted with a more complete and clear description. The birth signal can be expressed in terms of kinematic variables: extensional rate, shear rate, vorticity and their derivatives. In the case of Newtonian fluids, the conditions may be defined in terms of dynamic variables: shear stress, pressure and their derivatives.

The results from the analysis of vortex birth, a local flow feature condition, can be used to find the flow parameters at which a vortex birth inside the flow occurs. The connection from the global flow conditions to the local kinematic conditions for vortex birth can be used to track the operating parameters at which recirculations appear or disappear in a flow. By using multi-parameter continuation, a line (or surface) in the parameter space that define the flow parameters at which a vortex emerges or vanishes may be constructed automatically, and a vortex-free region of the parameter space may be determined. Such regions of the parameter space are extremely important to define optimal operating parameters for process that may be ruined by the presence of vortex, such as continuous liquid coating of substrates.
2.2 Analysis of flow near a stagnation point to find the vortex birth

The description of a fluid particle motion inside a flow can be decomposed into three independent parts — translation, deformation and rotation. With the proper choice of the frame of reference, the translation can be eliminated from the description. In this frame of reference, a flow recirculation is characterized by closed streamlines, and the axis of rotation of the vortex, the vortex center, is at rest relative to the observer. Hence the birth of a vortex is depicted as the splitting of stagnation point(s) to spawn a new vortex center. However, not all stagnation points in a flow are vortex centers. Saddle points and cusp points of streamlines are also stagnation points. In order to identify the vortex birth, characteristics of stagnation points must be analyzed and classified.

When a steady-state flow system is considered, the velocity field around a stagnation point, $x_0$, can be expressed using Taylor series expansion,

$$
\mathbf{u}(x) = \mathbf{u}(x_0) + (x-x_0) \cdot \nabla \mathbf{u}(x_0) + \frac{1}{2} (x-x_0)(x-x_0) : \nabla \nabla \mathbf{u}(x_0) + \cdots.
$$

(2.1)

Because $x_0$ is the stagnation point, $\mathbf{u}(x_0)$ is zero. Therefore the characteristic of the flow field near the stagnation point, in general, depends on the linear term, $\nabla \mathbf{u}(x_0)$. However there are two possible scenarios at which the linear term does not govern the flow field near a stagnation point: no net contribution, $\text{det} \nabla \mathbf{u}(x_0) = 0$, or no contribution, $\nabla \mathbf{u}(x_0) = 0$.

When the linear term contributes to the flow field, and $x$ is close enough to stagnation point, $x_0$; higher order term in Eq. (2.1) can be neglected, and the equation that describes pathlines near a stagnation point $x_0$ is

$$
\frac{D \mathbf{x}}{D t} = \frac{D \mathbf{x}}{D(\vartheta t^*)} = \mathbf{u}(x) = (x-x_0) \cdot \nabla \mathbf{u},
$$

(2.2)

where $t$ is time, $\vartheta$ is a proper unit of time and $t^*$ is a dimensionless time. Note that a unit of time is immaterial, i.e. a choice of unit does not affect the solution of
2.2. ANALYSIS OF FLOW NEAR A STAGNATION POINT FOR VORTEX BIRTH

Eq. (2.2). The pathline is found by integrating Eq. (2.2),

$$\mathbf{x}(t) = \sum_{i=1}^{N} \exp(\lambda_i(x_0) t) \mathbf{z}_i(x_0) \mathbf{w}_i^\dagger(x_0) \cdot (\mathbf{x}(t = 0) - x_0) + x_0,$$

(2.3)

where \(N\) is the dimension of the flow (\(N = 2\) for two-dimensional flow), \(\lambda_i(x_0)\) is the eigenvalue of the velocity gradient, \(\mathbf{z}_i(x_0)\) is the corresponding unit eigenvector, and \(\mathbf{w}_i^\dagger(x_0)\) is the complex conjugate of the corresponding reciprocal eigenvector. As shown in Eq. (2.3) pathlines near the stagnation point depend on the eigenvalues and eigenvectors of the velocity gradient.

To simplify the notation from now on, if not specified otherwise, all variables are evaluated at the stagnation point, \(x_0\), e.g. \(u = u(x_0)\).

In this study, we consider steady-state flows only; the velocity field and its spacial derivatives depend only on position. Therefore, Eq. (2.2) is what is called an autonomous system in bifurcation theory (Hale and Kočak 1991): it is a system of ordinary differential equation which does not depend on the independent variables – in this case, \(t\). The analysis presented here is also restricted to two-dimensional incompressible flows.

### 2.2.1 Stagnation point inside a flow – “far-away” from a flow boundary

Using the irreducible representation of a dyadic, the velocity gradient can be decomposed into a symmetric part, \(S\) and an antisymmetric part, \(A\):

$$\nabla u = \frac{1}{2} \left[ \nabla u + (\nabla u)^T \right] + \frac{1}{2} \left[ \nabla u - (\nabla u)^T \right].$$

(2.4)

The symmetric part of the velocity gradient is called the rate of strain or the state of strain rate that characterizes the relative motion between a given particle and every other particles in its neighborhood: it represents deformation. In two-dimensional
flow and Cartesian coordinate, the symmetric part is

\[
S = i \frac{\partial u}{\partial x} + ij \left[ \frac{1}{2} \left( \frac{\partial v}{\partial x} + \frac{\partial u}{\partial y} \right) \right] + ji \left[ \frac{1}{2} \left( \frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right) \right] + j j \frac{\partial v}{\partial y} + ji \left( \frac{\partial v}{\partial x} \right) + ji \left( \frac{\partial u}{\partial y} \right) + j j \left( \frac{\partial v}{\partial x} \right) (2.5)
\]

\[
S = i (\dot{\varepsilon}_{xx}) + ij \frac{1}{2} (\dot{\gamma}_{xy} + \dot{\gamma}_{yx}) + ji \frac{1}{2} (\dot{\gamma}_{yx} + \dot{\gamma}_{xy}) + j j \left( \dot{\gamma}_{yy} \right) (2.6)
\]

\[
S = i \dot{\varepsilon} + i j \dot{\gamma} + j i \dot{\gamma} + j j (-\dot{\varepsilon}) (2.7)
\]

where \( \dot{\varepsilon}_{xx} = \frac{\partial u}{\partial x} \) and \( \dot{\varepsilon}_{yy} = \frac{\partial v}{\partial y} \) are extensional rates, and \( \dot{\gamma}_{xy} = \frac{\partial u}{\partial x} \) and \( \dot{\gamma}_{yx} = \frac{\partial v}{\partial y} \) are shear rates. Note that from the continuity equation, \( \frac{\partial v}{\partial y} = -\dot{\varepsilon}_{yy} = -\frac{\partial u}{\partial x} = -\dot{\varepsilon}_{xx} \). For a simpler representation, the average shear rate, \( \dot{\gamma} = \left( \dot{\gamma}_{xy} + \dot{\gamma}_{yx} \right) / 2 \), and the extensional rate, \( \dot{\varepsilon} = \dot{\varepsilon}_{xx} \), are used from now on.

The determinant of the symmetric part of the velocity gradient is

\[
\text{det} S = -(\dot{\varepsilon}^2 + \dot{\gamma}^2). (2.8)
\]

It is always negative, and, consequently the eigenvalues are two distinct real numbers with opposite sign:

\[
\lambda_{\pm}^S = \pm \sqrt{\dot{\varepsilon}^2 + \dot{\gamma}^2} = \pm \sqrt{\text{det} S}, (2.9)
\]

and the corresponding eigenvectors are

\[
z_{\pm}^S = i (\dot{\varepsilon} \pm \sqrt{\dot{\gamma}^2 + \dot{\varepsilon}^2}) + j \dot{\gamma}. (2.10)
\]

The eigenvectors used in this study are the left eigenvectors, i.e. \( z^S \cdot S = \lambda^S z^S \), and they are consistent with the Taylor series expansion used in Eq. (2.3). To simplify the representation, the eigenvectors are not normalized. They capture a qualitative flow feature at the stagnation point, because they represent the characteristic directions at that point. If the velocity gradient has only the symmetric part, there are two distinct eigenvectors in the plane of the flow. Two different pathline cross at the stagnation point. Along one of the pathlines, liquid flows towards the stagnation point, and flows away from it along the other pathline. The flow field near the stagnation point consists of both expansion (or contraction) and shear deformation without a circular motion. This stagnation point is known as a saddle point.
2.2. ANALYSIS OF FLOW NEAR A STAGNATION POINT FOR VORTEX BIRTH

The antisymmetric part of the velocity gradient describes a local solid-body-like rotation around a point:

\[ \mathbf{A} = \mathbf{i} \left[ \frac{1}{2} \left( \frac{\partial v}{\partial x} - \frac{\partial u}{\partial y} \right) \right] + \mathbf{j} \left[ -\frac{1}{2} \left( \frac{\partial v}{\partial x} - \frac{\partial u}{\partial y} \right) \right], \]

\[ = \mathbf{i} \mathbf{j} (\omega) + \mathbf{j} (-\omega), \quad (2.11) \]

where \( \omega = \left( \frac{\partial v}{\partial x} - \frac{\partial u}{\partial y} \right) \) is the vorticity of the flow. The antisymmetric component of the velocity gradient is sometimes called the vorticity dyadic. Its determinant is

\[ \det \mathbf{A} = \left( \frac{\omega}{2} \right)^2. \quad (2.12) \]

It is always positive. There are two complex conjugate eigenvalues:

\[ \lambda_{\pm}^A = \pm i \frac{\omega}{2} = \pm i \sqrt{\det \mathbf{A}}, \quad (2.13) \]

and the corresponding eigenvectors are

\[ \mathbf{z}_{\pm}^A = \mathbf{i} (\pm i) + \mathbf{j}. \quad (2.14) \]

Note that not only the eigenvalues but also the eigenvectors are complex. Because the eigenvalues are pure imaginary numbers, pathlines around the stagnation point are concentric circles, and the flow field shows swirling or eddying motion. Therefore closed loop pathlines near a stagnation point are closely related to the antisymmetric part of the velocity gradient.

In order to identify the characteristic of the stagnation point, both parts – symmetric and antisymmetric – should be evaluated together. In terms of the kinematic variables, the velocity gradient is expressed as:

\[ \nabla \mathbf{u} = \mathbf{i} \dot{\varepsilon} + \mathbf{j} \left( \dot{\gamma} + \frac{\omega}{2} \right) + \mathbf{j} \left( \dot{\gamma} - \frac{\omega}{2} \right) + \mathbf{j} (-\dot{\varepsilon}). \quad (2.15) \]

The determinant, eigenvalues and eigenvectors of the velocity gradient are a combination of the determinant, eigenvalues and eigenvectors of the symmetric and antisymmetric components.

\[ \det \nabla \mathbf{u} = -\left( \dot{\varepsilon}^2 + \dot{\gamma}^2 \right) + \left( \frac{\omega}{2} \right)^2 = \det \mathbf{S} + \det \mathbf{A}. \quad (2.16) \]
2.2. ANALYSIS OF FLOW NEAR A STAGNATION POINT FOR VORTEX BIRTH

\[ \lambda_{\pm} = \pm \sqrt{(\dot{\varepsilon}^2 + \dot{\gamma}^2) - \left(\frac{\omega}{2}\right)^2} = \pm \sqrt{-\det \nabla \mathbf{u}}, \quad (2.17) \]

\[ \mathbf{z}_{\pm} = \mathbf{i} \left( \dot{\varepsilon} \pm \sqrt{-\det \nabla \mathbf{u}} \right) \pm \mathbf{j} \left( \dot{\gamma} - \frac{\omega}{2} \right). \quad (2.18) \]

Note that eigenvalues and eigenvectors have imaginary parts when \( \det \nabla \mathbf{u} \) is positive. Eq. (2.16) implies that the sign of the determinant depends on the competition between the symmetric and the antisymmetric part of the velocity gradient.

The competition can be categorized in four possible scenarios:

1. **Symmetric part dominant** \(| \det \mathbf{S} | > | \det \mathbf{A} | \)
   
   In this scenario, the determinant of the velocity gradient is negative and the system has two distinct real eigenvalues. The deformation predominates over the solid body rotation. The condition can be expressed as an inequality between the magnitude of the extensional and shear rates, and the vorticity:
   
   \[ \dot{\varepsilon}^2 + \dot{\gamma}^2 > \left( \frac{\omega}{2} \right)^2, \quad (2.19) \]
   
   In this case, the stagnation point is a saddle point.

2. **Anti-symmetric part dominant** \(| \det \mathbf{S} | < | \det \mathbf{A} | \)
   
   The determinant of the velocity gradient is positive and the system has two pure imaginary conjugate eigenvalues. In contrast to the first scenario, the deformation is overwhelmed by the rotational motion. In this scenario, the vorticity is larger than the magnitude of the deformation rate.
   
   \[ \dot{\varepsilon}^2 + \dot{\gamma}^2 < \left( \frac{\omega}{2} \right)^2. \quad (2.20) \]
   
   The stagnation point is a vortex center.

3. **Equal and opposite influence from symmetric and anti-symmetric parts** \(| \det \mathbf{S} | = | \det \mathbf{A} | \) with \( \mathbf{S} \neq \mathbf{0} \) and \( \mathbf{A} \neq \mathbf{0} \)
   
   In contrast to the previous two scenarios, the deformation is set off by the rotation. Consequently, the magnitude of the extensional and the shear rate, and the vorticity are exactly equal:
   
   \[ \dot{\varepsilon}^2 + \dot{\gamma}^2 = \left( \frac{\omega}{2} \right)^2. \quad (2.21) \]
2.2. ANALYSIS OF FLOW NEAR A STAGNATION POINT FOR VORTEX BIRTH

The determinant of the velocity gradient is zero and the system has a zero eigenvalue with algebraic multiplicity two and geometric multiplicity one:

\[ \lambda_0 = 0, \quad z_0 = i \dot{\epsilon} + j \left( \dot{\gamma} - \frac{\omega}{2} \right). \]  \hspace{1cm} (2.22)

The vanishing determinant implies that the velocity gradient – the linear term in Eq. (2.1) – cannot determine the pattern of pathlines near the stagnation point and the characteristics of the stagnation point. They are determined by the higher order terms.

4. Both parts have no contribution \((S = A = 0)\)

Because the velocity gradient is exactly zero, the determinant is meaningless and there is no deformation nor rotation:

\[ \dot{\epsilon} = 0, \quad \dot{\gamma} = 0, \quad \omega = 0. \]  \hspace{1cm} (2.23)

Note that these satisfy Eq. (2.21) automatically. As in the previous scenario 3, the pattern of pathlines near the stagnation point and the type of stagnation point depend on higher order term; however, there is no eigenvector and eigenvalue associated with the velocity gradient.

Stagnation points of the third and fourth scenarios are called degenerate stagnation points. According to Brøns (1994), the third scenario is called a simple degeneracy case and the fourth is called a non-simple degeneracy case. In both scenarios, degeneracy is caused by the vanishing determinant of the velocity gradient.

For the simple degeneracy case, scenario 3, the stagnation point has three possible forms: a degenerated saddle, a degenerated vortex center and a cusp point at a pathline as shown by Brøns and Hartnack (1999), see Fig. 2.1. When the eigenvalue of the velocity gradient crosses zero, a saddle point becomes a vortex center or vice versa. And in this path, a stagnation point must pass through the degenerate state. At this state, both eigenvectors, \(z_\pm\), merge into a single eigenvector, \(z_0\). Because the sign of a eigenvector does not affect its direction, merging of eigenvectors represents that they become parallel or anti-parallel to each other.

Figure 2.2 shows three ways of how the eigenvectors evolve at a stagnation point as it becomes a simple degenerate point. Note that the flow considered is steady, two-dimensional and incompressible and only parallel degenerate eigenvectors are shown.
2.2. ANALYSIS OF FLOW NEAR A STAGNATION POINT FOR VORTEX BIRTH

**EIGENVALUE** of velocity gradient (For incompressible flow)

\[ \lambda = \pm \sqrt{-\det \nabla u} \]

![Diagram of eigenvalues](image)

**Figure 2.1:** Pathlines near a stagnation point inside a steady two dimensional incompressible flow ([Brøns and Hartnack, 1999](#)) and corresponding eigenvalue of the velocity gradient. Note that the eigenvalues are only a function of the determinant of the velocity gradient.

Each case represents a different vortex birth from a degenerate stagnation point: a vortex born from a saddle point, from a vortex center, and from a cusp point.

Except when the birth occurs from a cusp point, the eigenvectors transition from real to imaginary as the determinant of the velocity gradient changes sign. When the determinant changes from negative to positive, two real eigenvectors merge into a single eigenvector and it re-splits into two complex conjugate eigenvectors. This transition is illustrated in the first column of Fig. 2.2. In Fig. 2.2(a), the stagnation point that gives birth to a new vortex center is a saddle point. The determinant of the velocity gradient is negative; one of the eigenvalues has a positive real part and the other has a negative real part. The eigenvectors, that represent characteristic directions of the pathlines at the stagnation point, are two real vectors, as shown in Eq. 2.18. They are tangent to the streamline that divides the flow in the region inside and outside the vortex, the infinity-shape curve in Fig. 2.2(a). This line is known as the *separating streamline* or *separatrix* that defines a boundary in the phase space in bifurcation theory ([Hale and Kocak, 1991](#)). As the flow evolves from (a) to (b) in Fig. 2.2, the angle between the eigenvectors falls until the two vectors \( z_1 \) and
2.2. ANALYSIS OF FLOW NEAR A STAGNATION POINT FOR VORTEX BIRTH

There are three possibilities for a vortex birth: born from a saddle point, from a vortex center, and from a cusp point. Numer of eigenvector is two for a stagnation point in general, but they are merged into one and split again after passing a degenerate state when the determinant becomes zero. Note that, as a rule, flow inside dotted box, in (j), does not have stagnation point. Therefore the linear term in the Taylor series expansion, Equation (2), and the determinant cannot “mold” the flow pattern: the determinant can be either positive or negative.

Figure 2.2: Evolution of eigenvectors at a stagnation point as the determinant of the velocity gradient changes. “Im”, “Stag. pt.”, “Sad. pt.”, “Vor. ct.”, “Deg. sd.”, “Deg. ct.”, and “Cusp pt.” stand for imaginary axis, stagnation point, saddle point, vortex center, degenerate saddle point, degenerate vortex center, and cusp point, respectively. There are three possibilities for a vortex birth: born from a saddle point, from a vortex center, and from a cusp point.
2.2. ANALYSIS OF FLOW NEAR A STAGNATION POINT FOR VORTEX BIRTH

When \( \det \nabla \mathbf{u} = 0 \), the separating streamlines become tangent at the stagnation point. This is what is called a degenerate saddle point. As the flow continues to evolve from (b) to (c) in Fig. 2.2, the determinant of the velocity gradient becomes positive. The eigenvalues are a pair of complex conjugate values and the eigenvectors, a pair of complex conjugate vectors. The real and imaginary parts of the eigenvectors are

\[
\mathbf{z}_\pm = \mathbf{Z}_0 \pm i \mathbf{Z}_{im}, \quad (\det \nabla \mathbf{u} > 0).
\]

The real part \( \mathbf{z}_{Re} \) is equal to the eigenvector \( \mathbf{z}_0 \) at \( \det \nabla \mathbf{u} = 0 \). The saddle-vortex center-saddle set that is formed from the degenerate saddle point splits in the direction aligned with \( \mathbf{z}_{Re} = \mathbf{z}_0 \), as indicated in Fig. 2.2 (c). As the anti-symmetric part of the velocity gradient becomes stronger, the magnitude of \( \mathbf{z}_{Im} \) grows. The flow motion approaches a solid body rotation and the area enclosed by the separating streamline rises. The separating streamline surrounding the new vortex, which replaces the degenerated saddle point, is formed from the detachment of tangent streamlines.

The sequence of flow states as a vortex is born from another vortex is presented in Fig. 2.2 (f) – (e) – (d). The transition is similar to the one discussed before, but in a reverse order. The two complex conjugate eigenvectors \( \mathbf{z}_+ \) and \( \mathbf{z}_- \) associated with the vortex center in Fig. 2.2 (f) collapse into a single real eigenvector \( \mathbf{z}_0 \) as the determinant of the velocity gradient approaches zero, as illustrated in Fig. 2.2 (e). The stagnation point in this figure is called a degenerate vortex center. The angular velocity near the stagnation point vanishes and consequently the period of a particle located at a finite distance from the center to complete one loop around the recirculation approaches infinity. As the determinant becomes negative, the eigenvector \( \mathbf{z}_0 \) splits into two real eigenvectors and the degenerate center splits into two vortex centers and a saddle point between them, as illustrated in Fig. 2.2 (d).

The birth of a vortex from a cusp is sketched in the third column of Fig. 2.2. The transition is different from the previous situations; the flow represented in Fig. 2.2 (i) that leads to the cusp point does not have a stagnation point. Consequently, the flow pattern is not defined by the linear term in the Taylor series expansion of the...
velocity field. A cusp point is formed where a streamline bends and forms a cusp, such that the velocity vector there vanishes; the cusp point is a stagnation point. This condition is illustrated in Fig. 2.2 (h); there is only one real eigenvector $z_0$. As the determinant of the velocity gradient becomes negative, the two eigenvectors split into two real eigenvectors $z_-$ and $z_+$ and a new vortex center is formed, as shown in Fig. 2.2 (g).

As presented in this section, a new vortex that appears inside a flow, far from the boundaries, is born from a degenerate stagnation point. As discussed in the literature, the configuration of the flow after the transition can only be determined by higher order terms of the Taylor expansion around the stagnation point. However, the local condition that signals the vortex birth is the vanishing determinant of the velocity gradient. The following sections describe the vortex birth at flow boundaries. The associated boundary condition has to be satisfied together with the velocity expansion near the stagnation point, Eq. (2.1), restricting the possible flow patterns.

### 2.2.2 Stagnation point at a liquid-gas interface

A liquid-gas interface, also called a liquid free surface, is an impermeable and deformable boundary between a liquid and a gas phase. Like any boundary, it has characteristic directions, the tangent and normal directions to the surface. These directions can be chosen as the natural basis set to describe the flow near the interface. Curvature effects of the free surface are neglected, here the birth of a new vortex from a stagnation point depends only on the kinematics in a region very close the point at which the tangent and normal vectors are considered constant. The canonical basis set $B = (i,j)^T$ can be transformed to the natural basis set along the interface, $B' = (n,t)^T$, by a properly chosen rotational dyadic, $R = B^T B' = ni + tj$. In terms of the natural basis set, the position vector is $x = s_n n + s_t t$, and the velocity vector is $u = u_n n + u_t t$. Here $s_t$ and $s_n$ are the arc-length coordinate along the tangential and normal directions to the interface, and $u_t$ and $u_n$ are the tangential and normal components of the velocity. The symmetric and antisymmetric components of the velocity gradient becomes $S = nn \dot{\epsilon}_s + nt \dot{\gamma}_s + tn \dot{\gamma}_s + tt(-\dot{\epsilon}_s)$ and $A = nt(\omega_s/2) + tn(-\omega_s/2)$, where $\dot{\epsilon}_s = \partial u_n / \partial s_n$, $\dot{\gamma}_s = 1/2(\partial u_n / \partial s_t + \partial u_t / \partial s_n)$ and $\omega_s = \partial u_n / \partial s_t - \partial u_t / \partial s_n$. 
Physics at the free surface dictates that there is no flow across the interface, i.e.
\( u_n = n \cdot u = 0 \) along the boundary. Only vanishing tangential velocity component,
\( u_t = 0 \), is required to define a stagnation point. However, the no-penetration condition restricts not only the normal velocity component itself but also its derivative along the boundary, i.e. the derivative of normal velocity component along free surface,
\( \frac{\partial u_n}{\partial s_t} \), vanishes as well.

Using the above simplifications, the velocity gradient at the stagnation point written in the natural basis set becomes

\[
\nabla u = n n \dot{\varepsilon}_s + t n (2 \dot{\gamma}_s) + t t (-\dot{\varepsilon}_s).
\]

The determinant of velocity gradient is

\[
\det \nabla u = -\dot{\varepsilon}_s^2,
\]

its eigenvalues are

\[
\lambda^f_{\pm} = \pm \dot{\varepsilon}_s,
\]

and corresponding eigenvectors are

\[
z^f_\pm = t \dot{\gamma}_s + n \dot{\varepsilon}_s, \quad z^f_+ = t.
\]

The eigenvectors of the velocity gradient, Eq. (2.28), represent the two characteristic directions of the pathlines, or streamlines, at the separation point: \( z^f_- \) is the direction of the streamline that ends at the stagnation point located on the free surface (separating streamline) and \( z^f_+ \) is tangent to the free surface. The flow direction along the streamline that meets the free surface at the stagnation point is a function of the local extensional rate \( \dot{\varepsilon}_s \); for \( \dot{\varepsilon}_s < 0 \), fluid flows towards the stagnation point along the free surface and away from if along the separating streamline; for \( \dot{\varepsilon}_s > 0 \), the flow is reversed. When the local extensional rate \( \dot{\varepsilon}_s \) vanishes, the determinant of the velocity gradient at the stagnation point is zero and the point becomes a degenerate stagnation point. There are two classes of degenerate stagnation point depending on the shear rate \( \dot{\gamma}_s \):

1. \( \dot{\gamma}_s \neq 0 \). Velocity gradient is non-zero but its determinant is zero. Only one eigenvector is associated to the stagnation point, i.e., \( z^f_- \) collapses into \( z^f_+ \) and aligns
along free surface. Therefore the velocity gradient has one eigenvalue equal to
zero with a single eigenvector. This situation is called a simple degenerate case.

2. \( \gamma_s = 0 \). Velocity gradient is zero and there are no eigenvalue nor eigenvector.
This is called non-simple degenerate case.

The possible flow bifurcation patterns at stagnation point(s) located along a free
surface near the state of degeneracy were studied by Brøns (1994). Among those flow
structures, vortex birth happens only when the stagnation points are connected by
the same separating streamline after splitting from a degenerate separation point as
shown in Fig. 2.3 (c). Locally-closed flow occurs inside the separating streamline,
because a fluid particle cannot cross it. Therefore, at least a vortex center must exist.
The onset of splitting of a degenerate stagnation point into two stagnation points is
the condition for vortex birth.

Figure 2.3 illustrates different sequence of flow states that leads to the birth of a
vortex attached to a free surface. The first column corresponds to vortex birth from
a simple degenerate stagnation point. The sequence starts with a flow state without
a stagnation point on the free surface, shown in Fig. 2.3 (a). Figure 2.3 (b) illustrates
the situation where one degenerate stagnation point is present. The determinant of
the velocity gradient and the extensional rate \( \dot{\epsilon}_s \) all vanish at the stagnation point. In
general, the point is a simple degenerate stagnation point. When the shear rate \( \dot{\gamma}_s \) also
vanishes, it is a non-simple degenerate stagnation point. This degenerate stagnation
point splits into two stagnation points, located on the liquid-gas interface, and one
vortex center (also a stagnation point) that is detached from the free surface, as shown
in Fig. 2.3 (c). The reverse process, an evolution from the flow state shown in Fig. 2.3
(c) to the one shown in Fig. 2.3 (b), represents the death of a vortex attached to the
free surface. In this case, the two stagnation points on the free surface collapses into
a single one. Therefore, the local kinematics at the vortex birth corresponds to the
limit when two stagnation points on the free surface approach each other; i.e.

\[
\frac{du_t}{ds_t}(s_{t0}) = \dot{\epsilon}_s(s_{t0}) = \lim_{\delta s_t \to 0} \frac{u_t(s_{t0} + \delta s_t) - u_t(s_{t0})}{\delta s_t} = 0,
\]

where \( s_{t0} \) and \( s_{t0} + \delta s_t \) are arc-length coordinate values of the two stagnation points
and \( \delta s_t \) is the distance between them. Therefore the degenerate separation point has
2.2. ANALYSIS OF FLOW NEAR A STAGNATION POINT FOR VORTEX BIRTH

both zero tangential velocity and zero extensional rate that, according to Eq. (2.26),
leads to a velocity gradient determinant equal to zero. This argument also holds for
multiple stagnation points when they are collapsed to single stagnation point.

The second column of Fig. 2.3 corresponds to a vortex birth from a non-simple de-
generate stagnation point located at the free surface. The angle between the free
surface and the separating streamline that terminates at the stagnation point, $\theta_f$,
is the angle between the eigenvectors of the velocity gradient.

\[
\theta_f = \begin{dcases}
\arctan \left( \frac{\dot{\epsilon}_s}{\dot{\gamma}_s} \right) & ; \dot{\gamma}_s \neq 0 \\
\frac{\pi}{2} & ; \dot{\gamma}_s = 0
\end{dcases}
\]  

(2.30)

This shows that, in a non-simple degenerate stagnation point, shown in Fig. 2.3
(e), the separating streamline is perpendicular to the free surface. The non-simple
stagnation point splits into four saddle points and two vortex centers, as shown in
Fig. 2.3 (f).

In order to write the conditions at the birth or death of vortices in terms of the
stress components, constitutive equations are required to relate them to the local
kinematics. In general, there is no simple relation between shear stress and the state
of strain-rate for constitutive equations that describe the behavior of complex fluids.
However, this relation is possible for generalized Newtonian fluid. The state of stress
of generalized Newtonian fluid is

\[
\tau = 2\eta(II_S)S.
\]  

(2.31)

where $\eta$ is viscosity which is function of the second invariant of the strain-rate tensor,
$II_S$. From the stress boundary condition at the free surface, the shear stress $\tau_{tn}$ must
vanish:

\[
\tau_{tn} = t_n : \tau = 2\eta(II_S)\dot{\gamma}_s = 0.
\]  

(2.32)

Therefore $\dot{\gamma}_s$ must vanish along the free surface because negative and zero viscosity is
physically impossible. The boundary condition also implies zero vorticity along the
free surface, i.e. $\partial u_i / \partial n = \omega_s = 0$. In other words, the stress boundary condition
casts into zero shear rate and zero vorticity for generalized Newtonian fluid. Note
that zero vorticity condition comes form the fact that we have neglected curvature
2.2. ANALYSIS OF FLOW NEAR A STAGNATION POINT FOR VORTEX BIRTH

![Diagram of Vortex Birth at Free Surface](image)

**Figure 2.3:** A vortex birth at free surface. First column, from (a) to (c), represents the birth from a simple degenerate stagnation point. Second column, from (d) to (f), represents the birth from a non-simple degenerate stagnation point. "Sep. st. line.,” “St. pt.”, “Vor. ct”, and “Deg. st. pt.” stand for separating stream line, stagnation point and vortex center and degenerate stagnation point, respectively.
2.2. ANALYSIS OF FLOW NEAR A STAGNATION POINT FOR VORTEX BIRTH

effect along the free surface; vorticity at the free surface depends on the curvature of surface (Batchlor [1967]).

Because \( \dot{\gamma}_s = 0 \) along the free surface, all degenerate stagnation points are non-simple if the liquid behaves as a generalized Newtonian fluid. The angle between the free surface and a separating streamline is always \( \pi/2 \) as discussed by Lutz [1987]. When the extensional rate vanishes, the separation point becomes a degenerate stagnation point and the velocity gradient is zero. This marks the onset of a stagnation point split and vortex birth.

In summary, the condition at the onset of vortex birth at a free surface is the same as the vortex birth condition for a birth inside the flow: zero determinant of velocity gradient at a stagnation point. Due to the kinematic boundary conditions at the free surface, this condition corresponds to a zero extensional rate. For generalized Newtonian fluid, the angle between the separating streamline and free surface is always \( \pi/2 \), and the onset of separation point is always from a non-simple degenerate stagnation point.

For Newtonian fluid, Eq. (2.26) can be written in terms of stress component: \( \det(\nabla \mathbf{u}) = -\left(\tau_{tt}/2\mu\right)^2 \), where \( \mu \) is Newtonian viscosity. This is the same as the result obtained by Brøns [1994].

2.2.3 Stagnation point at stationary solid wall

As in the case of free surfaces, a solid wall also has two natural characteristic directions, normal and tangent to the wall. The unit vectors along these directions are the natural choice for a basis set to describe the kinematics of the flow near a point at the solid wall. In the analysis presented here, the wall curvature is neglected.

The boundary conditions along a stationary solid surface are the no-slip and no-penetration conditions: \( \mathbf{t} \cdot \mathbf{u} = u_t = 0 \) and \( \mathbf{n} \cdot \mathbf{u} = u_n = 0 \). Because the velocity components are all zero along the wall, the derivatives of the velocity components along the wall also vanish: \( \partial u_n / \partial s_t = 0 \), \( \partial u_t / \partial s_t = 0 \).
2.2. ANALYSIS OF FLOW NEAR A STAGNATION POINT FOR VORTEX BIRTH

The shear rate, vorticity and extensional rate are
\[ \dot{\gamma}_s = \frac{1}{2} \left( \frac{\partial \mathbf{u}_t}{\partial s_n} \right), \]
\[ \omega_s = -\frac{\partial \mathbf{u}_t}{\partial s_n}, \]
and \( \dot{e} = 0. \) Therefore the velocity gradient at the wall in terms of the basis set \( \mathbf{B'} = (n, t)^T \) is
\[ \nabla \mathbf{u} = n t \frac{\partial \mathbf{u}_t}{\partial s_n} = n t (2\dot{\gamma}_s). \] (2.33)

The determinant of the velocity gradient is identically zero and consequently, the kinematics near the wall is described by higher order term \( \nabla \nabla \mathbf{u} \).

Due to the fact that velocity does not change along the wall, some second spatial derivatives of the velocity components vanish as well:
\[ \frac{\partial^2 u_n}{\partial t \partial n} = \frac{\partial}{\partial t} \left( \frac{\partial u_n}{\partial n} \right) = -\frac{\partial}{\partial t} \left( \frac{\partial u_t}{\partial t} \right) = 0, \]
\[ \frac{\partial^2 u_n}{\partial t^2} = \frac{\partial}{\partial t} \left( \frac{\partial u_n}{\partial t} \right) = 0, \]
\[ \frac{\partial^2 u_t}{\partial t^2} = \frac{\partial}{\partial t} \left( \frac{\partial u_t}{\partial t} \right) = 0. \] (2.34)

The second order term \( \nabla \nabla \mathbf{u} \) can be simplified to
\[ \nabla \nabla \mathbf{u} = n n n \left( -\frac{\partial^2 u_t}{\partial s_n \partial s_t} \right) + n n t \left( \frac{\partial^2 u_t}{\partial s_n \partial s_t} \right) + n t t \left( \frac{\partial^2 u_t}{\partial s_n \partial s_t} \right) \]
\[ = n n n \left( 2 \frac{\partial \dot{\gamma}_s}{\partial s_t} \right) + n n t \left( 2 \frac{\partial \dot{\gamma}_s}{\partial s_t} \right) + n t t \left( -2 \frac{\partial \dot{\gamma}_s}{\partial s_t} \right). \] (2.35)

Using these informations, the linear and the quadratic terms of the Taylor series expansion of velocity become,
\[ (\mathbf{x} - \mathbf{x}_0) \cdot \nabla \mathbf{u} = t \left[ (2\dot{\gamma}_s)(s_n - s_{n0}) \right], \]
\[ (\mathbf{x} - \mathbf{x}_0)(\mathbf{x} - \mathbf{x}_0) : \nabla \nabla \mathbf{u} \]
\[ = n \left( 2 \frac{\partial \dot{\gamma}_s}{\partial s_t} \right) (s_n - s_{n0})^2 + t \left( 2 \frac{\partial \dot{\gamma}_s}{\partial s_n} \right) (s_n - s_{n0})^2 + t \left( -4 \frac{\partial \dot{\gamma}_s}{\partial s_t} \right) (s_n - s_{n0})(s_t - s_{t0}) \]
\[ = (\mathbf{x} - \mathbf{x}_0) \cdot \left[ n n \left( 2 \frac{\partial \dot{\gamma}_s}{\partial s_t} \right) + n t \left( 2 \frac{\partial \dot{\gamma}_s}{\partial s_n} \right) + t t \left( -4 \frac{\partial \dot{\gamma}_s}{\partial s_t} \right) \right] (s_n - s_{n0}) \]
\[ = (\mathbf{x} - \mathbf{x}_0) \cdot \mathbf{Q}(s_n - s_{n0}), \] (2.36)
where \( \mathbf{x}_0 \) is a point along the wall, \( \mathbf{x} = ts_t + n_s n \) is a point near \( \mathbf{x}_0 \) and \( \mathbf{Q} \) is the reduced quadratic term derived from \( \nabla \nabla \mathbf{u} \) and the simplifications associated with the boundary conditions along the wall.

Because all velocity components are zero along the wall, the Taylor series expansion of the velocity field, Eq. (2.2), becomes

\[
\frac{D \mathbf{x}}{Dt} = (\mathbf{x} - \mathbf{x}_0) \cdot \nabla \mathbf{u} + \frac{1}{2} (\mathbf{x} - \mathbf{x}_0)(\mathbf{x} - \mathbf{x}_0) : \nabla \nabla \mathbf{u} \\
= (s_n - s_{n0}) \left[ t (2 \dot{\gamma}_s) + \frac{1}{2} (\mathbf{x} - \mathbf{x}_0) \cdot \mathbf{Q} \right].
\] (2.37)

It is important to notice that the reduced quadratic term acts like a linear term: the quadratic term in Taylor series expansion, \( (\mathbf{x} - \mathbf{x}_0)(\mathbf{x} - \mathbf{x}_0) : \nabla \nabla \mathbf{u} \), becomes linear, \( (\mathbf{x} - \mathbf{x}_0) \cdot \mathbf{Q} \), due to the simplifications coming from the boundary conditions.

As expected, the velocity vanishes at \( s_n = s_{n0} \), i.e. \( \mathbf{u}(s_n = s_{n0}) = 0 \). As treated by Bakker (1991) and Hartnack (1999), one can choose an unit of time, \( \vartheta \), proportional to \( (s_n - s_{n0}) \) such that the singular character at the wall, caused by the \( (s_n - s_{n0}) \) term, can be eliminated. Then Eq. (2.37) becomes

\[
\frac{D \mathbf{x}}{Dt^*} = \left[ t (2 \dot{\gamma}_s) + \frac{1}{2} (\mathbf{x} - \mathbf{x}_0) \cdot \mathbf{Q} \right].
\] (2.38)

This linear system governs the local flow field at a point near the wall. The zeroth order term of Eq. (2.38) vanishes if the shear rate, \( (\partial u_t / \partial s_n) = 2 \dot{\gamma}_s \), is zero. In this case, the velocity profile perpendicular to the wall shows an inflection point at the wall. The point along the wall at which the shear rate vanishes is called a separation point, because flow is detached or attached at that point.

The determinant of the reduced quadratic term, \( \mathbf{Q} \), is

\[
\det \mathbf{Q} = -8 \left( \frac{\partial \dot{\gamma}}{\partial s_t} \right)^2.
\] (2.39)

The eigenvalues are

\[
\lambda^- = -2 \left( \frac{\partial \dot{\gamma}}{\partial s_t} \right), \quad \lambda^+ = \left( \frac{\partial \dot{\gamma}}{\partial s_t} \right),
\] (2.40)

which are positive or negative real numbers depending on the sign of the spatial derivative of the shear rate along the wall. The corresponding eigenvectors are

\[
\mathbf{z}_- = t \left( \frac{\partial \dot{\gamma}}{\partial s_n} \right) + n \left( -3 \frac{\partial \dot{\gamma}}{\partial s_t} \right), \quad \mathbf{z}_+ = t,
\] (2.41)
Vortex birth from stationary solid wall

\( \text{det} \, Q = 0 \)

(\( \frac{\partial^2 z}{\partial n^2} = 0 \))

(\( \text{No eigenvector when} \ \frac{\partial^2 z}{\partial n^2} = 0. \))

(\( \frac{\partial^2 z}{\partial t^2} = 0 \))

(b) Onset of sep. pt.

\( \text{det} \, Q < 0 \)

\( \frac{\partial^2 z}{\partial n^2} < 0 \)

\( \frac{\partial^2 z}{\partial t^2} > 0 \)

(c) Two sep. pt. and one vor. ct.

\( A: x(s_{i0}) \)

\( B: x(s_{i0} + \delta s_t) \)

\( \theta_A \)

\( \theta_B \)

\( \delta s_t \)

\( z^{w,A}_{\cdot} \)

\( z^{w,B}_{\cdot} \)

\( z^{w,A}_{\cdot} + \text{Vor. ct.} \)

**Figure 2.4:** Onset of vortex birth attached to a stationary solid surface from a simple degenerate point. “Sep. pt.”, “vor. ct.” and “deg. sep. pt.” stand for separation point, vortex center and degenerate separation point, respectively. Vortex birth is marked by the onset of the separation point, (b). After the onset, two separation points, \( A \) and \( B \), split from the degenerate point shown in (b), move away from each other and a vortex center is detached from the solid wall, as shown in (c).
2.2. ANALYSIS OF FLOW NEAR A STAGNATION POINT FOR VORTEX BIRTH

where $\mathbf{z}_w^-$ is the direction of the separating streamline at the wall and $\mathbf{z}_w^+$ is the direction tangent to the wall, as indicated in Fig. 2.4(c). The angle between the wall and the separating streamline $\theta_w$ is,

$$\theta_w = \begin{cases} \arctan \left( -3 \frac{\partial \dot{\gamma}}{\partial s} \frac{\partial \dot{\gamma}}{\partial s_n} \right) , & \frac{\partial \dot{\gamma}}{\partial s_n} \neq 0 \\ \frac{\pi}{2} , & \frac{\partial \dot{\gamma}}{\partial s_n} = 0 \end{cases}$$

(2.42)

If the derivative of the shear rate along the wall $\frac{\partial \dot{\gamma}}{\partial s}$ vanishes, the determinant of the reduced quadratic term, $\det \mathbf{Q}$, also vanishes, and the eigenvalues are identically zero. The separation point becomes degenerate. As in the free surface case, there are two kinds of degenerate separation point:

1. $\frac{\partial \dot{\gamma}}{\partial s_n}$ is non-zero.

   The reduced quadratic term, $\mathbf{Q}$, is non-zero but its determinant vanishes. The reduced quadratic term has eigenvalues equal to zero and both eigenvectors $\mathbf{z}_\pm^w$ are collapsed into a single eigenvector $\mathbf{z}_w^0 = t$. This is called the simple degenerate case. Figure 2.4 illustrates the flow evolution as a vortex is born or disappear from a separation point at a wall. The simple degenerate stagnation point splits into two separation points with two eigenvectors are $\mathbf{z}_\pm^{w,A}$ and $\mathbf{z}_\pm^{w,B}$, as shown in Fig. 2.4(c). The evolution in the reverse order, from Fig. 2.4(c) to (a), corresponds to merging two separation points into a single one. The two eigenvectors collapse into a single eigenvector tangent to the wall. The presence of a simple degenerate separation point at a stationary wall is the condition that marks the birth (or death) of a vortex.

2. $\frac{\partial \dot{\gamma}}{\partial s_n}$ is zero.

   In this case the reduced quadratic term is identically zero, there is no eigenvalue nor eigenvector associated with the separation point. This is called the non-simple degenerate case. Figure 2.5 sketches the appearance of this non-simple degenerate separation point from a flow pattern that contains two vortices attached to each other and to a wall. As the recirculations shrink, the four saddle
points approach each other until they collapse into a single non-simple degenerate stagnation point.

As in the free surface case, the two cases of degenerate separation point discussed before mark the birth (or death) of a vortex. However, a degenerate separation point does not always represent the birth of a vortex. Bakker (1991) showed that a simple degenerate point at a wall may occur by the collapse of two separation points that are not connected by the same streamline, as sketched in Fig. 2.6. The birth (or death) of a vortex occurs only when two separation points that are connected by a streamline.

![Flow past cylinder](image)

**Figure 2.5:** Onset of vortex birth attached to a stationary solid surface from a non simple degenerate stagnation point. "Stag. pt.", "Sep. pt.", "vor. ct." and "deg. st. pt." stand for stagnation point, separation point, vortex center and degenerate stagnation point, respectively. This situation occurs in the flow past a cylinder. The situation sketched in (a) shows three stagnation points along the wall, two vortices and one saddle point inside the flow. As flow parameter changes, point $A$, $B$, and $D$ approaches $C$. Eventually all six points are merged into a single non-simple degenerated stagnation point, shown in (b). Progress from (a) to (b) stands for a "death" of vortex; the reverse process corresponds to a vortex birth.
2.2. ANALYSIS OF FLOW NEAR A STAGNATION POINT FOR VORTEX BIRTH

Detachment of saddle point from stationary solid wall

(a)

Separating streamline

(b)

Degenerate separation point

(c)

Saddle point

Figure 2.6: Saddle point birth from a solid wall [Bakker 1991]. When two adjacent separation points that are not connected by the same separating streamline collapse, a vortex does not disappear, as sketched in Fig. 2.4. In this case, the collapse represents a saddle point birth, as indicated in (c).

Split (or collapse) into a single degenerate separation point, as illustrated in Fig. 2.4 and 2.5. Mathematically, the birth condition can be represented by

$$\frac{d\gamma}{ds_t}(s_{t0}) = \lim_{\delta s_t \to 0} \frac{\gamma(s_{t0} + \delta s_t) - \gamma(s_{t0})}{\delta s_t} = 0,$$

(2.43)

where $s_{t0}$ is arc-length coordinate along the wall, and $\delta s_t$ is the distance between the two separation points. Note that this condition implies in a zero determinant of the reduced quadratic term.

In general, it is impossible to use dynamic variables, such as pressure, shear stress and derivative of shear stress, to describe the triadic components of $\nabla \nabla \mathbf{u}$, due to the
2.2. ANALYSIS OF FLOW NEAR A STAGNATION POINT FOR VORTEX BIRTH

fact that complex fluids, including generalized Newtonian fluids, show non-linear relationship between the state of stress and the state of strain-rate. However the vortex birth condition can be written in terms of the stress field for Newtonian fluids. From the steady-state Navier-Stokes equation, the pressure gradient along the tangential direction can be written in terms of the derivative of the shear rate with respect to the arc length normal to the wall.

\[
\frac{\partial P}{\partial s_t} = \mathbf{t} \cdot \nabla P = \mathbf{t} \cdot (-\rho \mathbf{u} \cdot \nabla \mathbf{u} + \nabla \cdot \tau)
\]

\[
= \mathbf{t} \cdot \mu \nabla^2 \mathbf{u} = \mu \frac{\partial^2 u_t}{\partial s_n^2} = \mu \frac{\partial \dot{\gamma}}{\partial s_n},
\]

(2.44)

where \(\mu\) is Newtonian viscosity. Note that at the solid surface the inertial term is absent due to the no-slip boundary condition. Hence, for Newtonian fluid, non-zero pressure gradient corresponds to a simple degeneracy case and zero pressure gradient along the wall corresponds to a non-simple degeneracy case.

The shear stress and its derivative along the wall can be expressed as

\[
\tau_{tn} = 2\mu \dot{\gamma}, \quad \frac{\partial \tau_{tn}}{\partial s_t} = \mu \left( \frac{\partial^2 u_t}{\partial s_t \partial s_n} \right) = \mu \left( 2 \frac{\partial \dot{\gamma}}{\partial s_t} \right)
\]

(2.45)

Using Eq. (2.44) and (2.45), the birth of vortex condition becomes zero shear stress at the wall, \(\tau_{tn} = 0\), and vanishing derivative of shear stress along the wall, \(d \tau_{tn}/ds_t = 0\).

The eigenvalues of the reduced quadratic term \(\mathbf{Q}\) can be written in terms of the derivative of the shear stress and pressure along the wall:

\[
\lambda_-^w = -2 \left( \frac{\partial \tau_{tn}}{\partial s_t} \right), \quad \lambda_+^w = 1 \left( \frac{\partial \tau_{tn}}{\partial s_t} \right),
\]

(2.46)

and eigenvectors

\[
\mathbf{z}_-^w = \mathbf{t} \frac{\partial P}{\partial s_t} + \mathbf{n} \left( -3 \frac{\partial \tau_{tn}}{\partial s_t} \right), \quad \mathbf{z}_+^w = \mathbf{t}.
\]

(2.47)

Equation (2.42) becomes

\[
\theta^w = \begin{cases} 
\arctan \left( -3 \frac{\partial \tau_{tn}}{\partial s_t} \right), & \frac{\partial P}{\partial s_t} \neq 0 \\
\frac{\pi}{2}, & \frac{\partial P}{\partial s_t} = 0
\end{cases}
\]

(2.48)
This equation is known as Ostwatch-Legendra solution (Bakker, 1991; Hartnack, 1999).

For Newtonian liquid, the presence and collapse of a recirculation can be defined in terms of the shear stress and pressure field. A recirculation attached to the wall, as the one sketched in Fig. 2.4 (c), only occurs if the adverse pressure gradient is strong enough. As the pressure gradient falls, the two separation points approach each other until they collapse into a simple degenerate separation point. The non-simple degenerate separation point, illustrated in Fig. 2.5, occurs when the pressure gradient is zero, as in the separation point located in the symmetry line behind a cylinder.

2.3 Final remarks

In a two dimensional, steady and incompressible flow, a stagnation point can be a vortex center, a saddle point or a separation point. Local flow features near a stagnation point can be analyzed by Taylor series expansion of the velocity field around that point. The birth of a vortex in a flow as parameters are changed occurs when multiple stagnation points split off from a degenerate stagnation point, usually accompanied by change of type of the stagnation point. Except at stationary solid walls, this event occurs when the determinant of the velocity gradient at the stagnation point vanishes. Along stationary solid walls, because of the no-slip and no-penetration, the local flow feature is described by the quadratic term of the Taylor series expansion. This quadratic term can be simplified leading to a reduced quadratic term.

A comprehensive view of vortex birth in flow was presented. The description of the local flow features shows that the birth of a vortex inside the flow, at a free surface or a wall is marked by changes on the eigenvalues and eigenvectors of the velocity gradient or the reduced quadratic term, in the case of vortex birth from a wall. The eigendirections are directly related to the directions of separating streamlines at the separation points. The conditions at which they occur can be expressed in terms of kinematic variables. The are summarized in Table 2.1.
### Table 2.1: Summary of vortex birth condition in steady two-dimensional incompressible flow

<table>
<thead>
<tr>
<th>Birth type</th>
<th>Vortex birth condition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inside flow</td>
<td>$\mathbf{u} = 0, \det(\nabla \mathbf{u}) = 0$</td>
</tr>
<tr>
<td>From free surface</td>
<td>$u_t = 0, \quad u_z = 0, \quad \dot{\varepsilon}<em>s = 0, \quad \tau</em>{tt} = 0$</td>
</tr>
<tr>
<td>From solid surface</td>
<td>$\dot{\gamma}<em>s = 0, \quad \tau</em>{tn} = 0, \quad \partial \dot{\gamma}<em>s / \partial s_t = 0, \quad \partial \tau</em>{tn} / \partial s_t = 0$</td>
</tr>
</tbody>
</table>
Chapter 3

Tracking birth of vortex in flows

3.1 Introduction

Vortices in flows are characterized by the rotating motion of material particles around a common center (Lugt, 1983). They can be visualized by examining pathlines constructed from theoretical models or experiments. Numerically or analytically computed streamlines or pathlines illustrate the presence of a vortex by generating multiple-closed loops around a common center. However, streamlines and pathlines are not invariant under a change of frame of reference. For example, a moving sphere in a stationary fluid has closed streamlines varying with time in a laboratory reference; however when the observer moves with the sphere, the flow becomes time-independent and the closed loop streamlines disappear (Lugt, 1983). This example shows that closed streamlines do not guarantee the existence of a vortex: without a proper frame of reference, a vortex can be invisible. In general, vortices show three dimensional spiral structure. But when the pathlines are perpendicular to the axis of rotation, they can be considered plane vortices.

The presence of vortices or recirculations in flows can lead to undesired effects in many industrial processes. In liquid coating, for example, the flow near the region where the liquid meets the moving substrate can develop microscopic gyres that are
3.1. INTRODUCTION

![Diagram of vortex in coating flows](image)

**Figure 3.1:** Vortex in coating flows. Roll coating, slide coating, and slot coating flow are illustrated, and possible locations of vortex for each flow system are specified here. Each case the schematic diagram comes from visualization of coating flows (Coyle 1984, Schweizer 1988, Sartor 1990).

intense and typically extend across the entire coating bead. These were found in experiments by Coyle (1984), Schweizer (1988), Sartor (1990) and others, as shown in Fig. 3.1. They tend to centrifuge denser particles, to desorb dissolved gas, to collect and discharge bubbles, to hold formulations long enough for unwanted flocculation or polymerization, and to become nodular along their length and thereby detract from cross-wise coating uniformity. Therefore the set of operating parameters at which the flow does not present vortices are preferred in order to avoid the problems listed above. It is crucial to map these conditions in the parameter space, and construct the so called vortex-free operating window. The appropriate frame of reference to analyze the presence of undesired vortices in coating flows is the laboratory frame of reference, at which the geometry of the flow does not change with time.

This vortex-free window in the parameter space can be simply obtained by post-processing a large number of steady state solutions at different flow conditions, covering the range of interest in all the flow parameters. This procedure can be called vortex birth capturing and it is extremely expensive, the number of solutions needed to construct the window can easily reach 1,000 and post-processing each of these solutions can be very tedious. We propose an alternative efficient way to construct the vortex-free window of a flow.
3.1. **INTRODUCTION**

Historically, vortex analysis is closely related to flow topology analysis (Bakker 1991; Lugt 1987; Brøns and Hartnack 1999). In two dimensional steady-state incompressible flows, the velocity field near a stagnation point may be described by Taylor series expansion around the stagnation point. The characteristics of the expansion terms define the structure of the pathlines in that region. When the stagnation point becomes degenerate, the structure of the pathlines around it changes dramatically. Some of these changes imply the birth of a new vortex.

The event of vortex birth, or death, is signalized by changes in the eigenvalues and eigenvectors of the linear term of the Taylor series expansion of the velocity field around a stagnation point. Three types of vortex birth – inside the flow, from free surface, and from wall – are examined, and local birth conditions are defined in terms of kinematic variables, such as extensional rate, shear rate and vorticity. In the previous chapter (Chap. 2), we present the vortex birth conditions, which are local flow features evaluated at stagnation points in the flow. In order to determine the set of flow parameters at which a vortex is born (or disappears), we need to find at which flow conditions the local vortex birth condition at stagnation points is satisfied, i.e. the local flow condition near stagnation points need to be coupled with the global flow parameters. They are summarized in Table 2.1.

In this study, we construct the vortex-free window by tracking the global flow conditions at which a recirculation appears (or disappears). This is done by performing a multiparameter continuation to obtain solutions only at the set of conditions at which the local vortex birth condition at stagnation points is satisfied. Therefore, the line (or surface) in the parameter space that defines the boundary of the vortex-free window is constructed automatically. The savings compared to the determination of the window by post-processing a large number of solutions is enormous.

The formulation of the augmented system of equations used in the multiparameter continuation is discussed as well as the solution method used. Three examples are presented, one where a vortex is born from a cusp point inside the flow, a second where it is born from a stationary wall, and a third where it is born from a free surface. All the examples are related to coating flows.
3.2 Automated tracking of vortex birth in flows

The multiparameter continuation described previously was done by augmenting the Navier–Stokes equation by equations that describe the vortex-birth condition. The solution of this augmented system was obtained by Galerkin’s finite element method. The formulation and solution method for the multiparameter continuation is described in this section.

3.2.1 Navier–Stokes system for viscous free surface flows

The velocity and pressure fields of incompressible pressure fields of two-dimensional, steady state flow of a Newtonian liquid are governed by the continuity and momentum equations:

\[
\nabla \cdot \mathbf{u} = 0, \quad \rho \mathbf{u} \cdot \nabla \mathbf{u} = \nabla \cdot \mathbf{T},
\]

where \( \rho \) is the liquid density and \( \mathbf{T} \) is stress tensor. For Newtonian liquid, it is given by \( -\mathbf{T} = -p \mathbf{I} + \mu \left[ \nabla \mathbf{u} + (\nabla \mathbf{u})^T \right] \), where \( p \) is the pressure and \( \mu \) is the liquid viscosity.

Boundary conditions are need to solve the Navier–Stokes system. In most case, the flow domain is bounded by inflow and outflow synthetic planes, solid wall and free surface (gas-liquid interface).

At inflow and outflow boundaries, either the velocity components are specified, or a fully developed flow condition is imposed, or the value of pressure is prescribed. The corresponding boundary conditions are:

\[
\mathbf{u} = \mathbf{V}_b(x), \quad n_b \cdot \nabla \mathbf{u} = 0, \quad p = p_b.
\]

\( \mathbf{V}_b(x) \) is the imposed velocity profile, \( p_b \) is the imposed pressure, and \( n_b \) is the unit normal vector to the boundary.
At rigid solid walls, the no-slip and no-penetration conditions are imposed:

\[ t_w \cdot u = V_{wall}, \quad n_w \cdot u = 0, \quad (3.5) \]

where \( V_{wall} \) is the solid wall velocity, \( n_w \) and \( t_w \) are the unit normal and tangent vectors to the wall.

Along free surfaces, a force balance is imposed and the no-penetration condition (kinematic condition) is used:

\[ n_f \cdot T = \sigma \frac{dt_f}{ds} - n_f P_{amb}, \quad (3.6) \]
\[ n_f \cdot u = 0, \quad (3.7) \]

where \( t_f \) and \( n_f \) are the local unit tangent and unit normal to the free surface, \( s \) is the arc-length coordinate along the interface, \( \sigma \) is the liquid surface tension and \( P_{amb} \) is the ambient pressure.

Flows with free surface give rise to a free boundary problem. The flow domain is unknown a priori and it is part of the solution.

To solve a free boundary problem by means of standard techniques for boundary value problems, the set of differential equations and boundary conditions posed in the unknown physical domain have to be transformed to an equivalent set defined in a known, fixed computational domain. This transformation is made by a mapping \( x = x(\xi) \) that connects the two domains. The physical domain is parameterized by the position vector \( x = (x, y) \), and the reference domain, by \( \xi = (\xi, \eta) \). The mapping used here is the one described by de Santos (1991). The inverse mapping is governed by a system of elliptic differential equations identical to those encountered in the dilute regime of diffusional transport.

\[ \nabla \cdot D_\xi(\xi, \eta) \nabla \xi = 0, \quad \nabla \cdot D_\eta(\xi, \eta) \nabla \eta = 0. \quad (3.8) \]

\( D_\xi \) and \( D_\eta \) are mesh diffusivities which control the steepness of gradients in the node spacing by adjusting the potentials \( \xi \) and \( \eta \). Curves of constant \( \xi \) and \( \eta \) define the boundaries of elements used to describe the domain. The cross point of these curves of sets the position of a node. Boundary conditions are needed to solve the second-order
3.2. AUTOMATED TRACKING OF VORTEX BIRTH IN FLOWS

differential equations (3.8). Solid walls and inflow and outflow planes are described by
the function that defines their geometry and nodes were distributed along them by a
specified stretching function. The location of the free surface is implicitly determined
by the kinematic condition, Eq. (3.7). The discrete version of the mapping equations is
generally referred to as mesh generation equations. Detailed procedure and boundary
conditions for mesh equations are discussed in de Santos (1991) and Benjamin (1994).

3.2.2 Solution of the Navier–Stokes system for free surface
flow by G/FEM

Galerkin finite element method is used to solve the Navier–Stokes system coupled
with the mesh generation equation, equations (3.1) and (3.8). Each independent
variable, velocity, pressure and position, is approximated by a linear combination of
a finite number of basis functions:

\[ u = \sum_i U_i \phi_i(\xi, \eta), \]  
\[ p = \sum_k P_k \psi_k(\xi, \eta), \]  
\[ x = \sum_i X_i \phi_i(\xi, \eta). \]

\( U_i, P_k \) and \( X_i \) are the basis function coefficients, the unknowns of the discretized
problem. The velocity and nodal position are represented in terms of Lagrangian
bi-quadratic function \( \phi_i(\xi, \eta) \), and the pressure in terms of linear discontinuous basis
function \( \psi_k(\xi, \eta) \).

The weak form of equations (3.1) and (3.8) are obtained by multiplying each equa-
tion by weighting functions, integrating over the physical domain, and applying the
divergence theorem to the appropriate terms with divergence. In Galerkin’s method,
3.2. AUTOMATED TRACKING OF VORTEX BIRTH IN FLOWS

The weighting and basis functions are the same. The weighted residuals are:

\[ R_m = \int_A \phi^i \rho \mathbf{u} \cdot \nabla \mathbf{u} + \mathbf{T} \cdot \nabla \phi^i \, dA - \int_{\partial A} \phi^i \mathbf{n} \cdot \mathbf{T} \, ds, \]  
\[ R_c = \int_A \psi^k \nabla \cdot \mathbf{u} \, dA, \]  
\[ R^\xi_x = \int_A D_\xi \nabla \xi \cdot \nabla \phi^i \, dA + \int_{\partial A} \phi^i D_\xi \mathbf{n} \cdot \nabla \xi \, ds, \]  
\[ R^\eta_y = \int_A D_\eta \nabla \eta \cdot \nabla \phi^i \, dA + \int_{\partial A} \phi^i D_\eta \mathbf{n} \cdot \nabla \eta \, ds, \]  

where \( A \) is the flow domain and \( \partial A \) is the domain boundary and \( s \) is arc-length coordinate along the boundary. The superscripts, \( m, c \) and \( x \), denote momentum, continuity and mesh residuals, respectively. Essential boundary conditions were imposed by replacing the corresponding weighted residual equation with the desired velocity or node specification. Natural boundary conditions were applied through the boundary integrals in equations (3.12) and (3.14).

In sum, the Galerkin finite element method reduces the Navier–Stokes and mesh generation differential equations to a set of nonlinear algebraic equations on the basis functions coefficients.

\[ R(z, \lambda) = 0, \]  

where \( z \) is the solution vector which consist of velocity \( \mathbf{u} \), pressure \( P \) and position \( \mathbf{x} \), and \( \lambda \) is a vector that contains the \( M \) parameters on which the system depends. Equation (3.15) is solved iteratively by Newton’s method:

\[ J^{(i)}(z^{(i)}, \lambda) \delta z^{(i)} = -R^{(i)}(z^{(i)}, \lambda), \]  
\[ z^{(i+1)} = z^{(i)} + \delta z^{(i)}, \]  

the index \( i \) and \( i + 1 \) indicate the current and next Newton’s step. \( J^{(i)} \equiv \partial R^{(i)}/\partial z^{(i)} \) is the Jacobian matrix. The iteration continues until \( ||R^{(i)}||_2 < \epsilon \). Here we choose \( 10^{-8} \) as \( \epsilon \).
3.2. AUTOMATED TRACKING OF VORTEX BIRTH IN FLOWS

3.2.3 Tracking the vortex birth condition

Following the procedure described in the previous section, one can get a flow state at specified set of flow parameters.

In order to use Galerkin’s finite element method to solve the augmented system, the conditions described in Table 2.1 need to be written in terms of the finite element basis functions. For a vortex birth inside the flow, the discretized birth conditions are:

\[
\begin{align*}
  u(x^*, y^*) &= \sum_i U_i \phi_i(x^*, y^*) = 0, \\
  v(x^*, y^*) &= \sum_i V_i \phi_i(x^*, y^*) = 0, \\
  \det \nabla u(x^*, y^*) &= \frac{\partial u}{\partial x}(x^*, y^*) \frac{\partial v}{\partial y}(x^*, y^*) - \frac{\partial v}{\partial x}(x^*, y^*) \frac{\partial u}{\partial y}(x^*, y^*) \\
  &= \left( \sum_i U_i \frac{\partial \phi_i}{\partial x}(x^*, y^*) \right) \left( \sum_i V_i \frac{\partial \phi_i}{\partial y}(x^*, y^*) \right) \\
  &\quad - \left( \sum_i V_i \frac{\partial \phi_i}{\partial x}(x^*, y^*) \right) \left( \sum_i U_i \frac{\partial \phi_i}{\partial y}(x^*, y^*) \right) = 0, \quad (3.17)
\end{align*}
\]

where \( u \) and \( v \) are the velocity component in \( x \) and \( y \) direction, \( U_i \) and \( V_i \) are the nodal value of velocity components, and \((x^*, y^*)\) stands for the coordinate of the vortex birth point.
3.2. AUTOMATED TRACKING OF VORTEX BIRTH IN FLOWS

The discretized conditions for a vortex birth from free surface are:

\[
\begin{align*}
    u_t(x^*, y^*) &= (t \cdot u)(x^*, y^*) \\
    &= \left( t_x \sum_i U_i \phi_i \right)_{|_{(x^*, y^*)}} + \left( t_y \sum_i V_i \phi_i \right)_{|_{(x^*, y^*)}} = 0, \\
    \tau_{tt}(x^*, y^*) &= (tt : T)(x^*, y^*) \\
    &= \left[ t_x t_x \mu \left( 2 \sum_i U_i \frac{\partial \phi_i}{\partial x} \right) + t_x t_y \mu \left( \sum_i U_i \frac{\partial \phi_i}{\partial y} \sum_i V_i \frac{\partial \phi_i}{\partial x} \right) \\
    &\quad+ t_y t_x \mu \left( \sum_i U_i \frac{\partial \phi_i}{\partial y} \sum_i V_i \frac{\partial \phi_i}{\partial x} \right) \right]_{|_{(x^*, y^*)}} = 0,
\end{align*}
\]

(3.18)

where \( t \) is the tangent vector to the free surface.

The birth conditions from a solid surface are related to the shear stress and its derivative with respect to the arc-length coordinate, \( \tau_{tn} = 0 \) and \( \partial \tau_{tn} / \partial s_t = 0 \). When quadratic basis functions are used to expand the velocity field, the stress is linear inside each element and its derivative is constant. Therefore, it is impossible to locate a point inside an element from which a vortex is born. Instead of this condition, we use an “alleviated” version. The vortex birth (or death) condition at solid surface can be interpreted as two (or more) adjacent stagnation points that split (or collapse) from (into) a single degenerate stagnation point. Therefore the birth condition at a solid wall can be approximated as

\[
\frac{\partial \tau_{tn}(s_t^*)}{\partial s_t} \approx \frac{\tau_{tn}(s_t^* + \delta s_t) - \tau_{tn}(s_t^*)}{\delta s_t} = 0,
\]

(3.19)

where \( s_t^* \) is the arc-length coordinate along the solid surface at the vortex birth point, \( \delta s_t \) is the distance between the two separated but close stagnation points. Therefore, vortex birth is approximated as the condition at which adjacent stagnation points are located close enough to each other. Using this fact, the vortex birth condition for
Newtonian liquid becomes,

\[
\tau_{tn}(s^*_t) = (\mathbf{t} : \mathbf{T})(s^*_t) \\
= \left[ t_x n_x \mu \left( 2 \sum_i U_i \frac{\partial \phi_i}{\partial x} \right) + t_x n_y \mu \left( \sum_i U_i \frac{\partial \phi_i}{\partial y} \sum_i V_i \frac{\partial \phi_i}{\partial x} \right) \\
+ t_y n_x \mu \left( \sum_i U_i \frac{\partial \phi_i}{\partial y} \sum_i V_i \frac{\partial \phi_i}{\partial x} \right) \\
+ t_y n_y \mu \left( 2 \sum_i V_i \frac{\partial \phi_i}{\partial y} \right) \right]_{(s_t, s^*_t)} = 0
\]

\[
\tau_{tn}(s^*_t + \delta s_t) = (\mathbf{t} : \mathbf{T})(s^*_t + \delta s_t) = 0,
\] (3.20)

where \( \mathbf{t} = t_x \mathbf{i} + t_y \mathbf{j} \) and \( \mathbf{n} = n_x \mathbf{i} + n_y \mathbf{j} \) are the tangent and normal vector to the solid surface. This condition is the one used by Yeckel and Scriven (1992) and Yeckel (1998) to describe the birth of a vortex inside the feed slot of a slot coating flow. In this study, we set \( \delta s_t \) to be equal to the size of the largest element along the solid wall boundary. It is impossible to have more than one root of the shear stress distribution along a single element because of the quadratic basis function used for the velocity field.

### 3.2.4 Augmented Navier–Stokes system

The augmented system of algebraic equations consists of the algebraic equations that relate the finite element coefficients of the unknown fields and the conditions of vortex birth. The set of unknowns is also augmented by the number of algebraic equations that define the vortex birth condition, Three in the case of vortex birth inside the flow – Eqs. (3.17), and two in the case of vortex birth at a free surface or solid wall – Eqs. (3.18) or (3.20). The extra unknowns are the position of the stagnation point at which a vortex is born from and one of the flow parameters \( \lambda_1 \) that is not fixed. Actually, the solution of the augmented system will give the value of this parameter at which the vortex birth occurs. For a birth inside a flow, two coordinates \((x^*, y^*)\) are required to locate the stagnation point. For a birth attached to a flow boundary, either solid surface or free surface, only one coordinate is enough to locate the birth point.
The augmented system of equations can be
\[
\begin{align*}
\mathbf{R}(z, \lambda, p) &= 0 \\
\mathbf{A}(z, \lambda, p) &= 0
\end{align*}
\]

where \( z \) and \( \lambda \) are the vectors that contain the finite element coefficients and fixed flow parameters, as defined in Eq. (3.16), and \( p \) is a vector that contains the extra set of unknowns of the augmented problem, e.g. \( p = (x^*, \lambda_1) \). \( A \) is the set of algebraic equations that defines the vortex birth condition.

This non-linear system is solved by Newton’s method, which requires the evaluation of the Jacobian matrix of the system:

\[
\begin{bmatrix}
\frac{\partial \mathbf{R}}{\partial z} & \frac{\partial \mathbf{R}}{\partial p} \\
\frac{\partial \mathbf{A}}{\partial z} & \frac{\partial \mathbf{A}}{\partial p}
\end{bmatrix}
\begin{bmatrix}
\delta z \\
\delta p
\end{bmatrix}
= -
\begin{bmatrix}
\mathbf{R} \\
\mathbf{A}
\end{bmatrix}
\] (3.21)

where \( \frac{\partial \mathbf{R}}{\partial p} \) are the sensitivity of the residual equations to the extra unknowns and \( \frac{\partial \mathbf{A}}{\partial p} \) are the sensitivity of the vortex birth conditions to the flow field (finite element coefficients) and extra unknowns, respectively. \( \frac{\partial \mathbf{R}}{\partial z} \) is the Jacobian matrix of the original problem.

Because the new three blocks are usually small sub-matrices that are generally densely populated, the fill structure of the new matrix is that of an arrow head, which destroys the benefit of most direct sparse linear solver \cite{Musson2001}. This difficulty can be alleviated by employing the bordering algorithm \cite{Keller1977}, or block elimination on the solution of the linear system at each Newton iteration. The basic idea is to decompose the matrix-vector equations and solve them sequentially.

To construct the augmented Jacobian matrix, we need to evaluate these three new blocks. The sensitivity of the vortex birth conditions to the finite element coefficients \( \frac{\partial \mathbf{A}}{\partial z} \) is easily calculated, because the velocity and pressure field explicitly appear in the algebraic equations that define a vortex birth.

The derivative of the Navier–Stokes residual with respect to the auxiliary parameters \( p \) is not readily available. When a given parameter explicitly appears in the Navier–Stokes equation, the evaluation of the sensitivity is straightforward. For example,
if the parameter $\lambda_1$ that belongs to $\mathbf{p}$ is the Reynolds number ($\lambda_1 = N_{Re}$), analytical entries for $\partial \mathbf{R}/\partial N_{Re}$ can be easily computed. If the set of equations depends on the parameter implicitly, for example, if the free parameter is a geometrical parameter that defines a flow boundary, it is impossible to construct $(\partial \mathbf{R}/\partial \lambda_1)$ analytically. However, it can be easily approximated by a finite difference quotient, as the central difference secant approximation to the tangent (Musson [2001]):

$$\frac{\partial \mathbf{R}}{\partial \lambda_1} \approx \frac{\mathbf{R}(\lambda_1 + \epsilon_\lambda) - \mathbf{R}(\lambda_1 - \epsilon_\lambda)}{2 \epsilon_\lambda}. \quad (3.22)$$

Here $\epsilon_\lambda$ is a small number, say, $\epsilon_\lambda = \lambda_1 \times 10^{-6}$, or $10^{-6}$ when $\lambda_1$ is zero.

The derivative of the augment equations with respect to the extra unknowns $\partial \mathbf{A}/\partial \mathbf{p}$ can be a problem. The augment equations used here are based on local flow features near stagnation point. In finite element context, the equations that describe the conditions for vortex birth are evaluated inside or along the element which contains the stagnation point “under surveillance”. However, the entire flow field is affected by changes of the flow parameter $\lambda_1$. Therefore the dependence of the augment equations on the unknown flow parameter $\lambda_1$ and on the location of the stagnation point $(x^*, y^*)$ is not explicit, and the evaluation of the sensitivity of augment equations with respect to extra unknown vector $\partial \mathbf{A}/\partial \mathbf{p}$ is not straight forward to compute.

For example, the determinant of velocity gradient during $i^{th}$ Newton’s step can be represented as $(\text{det} \nabla \mathbf{u}^{(i)}(\mathbf{z}^{(i)}(\lambda_1), x^{*(i)}, y^{*(i)})$. The first argument indicates the effect of changes in the parameter $\lambda_1$ on the solution vector — the indirect dependence. The second and third arguments indicate the direct dependence on the $i^{th}$ step vortex birth position, $(x^{*(i)}, y^{*(i)})$. In this case, the augmented Jacobian entries corresponding to
extra unknowns can be approximated as
\[
\frac{\partial (\text{det} \, \nabla u)^{(i)}}{\partial \lambda_1} (z^{(i)}(\lambda_1), x^{s(i)}, y^{s(i)}) \\
\approx \frac{\text{det} \, \nabla u)^{(i)}}{\partial x^*} (z^{(i)}(\lambda_1), x^{s(i)}, y^{s(i)}) \\
\approx \frac{\text{det} \, \nabla u)^{(i)}}{\partial y^*} (z^{(i)}(\lambda_1), x^{s(i)}, y^{s(i)})
\]
where \(\epsilon_x\) and \(\epsilon_y\) are \(10^{-6}\) in this study.

The evaluation of Eqs. (3.24) and (3.25) are straight forward: each term in numerator are just evaluated at different positions at the given parameter \(\lambda_1\). However, Eq. (3.23) needs a special treatment because it is not trivial to obtain \(z^{(i)}(\lambda_1 + \epsilon_\lambda)\) which means the \(i^{th}\) step solution vector with the perturbed parameter. Once you solve the system at \(\lambda_1 + \epsilon_\lambda\), the resulting solution vector does not belonged to \(i^{th}\) Newton step of the system at \(\lambda_1\) anymore!

A simple way to approximate \(z^{(i)}(\lambda_1 + \epsilon_\lambda)\) is to use Taylor series expansion around \(\lambda_1\):
\[
z^{(i)}(\lambda_1 + \epsilon_\lambda) = z^{(i)}(\lambda_1) + \epsilon_\lambda \frac{\partial z^{(i)}(\lambda_1)}{\partial \lambda_1} + \epsilon_\lambda^2 \frac{\partial^2 z^{(i)}(\lambda_1)}{\partial \lambda_1^2} + \ldots
\]
Because \(\epsilon_\lambda\) is a small number, higher order term can be neglected. \(z^{(i)}(\lambda_1)\) is already known by solving Eq. (3.16) of \((i - 1)^{th}\) step. \(\partial z^{(i)}(\lambda_1)/\partial \lambda_1\) can be obtained by
\[
J^{(i)}(z^{(i)}(\lambda_1)) \frac{\partial z^{(i)}(\lambda_1)}{\partial \lambda_1} = -\frac{\partial R^{(i)}}{\partial \lambda_1} (z^{(i)}(\lambda_1)).
\]

The Jacobian matrix is the same as Eq. (3.16) and the right hand side can be approximated as in Eq. (3.22). This equation will appear in the predictor step of the
continuation procedure in direct tracking algorithm — the result can be reused, improving the efficiency of the method.

### 3.2.5 Direct tracking algorithm

Using the augmented Navier–Stokes system and the solution strategy discussed on Sec. 3.2.4, one can perform direct tracking of vortex birth: an automated vortex birth tracking algorithm. Multiparameter continuation is the heart of this algorithm.

The basic scheme is shown in Fig. 3.2. \( \alpha \) and \( \beta \) stand for chosen operating or design parameters from the parameter space that define a plane at which a vortex-free window will be constructed. This plane is a cut on a multi-dimensional parameter space. Specifically, \( \alpha \) is the parameter which will remain part of the solution of the augmented Navier–Stokes system, and \( \beta \) is the control variable, which is changed by a user-defined rule during continuation.

Step I in Fig. 3.2 represents the evaluation of a flow state at a fixed set of parameters \( \alpha \) and \( \beta \) which is close enough to a vortex birth state. Either solution, with or without vortex, will work but “close enough” means it should provide a good initial guess for step II.

Step II corresponds to the search of the first vortex birth state at a fixed value of \( \beta \), that is, finding the critical value of \( \alpha \) at which the vortex birth condition is satisfied at a fixed \( \beta \). For solving the augmented Navier–Stokes system, as explained in Sec. 3.2.4, Newton’s method is used until the \( \mathcal{L}_2 \) norm of the augmented residual, \( \| R^A(z) \|_2 \), is less than \( 10^{-8} \). This step gives the starting point of the direct tracking procedure.

Steps III to V is the multi-parameter continuation: increase or decrease \( \beta \) based on a specified rule, and find the critical value of \( \alpha \) at fixed \( \beta \) at which a vortex is born. Cycling process through III to V will continue to construct the boundary of the vortex-free window.

Step III corresponds to changing \( \beta \) based on a given rule. One can apply an adaptive step-size control strategy in this step based on the result from the predictor or cor-
3.2. **AUTOMATED TRACKING OF VORTEX BIRTH IN FLOWS**

**Figure 3.2:** Schematic procedure of direct tracking algorithm. $\alpha$ and $\beta$ are the chosen operating or design parameter and NS stands for Navier–Stokes system.
3.2. AUTOMATED TRACKING OF VORTEX BIRTH IN FLOWS

rector step during the continuation, as described in Seydel (1994). In this study, we simply used \( \beta_{(i+1)} = \beta_{(i)} + \delta \beta \), where subscript stands for the number of continuation step and \( \delta \beta \) is a given step size that depends on the flow.

Step IV is the predictor step in the continuation which generate the “good” initial guess for the corrector step. According to Seydel (1994), there are two types of predictor: tangent predictor or polynomial extrapolation. In this study, we use the tangent predictor type. Because the control variable is \( \beta \), the equation used to evaluate the tangent, \( \partial z^A / \partial \beta \), is,

\[
\begin{bmatrix}
J & \partial R \\
\partial z^A & \partial A \\
\partial p & \partial p
\end{bmatrix} \begin{bmatrix}
\partial z \\
\partial \beta
\end{bmatrix} = -\begin{bmatrix}
\partial R \\
\partial A \\
\partial \beta
\end{bmatrix}.
\]

Note that the Jacobian of the augmented system is the same as Eq. (3.21). If \( \beta \) explicitly appears on \( A \), one can use an analytic expansion for \( \partial A / \partial \beta \). If not, one can use the same technique used in Eq. (3.22) to approximate the derivative by a secant. The other blocks were already discussed in Section 3.2.4. Using the tangent predictor, an initial guess for the corrector iterations is computed by \( \tilde{z}^A_{(i+1)} = z^A_{(i)} + \delta \beta (\partial z^A_{(i)} / \partial \beta) \), where \( \tilde{z}^A_{(i+1)} \) stands for the initial guess for \( (i+1)^{th} \) continuation step.

Step V is the corrector step. The type of corrector depends on how the curve is parameterized. The parameter used for describing the curve is called the curve parameter (Seydel 1994). What we called first-order or natural continuation is that the curve parameter is the control variable, \( \beta \). In this case, Jacobian matrix and the residual vector of corrector step are basically the same as augmented Navier–Stokes system, Eq. (3.21). Step V is similar to step II. To avoid singular points on the branch, as turning points, the curve need to be parameterized properly. Typically, pseudo arc-length parameterization is used: add one more equation, arc-length definition, to augment the residual, and one more parameter, arc-length \( s \), to the auxiliary parameter vector. Detailed methods about pseudo arc-length continuation are described in Chan (1984). In this study, we use first-order continuation to construct the vortex birth curve of a given flow system.
3.3 Examples

As mentioned before, vortex in flows can cause problems in industrial processes, especially continuous coating of substrates. In this study, we choose two coating systems, slot coating and forward roll coating, as examples of automated tracking of vortex birth. By using the vortex birth tracking, we were able to create vortex-free windows which can be used as guidelines for operating conditions or design parameters in a very efficient way.

3.3.1 Single-layer slot coating

Single-layer slot coating is a high-precision coating method used to deposit a thin liquid film onto a moving substrate. It is classified as a pre-metered coating method: the coated thickness is directly controlled by the flow rate and web speed and independent of other process variables. However, the liquid flow in the application region, so called the *coating bead*, is strongly affected by operating parameters, liquid properties, and design parameters; such as web speed, surface tension, and geometry of the coating die. Extensive research has been done to find the region in the parameter space of acceptable coating quality (Higgins and Scriven, 1980; Sartor, 1990; Gates, 1999; Carvalho and Kheshgi, 2000). But here, we focused on the vortex birth inside coating bead and tracked it using direct-tracking.

In this study, the coating liquid is considered Newtonian. The geometry of the flow and the boundary conditions are summarized in Fig. 3.3(a).

The geometric parameters of the die shape considered here are the upstream gap $h_{ug}$, the downstream gap at the upstream corner $h_{dg,u}$ and the downstream gap at the downstream corner $h_{dg,d}$ and the length of the upstream and downstream lips $L_u$ and $L_d$, as indicated in Fig. 3.3(b). Note that the downstream die-lip angle $\theta$ is defined in terms of these variables.

$$\theta = \tan^{-1}\left(\frac{h_{dg,u} - h_{dg,d}}{L_d}\right).$$  \hspace{1cm} (3.29)
3.3. EXAMPLES

(a) Sketch of flow domain and boundary conditions used for a single-layer slot coating flow.

(b) Geometry parameters used for a single-layer slot coating flow.

(c) Mesh used to solve the governing equations by G/FEM 920 elements and 18300 unknown coefficients of the finite element basis functions are used to discretize the system.

**Figure 3.3:** single-layer slot coating: boundary condition, design parameter and mesh.
### Table 3.1: Parameters used in single-layer slot coating.

<table>
<thead>
<tr>
<th>Name</th>
<th>Unit</th>
<th>Base case</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Operating parameters</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Film thickness ((h_\infty))</td>
<td>mm</td>
<td>0.05</td>
</tr>
<tr>
<td>Web speed ((V_w))</td>
<td>m/sec</td>
<td>1</td>
</tr>
<tr>
<td>Vacuum pressure ((P_{vac}))</td>
<td>Pa</td>
<td>-1000</td>
</tr>
<tr>
<td>Ambient pressure ((P_{amb}))</td>
<td>Pa</td>
<td>0</td>
</tr>
<tr>
<td>Surface tension ((\sigma))</td>
<td>dyne/cm</td>
<td>61</td>
</tr>
<tr>
<td>Viscosity ((\mu))</td>
<td>mPa.s</td>
<td>2.3</td>
</tr>
<tr>
<td>Density ((\mu))</td>
<td>g/cm³</td>
<td>1.2</td>
</tr>
<tr>
<td>Dynamic contact angle ((\theta_d))</td>
<td>deg.</td>
<td>128</td>
</tr>
<tr>
<td>Static contact angle ((\theta_s))</td>
<td>deg.</td>
<td>62</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Name</th>
<th>Unit</th>
<th>Base case</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Model geometry parameters</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Upstream gap height ((h_{ug}))</td>
<td>mm</td>
<td>0.1</td>
</tr>
<tr>
<td>Downstream gap height at downstream corner ((h_{dg,d}))</td>
<td>mm</td>
<td>0.1</td>
</tr>
<tr>
<td>Downstream gap height at upstream corner ((h_{dg,u}))</td>
<td>mm</td>
<td>0.1</td>
</tr>
<tr>
<td>Downstream lip angle ((\theta))</td>
<td>deg.</td>
<td>0</td>
</tr>
<tr>
<td>Upstream lip length ((L_u))</td>
<td>mm</td>
<td>0.6</td>
</tr>
<tr>
<td>Downstream lip length ((L_d))</td>
<td>mm</td>
<td>0.6</td>
</tr>
<tr>
<td>Fully developed length ((L_\infty))</td>
<td>mm</td>
<td>1.1</td>
</tr>
<tr>
<td>Feed slot width ((h_s))</td>
<td>mm</td>
<td>0.1</td>
</tr>
<tr>
<td>Feed slot height ((h_{f,\infty}))</td>
<td>mm</td>
<td>0.6</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Name</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dimensionless flow rate ((q^*))</td>
<td>(\frac{h_\infty}{h_{ug}})</td>
</tr>
<tr>
<td>Dimensionless vacuum ((P_{vac}^*))</td>
<td>(\frac{P_{vac}}{\mu V_w h_{ug}})</td>
</tr>
</tbody>
</table>
3.3. EXAMPLES

Figure 3.4: Possible vortices inside slot coating and the flow parameters which can induce vortex birth. Three figures, (a), (b) and (c), are streamline plots of steady slot coating flows at different conditions (a) represents slot coating operation without vortex, and (b) and (c) represent upstream and downstream die lip region with vortices. Note that vortex inside upstream coating bead attached to saddle point, and vortex under downstream die lip is enclosed by a separating streamline, attached to the die lip.

Operating and geometric parameters and their values at a base case used in this study are summarized in Table 3.1.

The finite element mesh used to computed the results presented here is shown in Fig. 3.3(c). It consisted of 920 elements and 18300 degrees of freedom.

In slot coating operation, as shown in Fig. 3.1, recirculation usually occurs in two different regions of the flow; under the upstream die lip and under the downstream die lip. In the first case, the turn around flow with zero net flow rate may lead to a vortex formation. Under the downstream lip, recirculation attached to the wall may appear if the adverse pressure gradient is high enough.
3.3. EXAMPLES

Tracking cusp point under upstream die lip

In typical slot coating operation, vacuum pressure is applied at the upstream meniscus of a slot coater in order to move the meniscus away from the feed slot and to obtain thin coatings (Sartor, 1990). The net flow rate under the upstream die lip is zero. The liquid flows upstream because of the pressure difference and flow upstream dragged by the moving web. The length of the die lip that is wetted by the liquid is a function of the imposed pressure difference. The lower the upstream pressure, the longer is the upstream coating bead. If the upstream bead is short the flow turns around without forming a vortex. If the upstream bead is too long, a vortex is formed inside the flow. This transition between these two states implies a cusp point formation as sketched in Fig. 3.4.

In this study, we focus on two parameters which affect the length of the upstream coating bead and consequently the presence of a vortex: flow rate and vacuum pressure. High flow rates lead to long coating beads. We choose vacuum pressure $P_{\text{vac}}$ as the control variable for the continuation method, and flow rate $q$ as the auxiliary parameter for the augmented Navier–Stokes system. Augmented residuals are Eqs. (3.17), and the additional auxiliary parameters for the augment Navier–Stokes system are the two coordinates $x$ and $y$ of the cusp point and the flow rate $q$. The remaining flow parameters are the ones listed in Table 3.1.

The path of solution of the augmented Navier–Stokes system in the plane of dimensionless flow rate $q^* = q/(V_w h_{ug})$ and dimensionless vacuum pressure $P_{\text{vac}}^* = P_{\text{vac}}/(\mu V_w/h_{ug})$ is presented in Fig. 3.5. All the flow states over the constructed path present a cusp point, as illustrated in insert (a) of the figure, that marks the onset of a vortex birth. As discussed before, this curve is the boundary of the vortex-free window in this plane of parameters. For the purpose of illustration, we computed solutions at a set of parameters away from the constructed path. These flow states are illustrated in inserts (b) and (c) of the figure. Flow state (b) was obtained at a set of parameters above the computed path; the presence of a vortex is clearly observed. Flow state (c) was computed at a set of parameters below the curve; the turn around flow does not present a vortex.
3.3. EXAMPLES

Figure 3.5: Solution path of the augmented Navier–Stokes system. All the states on the path satisfy the vortex birth condition and, consequently present a cusp point, as indicated in (a), (b) and (c) are streamlines at a set of parameters above and below the computed path.
3.3. EXAMPLES

Vortex appears when the length of the upstream bead is long enough, i.e. at high flow rates and high vacuum (more negative) pressure.

With the discretization used here, e.g. 18300 degree of freedom, each solution of the Navier–Stokes system alone took approximately 60 s in a 1.7 GHz IBM power 4 system (about 6 iterations per solution, \( \sim 9.96 \text{s} \) per iteration). The solution of each point along the vortex birth path, i.e. the solution of the Navier–Stokes system augmented by the vortex birth condition: took approximately 220 s in the same machine. (about 20 iterations per solution, \( \sim 11.1 \text{s} \) per iteration). The line that defines the flow states at which vortex birth occurs was constructed by using 20 different values of vacuum pressure \( P_{\text{vac}} \), leading to a total computational time of approximately \( 20 \times 220 \text{s} = 4400 \text{s} \). Assuming that in order to find the critical flow rate at each value of vacuum pressure at which a vortex is born from a cusp point, 20 different solutions of the Navier–Stokes are needed, the total cost to predict the vortex free window by vortex capturing is \( 20 \times 20 \times 60 \text{s} = 24000 \text{s} \). The time required to post-process the solutions is not included in this estimation. The method described in this work is faster by a factor of \( \approx 5.5 \) even without considering the necessary and tedious user intervention associated with post-processing the computed flow fields. Furthermore, the accuracy of the critical flow states computed by tracking the vortex birth is much higher than what can be obtained by post-processing solutions at different set of flow parameters.

Tracking onset of separation point at solid wall

According to Sartor (1990), a converging downstream die lip geometry broaden the possible operating ranges of vacuum pressure and coating speed. The converging channel causes the formation of a pressure hill in this part of the flow. This high pressure helps to push the upstream meniscus away from the feed slot enabling thin coating at lower vacuum pressure. However, this adverse pressure gradient can create a flow detachment at the downstream die lip leading to a vortex formation, as illustrated in Fig. 3.4. The presence of vortex may limit the operating ranges when coating quality is important.

In this study, we focus on the effects of two parameters: flow rate \( q \) and downstream die lip angle \( \theta \), as shown in Figure 3.4. At a set die lip angle, a smaller flow rate
3.3. EXAMPLES

Figure 3.6: Solution path of the augmented Navier–Stokes system. The computed path marks the boundary of the vortex free window. The states on the path present a degenerate stagnation point at the wall, as indicated in (a), (b) and (c) are flow states above and below the line.

leads to a stronger adverse pressure gradient which can cause flow detachment. For direct tracking of vortex birth from the die lip, we choose the downstream die lip angle \( \theta \) as the control variable for continuation method, and flow rate \( q \) as the auxiliary parameter for augment Navier–Stokes system. Augmented residuals are the conditions for vortex birth at a solid wall, e.g. Eqs. (3.20), and the additional auxiliary parameter for augment Navier–Stokes system is one the coordinate \( x \) for the birth point along the downstream die lip and the flow rate. The remaining flow parameters are the ones listed in Table 3.1.

The path of solution of the augmented Navier–Stokes system in the plane of dimensionless flow rate \( q^* = q/(V_w h_{ug}) \) and die lip angle \( \theta \) is presented in Fig. 3.6. All the flow states over the solution path present a degenerate separation point, as shown in insert (a) of the figure. The results show that at a given flow rate there is a critical die lip angle above which a vortex appears. As illustration, flow fields at a set of con-
3.3. EXAMPLES

ditions above (insert b) and below (insert c) the solution path are also presented. The computed path clearly defines the boundary of the vortex-free region of the parameter space.

3.3.2 Half-submerged-forward roll coating

Forward roll coating is a process whereby liquid flows into a narrow gap between two counter rotating rolls before it splits into two layers, one on each of the roll surfaces [Coyle et al. (1986)], as sketched in Fig. 3.7. The flow near the film-splitting zone is two-dimensional as shown by [Coyle et al. (1986)], and a recirculation may appear depending on operating conditions. We consider the upstream side of the roll submerged into the pool of liquid, so that the flow system has only one meniscus. This prototype forward roll coating is so called half-submerged-forward roll coating (HSFR), as discussed by Carvalho (1996) and Coyle et al. (1986).

The complex flow pattern of film-splitting flows between rigid rolls has been extensively studied in the past. Coyle (1984) described the flow near the film splitting meniscus using lubrication theory and the full Navier–Stokes equation. He found that at low capillary number and large gaps, a recirculation attached to the meniscus could be observed. He also pointed out that when the symmetry of the flow is broken (the speed ratio is different than one), topological changes in the film-splitting zone is observed: the two vortices attached to the free surface split and one of them moves upstream and is not attached to the meniscus, as illustrated in Fig. 3.8 (c).

Benjamin (1994) named them internal gyre and free-surface gyre, and captured the flow state changes with respect to parameters using post-processing. Gaskell et al. (1998) analyzed the vortex structures in forward roll coating and their dependence on the operating parameters. The analysis was done by post-processing a series of steady state solutions at different operating parameters. Wilson et al. (2001) also examined the evolution of vortex-saddle patterns in both the free surfaces as changing operating parameters.

In this study, we consider a Stokes flow of a HSFR situation and focus on the vortex birth near the film-splitting zone. In order to represent the detail flow structure in this
3.3. EXAMPLES

3.2 Half-submerged-forward roll coating

\[ \mathbf{n} \cdot \mathbf{T} = \sigma \frac{dt}{ds} - \mathbf{n} P_{\text{amb}} \]

Kinematic

\[ \mathbf{n} \cdot \mathbf{u} = 0 \]

Capillary pressure

\[ \mathbf{n} \cdot \nabla \mathbf{u} = 0 \]

<table>
<thead>
<tr>
<th>Flooded</th>
<th>No-slip / No penetration</th>
<th>&quot;Film-splitting zone&quot;</th>
</tr>
</thead>
<tbody>
<tr>
<td>( p = p_{\text{in}} )</td>
<td>( 2H_0 )</td>
<td>Fully developed</td>
</tr>
</tbody>
</table>

(a) Sketch of flow domain and boundary conditions and geometry parameters used for film-splitting flows in a half-submerged-forward roll coating.

(b) Mesh used to discretize the free surface flow. 540 elements and 10808 unknown coefficients of the finite element basis function are used to discretize the system.

**Figure 3.7:** Half-submerged-forward roll coating: boundary condition and mesh.
### Table 3.2: Parameters used in half-submerged forward roll coating.

<table>
<thead>
<tr>
<th>Operating parameters</th>
<th>Unit</th>
<th>Base case</th>
</tr>
</thead>
<tbody>
<tr>
<td>Upper roll angular speed ($\Omega^u_{roll}$)</td>
<td>rad/sec</td>
<td>9.5</td>
</tr>
<tr>
<td>Lower roll angular speed ($\Omega^l_{roll}$)</td>
<td>rad/sec</td>
<td>9.5</td>
</tr>
<tr>
<td>Ambient pressure ($P_{amb}$)</td>
<td>Pa</td>
<td>0</td>
</tr>
<tr>
<td>Inflow boundary pressure ($P_{in}$)</td>
<td>Pa</td>
<td>0</td>
</tr>
<tr>
<td>Surface tension ($\sigma$)</td>
<td>dyne/cm</td>
<td>58</td>
</tr>
<tr>
<td>Viscosity ($\mu$)</td>
<td>mPa.s</td>
<td>2.3</td>
</tr>
<tr>
<td>Half gap height ($H_0$)</td>
<td>mm</td>
<td>1</td>
</tr>
<tr>
<td>Upper roll radius ($R^u$)</td>
<td>mm</td>
<td>100</td>
</tr>
<tr>
<td>Upper roll radius ($R^l$)</td>
<td>mm</td>
<td>100</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Dimensionless variables used in vortex-free window</th>
</tr>
</thead>
<tbody>
<tr>
<td>Name</td>
</tr>
<tr>
<td>Dimensionless gap height ($H_0^*$)</td>
</tr>
<tr>
<td>Capillary number ($Ca$)</td>
</tr>
<tr>
<td>Speed ratio ($S$)</td>
</tr>
</tbody>
</table>
3.3. EXAMPLES

Figure 3.8: Possible vortex structure in half-submerged forward roll coating and flow parameters which can induce vortex birth. Three figures, (a), (b) and (c), are streamline plots of forward roll coating at different conditions. “Stag. pt.” means stagnation point. (a) represent forward roll coating without vortex. When the system is symmetric, i.e. roll speed ratio between upper and lower roll is one, vortex structure is also symmetric as in (b). When the system is asymmetric, i.e. roll speed ratio is not unity, vortex structure loses symmetricity as in (c).

zone, the finite element mesh is designed so that elements are concentrated near the film splitting meniscus, as shown in Fig. 3.7(b). Boundary conditions are summarized in Fig. 3.7(a). Table 3.2 lists the operating parameters considered as the base case. The synthetic inflow and outflow boundaries were located such that the solution was virtually independent of their locations. The values used here were $L_u = 50\, mm$ and $L_d = 60\, mm$.

In this study, we apply the tracking algorithm in both the symmetric flow, e.g. equal roll radii and speed ratio equal to one, and flow with speed ratio different than one. Possible streamline patterns of both cases are presented in Fig. 3.8(b) and (c).
3.3. EXAMPLES

Tracking onset of degenerate stagnation point at free surface (symmetric flow)

In the symmetric flow there is always a stagnation point at the intersection of the free surface, and the symmetry plane, even in the cases where there is no recirculation. This stagnation point gives birth to two vortices simultaneously, as sketched in Fig. 3.8(b).

The presence of the recirculation attached to the free surface depends on the strength of the adverse pressure gradient that slows down the liquid as it approaches the meniscus and on the location of the free surface.

In this study, we focus on the effects of two parameters on the flow pattern: the half-gap height $H_0$ and the roll angular speed $\Omega_{roll}$. For direct tracking of vortex birth from the free surface, we choose the roll angular speed as control variable for continuation, and the half-gap height as the auxiliary parameter for the augment Navier–Stokes system. Augmented residuals are the conditions for vortex birth attached to a free surface, e.g. Eqs. (3.18), and the additional auxiliary parameter for augment Navier–Stokes system is the coordinate $y$ of the birth point along the free surface and the half-gap height $H_0$. The remaining flow parameters are listed in Table 3.2.

The path of solutions of the augmented Navier–Stokes system is presented in the plane of capillary number $Ca \equiv \mu \Omega_{roll}/\sigma$ and dimensionless gap $H_0/R$ in Fig 3.9. All the computed flow states present a degenerate stagnation point, as shown in insert(a). At each gap there is a critical capillary number below which two vortices attached to the free surface appears, as illustrated in insert (b). As in the previous examples, the computed solution path defines the boundary of the vortex free window.

Tracking cusp point and onset of degenerate stagnation point (asymmetric case)

When the flow symmetry is broken, either by roll radius ratio or roll speed ratio, the two symmetrical vortices are disconnected, as shown in the Fig. 3.8(c). Only one of the vortices stays connected to the saddle point inside flow. This recirculation is
3.3. EXAMPLES

Figure 3.9: Solution path of the augmented Navier–Stokes system. The computed path marks the boundary of the vortex free window. The states on the path present a degenerate stagnation point at the meniscus, as indicated in (a). (b) and (c) are flow states above and below the line.
3.3. EXAMPLES

Figure 3.10: Solution paths of the augmented Navier–Stokes systems for vortex birth from saddle point (squares) and from a free surface (circles). The line define the regions of the parameter space without vortex (insert 1), with a single & vortex inside the flow (insert 3) and with two vortices (insert 5).
3.3. EXAMPLES

detached from the free surface and it is located closer to the slower moving roll. The vortex that stays attached to the free surface is located near the fast moving roll. Gaskell et al. (1998) explored these complex vortex/saddle structure near the film-splitting zone as a function of the capillary number, roll radius ratio, and roll speed ratio. They show examples of evolution of the flow pattern by presenting “snapshots” of steady state using post-processing. However, they did not capture the very moment of vortex birth or death.

Two types of vortex birth occur in the asymmetric flow: the vortex attached to the upstream saddle point comes from a cusp point inside the flow and the vortex attached to the free surface comes from a degenerate stagnation point at the free surface. The conditions at which the vortices appear is not the same. We track the birth of both vortices as a function of the capillary number and speed ratio.

For direct tracking of both vortex births, we choose the speed ratio as the control variable for continuation, and the lower roll angular speed as the auxiliary parameter for the augment Navier–Stokes system. For tracking the vortex birth from the cusp point, the augmented residual are the conditions given by Eqs. (3.17), and the additional auxiliary parameters for the augment Navier–Stokes system are the two coordinates $x$ and $y$ of the cusp point and the lower roll speed. For the tracking the vortex birth from the free surface, the augmented residuals are conditions given by Eqs. (3.18), and the additional auxiliary parameter for the augment Navier–Stokes system is the coordinate $y$ of the degenerate stagnation point along the free surface and the lower roll speed.

Both solution paths are presented in Fig. 3.10. The curve on the right corresponds to the flow states with a cusp point inside the flow, it defines the set of conditions at which a vortex inside the flow is about to appear, as shown in insert (2). The curve on the left defines the conditions at which a degenerate stagnation point attached to the free surface can be observed, as illustrated in insert (4) of the figure. It defines the set of conditions at which the vortex attached to the meniscus appears. These two solution paths of the corresponding augmented Navier–Stokes system define the boundaries of the vortex-free window, single vortex region and two vortices window.

The evolution of the streamline pattern as the capillary number (lower roll speed) falls
3.4. **Final Remarks**

Vortices in flow can cause undesired effects in many industrial processes, especially for continuous liquid coating on moving substrate. It is important to know the region of the operating parameters space at which vortices are not present in the coating flow, so these set of parameters can be avoided during operation. These regions are called vortex-free operating window. In computer-aided analysis and design, the most effective way to construct these is not to check a large set of solutions *a posteriori*, but to delineate the range of design parameters and operating conditions that define the boundary of the vortex-free window. This means tracking the birth (of death) of vortices of three kinds: *free* in the flow, *attached* to solid wall or liquid free surface. A way of doing this is to solve the Navier–Stokes or related governing equations, after augmenting the system with one or more equations that describe the local kinematic conditions at vortex birth and with an equal number of design parameters or operating conditions as new unknowns. Two liquid coating processes — single-layer slot coating and forward roll coating — were chosen as examples of automatic generations of vortex-free windows. In the slot coating, vortex birth under the upstream die lip and under the downstream die lip were tracked to give guidelines of preferred range of flow rate, vacuum pressure and die lip angles. In the roll coating, we focused on the generation of recirculations near film splitting zone. For symmetric flow, we were able to construct the path of multiple vortices birth in the parameter space represented by gap and capillary number. For asymmetric flow with roll speed ratio different from one, the birth of two different vortices near the meniscus were tracked to delineate the boundaries of the vortex-free window, single vortex region
3.4. **FINAL REMARKS**

and two vortices window in flow topology map.
Chapter 4

Mid-gap invasion in two-layer slot coating

4.1 Introduction

Continuous liquid coating is the main manufacturing step in the production of many different products such as adhesive tapes, specialty papers, optical films and many others. It is also a very strong candidate for mass production of nanoparticle assembly films, as discussed by Maenosono et al. (2003a). Slot coating is one of the many existing methods to obtain a thin uniform liquid layer over a moving substrate. The coating liquid is pumped to a coating die in which the liquid is distributed across the width of a narrow slot. Exiting the slot, the liquid forms a bridge between the moving web and die lips that is bounded by an upstream and downstream gas-liquid interfaces. Slot coating belongs to a class of coating methods known as pre-metered coating: the thickness of the deposited liquid layer is set by the flow rate fed to the coating die and the web speed, and is independent of other process variables.

Several products require more than one layer for optimal performance. The most efficient way to manufacture these products is by coating all the layers at once before they are solidified. Slot coating process can be easily adapted for coating two different layers as shown as Fig. 4.1.
4.1. INTRODUCTION

Figure 4.1: Two-layer dual slot coating schematic diagram. The flow between a moving web and a die lip can be divided into three regions: upstream gap, mid gap and downstream gap regions. Interlayer starts from mid gap region and pass through downstream coating gap. The layer closes to the substrate is called bottom layer (phase 2 in the diagram). \( H_g \) is the gap height between web and die lip. \( h_{w,1} \) and \( h_{w,2} \) are the wet thickness of layer 1 (top layer) and layer 2 (bottom layer). \( L_u, L_m \) and \( L_d \) are the upstream lip, mid lip and downstream lip lengths, respectively. \( L_{f,1} \) and \( L_{f,2} \) are the width of feed slot for top and bottom liquids. Like other pre-metered coating, the flow rate for both layers is the product of the wet thickness and web speed: \( q_1 = h_{w,1} U_w \) and \( q_2 = h_{w,2} U_w \).
4.1. INTRODUCTION

The coating die has two separate feed slots through which each layer is fed. The flow can be divided into three regions separated by the feed slots: upstream gap, mid gap and downstream gap. When both liquids are immiscible, they form an interlayer inside the coating bead. But, in several coating applications, both layers use solvents that are miscible and the apparent interface is truly an inter-diffusion zone. However, because of the relative high speed of the moving web (typically about 1 m/sec) and the small length of die lip (\(\sim 1\) mm), the residence time in the coating bead is extremely small and so is the thickness of the diffusion zone [Musson 2001]: it is so thin that it can be considered as a distinct interlayer with zero interfacial tension.

The interlayer starts at the separation point (or line, in three-dimension), located somewhere in the mid die block. The point usually lodges on the downstream corner of the mid lip (Fig. 4.2(a)), but it can transit from one corner to the other, as observed by Cohen (1993), as operating condition changes. Thus the interlayer and the top liquid layer can invade the mid-gap region, accompanied by dangerous turn-around flow and sometimes the appearance of microvortex (Fig. 4.2(b)), as reported by Cohen (1993), Sartor et al. (1999), and Musson (2001).

This microvortex is deleterious, because it may create a mixing zone between both layers which will spoil the functionality of the final product. Wilson et al. (2006) discussed how fluid recirculating for a long time in coating flows causes coating defects and stirring. The microvortex also may become nodular along their length and spoil the transverse uniformity of the interlayer, which is also critical, especially in optical products. Moreover, as it will be shown in this study, when the separation

![Figure 4.2: Sketch of interlayer configuration before and after mid-gap invasion.](image-url)
4.2. MID-GAP INVASION: FLOW VISUALIZATION

line is not located at the downstream corner of the mid die lip, its location is very sensitive to process conditions, leading to downweb variation on the coated layer, also compromising the final product quality. The observed oscillation may indicate the presence of multiple solutions or that a steady flow state does not exist. Armi (1986) has shown that for two layer flows through contractions, there are two distinct solutions.

Therefore, as discussed by Sartor et al. (1999), the ideal location of the separation point that leads to the desired steady flow is the downstream corner of the mid die lip, as shown in Fig. 4.2(a). The movement of the separation point from its desired location along the mid die lip is usually referred to as mid-gap invasion and its occurrence can be associated to the onset of coating defects. It is crucial to determine the set of process conditions at which it happens.

Here, we analysed mid-gap invasion by flow visualization and by the solution of the two dimensional Navier–Stokes equation for free boundary flows. Through both approaches, we determined the critical operating conditions at the onset of mid-gap invasion and the different mechanisms behind it. We realized that an approximate and simple criterion that defines the occurrence of mid-gap invasion can be derived using lubrication approximation. The reported results define the operability limits of two-layer slot coating flows related to interlayer uniformity.

4.2 Mid-gap invasion: flow visualization

Since mid-gap invasion is related to the movement of the interlayer as process conditions change, we visualized its location inside a dual slot coating flow as a function of flow parameters and liquid properties in order to determine the critical conditions at which mid-gap invasion occurs.
4.2. MID-GAP INVASION: FLOW VISUALIZATION

The bench-scale experiment coater, specially designed to visualize coating flows used in these experiment is shown in Fig. 4.3.

The coating solutions were stored in two separated tanks which were connected to calibrated precision gear pumps (Parker-Nichols Zenith, West Newton, MA). We installed filters (model type 54/95, Balston corp., Haverhill, MA) between the tanks and the pumps to prevent any residual particle from entering into the pumps. Both flow rates were controlled by the pump speed and measured with a Coriolis flow meter (model MFC 100/MFS 3000, Krohne America Inc, Peabody, MA). Downstream of both pumps, the liquid flows through a debubbler (Whitey 12FK088 304L-HOF4-1000cc, Swagelok co., Solon, OH) in order to eliminate any small bubble inside the liquid and to dampen pulsations from the pumps. Both coating liquids were fed to two separate cavities of the 4” wide stainless steel coating die. Inside the cavities, liquid is distributed along the width of the die and flows through a narrow feed slot. The feed slot heights were set to 250 µm by shims. The die was mounted such that...
the gap between the die and the roll was easily adjusted. As indicated in Fig. 4.3, the downstream die lip formed a slightly converging channel with the roll surface, with an inclination angle of $\theta_l = 5^\circ$. Both liquids were coated as a two-layer flow onto a 8" diameter high quality optical glass roll (Professional Instruments, Hopkins, MN). The speed of the roll surface could be controlled from $7 \text{m/min}$ up to $20 \text{m/min}$. Because no substrate was used, the coated liquid, except a trace residue, was removed from the roll surface by a Teflon squeegee. The used coating liquids were solutions of water and glycerin. Two different concentrations of glycerin solution were used (73% and 80% by weight) so that the viscosity could be changed from $28.5 \text{cP}$ to $57.0 \text{cP}$. The viscosity of the Newtonian solution was measured with a Brookfield viscometer (YULA-15 ULA Spindle, Model DV-II+, Brookfield Eng. Lab., Stoughton, MA).

Since both coating liquids are transparent, the interlayer was marked by the dark blue dye (FD&C blue no.1 food grade dye, Fischer united supply co., Minneapolis, MN) which was mixed with top layer coating liquid and injected by a syringe pump (model 230, KD scientific, Holliston, MA) through a small port in mid die block shoulder.

A video camera (model NX18A, NEC, Tokyo, Japan) with microscope lens (Magna-zoom 6000 with 0.5×adapter, Navitar, Rochester, NY) was focused on a right-angle mirror mounted inside the open end of the roll in order to visualize the flow in the coating bead through the glass wall as in Fig. 4.3. The interlayer separation line position could be marked by the end of the dye streakline in this view as in sketched Fig. 4.4. If the streakline only appeared downstream of the mid die lip, the separation point was located at the desired location and mid-gap invasion did not occur. After the onset of mid-gap invasion, the blue die was clearly observed under the mid lip, as shown in Fig. 4.4. The circular and transverse motion of the streakline is a definite evidence of the presence of the undesired microvortex under the mid lip, as was also observed by Cohen (1993).

### 4.2.2 Onset of mid-gap invasion

The onset of mid-gap invasion was determined at different gaps $H_g = 210, 260$ and $360 \mu\text{m}$ and viscosity ratios $m = \mu_1/\mu_2 = 0.5, 1$ and 2, where $\mu_1$ and $\mu_2$ are the viscosity
Figure 4.4: Flow visualization of mid-gap invasion thorough the glass roll. Inter-layer is marked by a color dye. Interlayer movement and evidence of micro-vortex at the mid lip were captured by the location of dye streakline. Near the critical mid-gap invasion conditions, interlayer oscillates along mid-lip.
of the top and bottom layer, respectively. The goal was to find the values of coating thickness (flow rate) of both layers at which mid-gap invasion occurs. The critical conditions were found by the following procedure. First, the roll speed and flow rate of each layer were adjusted such that the separation line was located at the downstream corner of the mid lip, i.e. at these conditions mid-gap invasion was not observed. Then, one of the three operating parameters, top-layer flow rate, bottom-layer flow rate or roll speed, was slowly varied, keeping the others constant. After each step change, we waited at least one minute to reach a new steady state before recording the position of the separation point.

Mid-gap invasion was not observed while changing the top layer flow rate for all gaps and viscosity ratios explored. As the top-layer flow rate rises, the static contact line at the downstream free surface (point A in Fig. 4.4) moves along the downstream die shoulder and the separation point remains at the downstream corner of the mid lip. Mid-gap invasion was only observed when lowering the bottom-layer flow rate or raising the roll speed, from the initial set of parameters. Near the critical conditions for mid-gap invasion, the interlayer position oscillated. Even with all the operating parameters fixed, dye streaklines under the mid lip appeared and disappeared periodically. We suspect that at these conditions, the position of the separation line is extremely sensitive to flow parameters and the small oscillations on flow rate, due to the gear pump, and gap, due to roll run-out, is enough to cause the observed behavior. We considered the critical condition at the onset of mid-gap invasion the set of parameters at which the oscillation of the separation point could be observed.

The results show that the parameter that determines the mid-gap invasion is the bottom-layer wet thickness $h_{w,2}$, which can be varied by changing both the pump rotation or the roll speed. Figure 4.5 shows the critical bottom layer thickness in units of coating gap $h_{w,2}^* = h_{w,2}/H_g$ at which mid-gap invasion occurred as a function of the total thickness $h_{w,t}^* = h_{w,t}/H_g = (h_{w,1} + h_{w,2})/H_g$ at different gaps (Fig. 4.5(a)), and viscosity ratios (Fig. 4.5(b)). The curves at different gaps and viscosity ratios fall on top of each other, showing that these parameters do not affect the critical value of $h_{w,2}^*$. Moreover, the effect of the total thickness is extremely weak. At small values of the total wet thickness, e.g. $h_{w,t}/H_g \approx 0.3$, the critical dimensionless bottom-layer is approximately $h_{w,2}^* \approx 0.26$; at large total thickness, e.g. $h_{w,t}/H_g \approx 0.55$, the critical
4.2. MID-GAP INVASION: FLOW VISUALIZATION

Figure 4.5: Critical bottom-layer thickness at mid-gap invasion as a function of the total wet thickness, gap height (a) and viscosity ratio (b).
value is \( h_{\text{w,2}}^* \approx 0.34 \).

In summary, the experimental results show that the bottom layer thickness in units of gap is the key operating parameter that triggers mid-gap invasion. The critical condition is a function of the bottom-layer flow rate \( q_2 \), web speed \( U_w \), and coating gap \( H_g ( h_{\text{w,2}}^* = q_2 / (H_g U_w) ) \), and it is independent of liquid properties. The results also show that near the critical condition, the separation line oscillate along the mid lip. The mechanism that drives this phenomenon and the evolution of flow states were examined here by solving the Navier–Stokes equation.

### 4.3 Mid-gap invasion: Navier–Stokes theory

The flow in coating bead is described by the complete two-dimensional, steady-state mass and momentum conservation equations for free surface flows. Here, there are three free boundaries, the upstream and downstream gas-liquid interfaces and interlayer between the two liquids. The goal of the numerical analysis is to determine the interlayer configuration as a function of the flow parameters and consequently determine numerically the onset of mid-gap invasion. In order to allow interlayer movement along the die lip surface, the edges of the coating die were described as rounded corners with prescribed radius of curvature, which are usually set by limitations on the machining process (Romero et al., 2006). Here, we changed the bottom layer flow rate, keeping the top layer flow rate constant at different viscosity ratios.

#### 4.3.1 Governing equation and boundary conditions

The velocity and pressure fields of incompressible two-dimensional, steady state flow of a Newtonian liquid are governed by the continuity and momentum equations:

\[
\nabla \cdot \mathbf{u} = 0, \quad \rho_i \mathbf{u} \cdot \nabla \mathbf{u} = \nabla \cdot \mathbf{T}_i, \tag{4.1}
\]

where \( \rho_i \) is the liquid density and \( \mathbf{T}_i \) is the stress tensor. For Newtonian liquid, it is given by \( \mathbf{T}_i = -p \mathbf{I} + \mu_i \left[ \nabla \mathbf{u} + (\nabla \mathbf{u})^T \right] \), where \( \mathbf{I} \) is the Identity tensor, \( p \) is the pressure.
4.3. MID-GAP INVASION: NAVIER–STOKES THEORY

and $\mu_i$ is the liquid viscosity. Here, subscript $i$ defines the two liquid phases, $i = 1$ for top layer and 2 for bottom layer.

Boundary conditions are needed to solve the Navier–Stokes system. In a two-layer slot coating flow, the domain is bounded by inflow and outflow planes, solid walls and free surfaces (gas-liquid interfaces) and the surface that separates the two liquids, the interlayer, as shown in Fig. 4.7. To simplify the geometry, we considered the downstream die lip parallel to the moving web in the numerical analysis. We verified that this small change in the geometry of the downstream die lip does not affect the critical condition at the onset of mid-gap invasion by comparing the predictions obtained with both configurations.

At the inflow planes, we imposed

$$\mathbf{u} = U_b(x),$$  \hspace{1cm} (4.2)

where $U_b(x)$ is the imposed parabolic velocity profile. At the outflow plane we impose

$$\mathbf{n}_b \cdot \nabla \mathbf{u} = 0,$$

$$p = p_b,$$  \hspace{1cm} (4.3) (4.4)

where $p_b$ is the imposed outflow pressure (ambient pressure), and $\mathbf{n}_b$ is the unit normal vector to the boundary. The total wet thickness $h_{w,t} = h_{w,1} + h_{w,2}$ is determined by the flow rate of each layer and the web velocity $U_w$. The outflow plane was located a distance equal to $10H_g$ downstream of the die corner. Moving the outflow plane in the downstream direction did not affect the free surface configuration. At rigid solid walls, the no-slip and no-penetration boundary conditions were imposed. Along the web,

$$\mathbf{t}_w \cdot \mathbf{u} = U_w, \quad \mathbf{n}_w \cdot \mathbf{u} = 0,$$  \hspace{1cm} (4.5)

where $U_w$ is the solid wall velocity, $\mathbf{n}_w$ and $\mathbf{t}_w$ are the unit normal and tangent vectors to the wall. Along the die surface, $\mathbf{t}_w \cdot \mathbf{u} = 0$ and $\mathbf{n}_w \cdot \mathbf{u} = 0$. At the dynamic contact point, where the upstream meniscus meets the moving web, Navier slip condition is used instead of no slip condition,

$$\frac{1}{\beta} \mathbf{t}_w \cdot (\mathbf{u} - U_w \mathbf{t}_w) = \mathbf{t}_w \mathbf{n}_w : \mathbf{T},$$  \hspace{1cm} (4.6)
where $\beta$ is slip coefficient. Here we choose $\beta = 0.1 \text{ g}^{-1} \text{sec}^{-1}$, based on the numerical tests reported by Sartor (1990).

Along the gas-liquid interfaces, a force balance and the no-penetration condition (kinematic condition) were imposed:

\begin{align*}
\mathbf{n}_f \cdot \mathbf{T}_i &= \frac{d\mathbf{t}_f}{ds} - \mathbf{n}_f P_{amb}, \\
\mathbf{n}_f \cdot \mathbf{u} &= 0,
\end{align*}

where $\mathbf{t}_f$ and $\mathbf{n}_f$ are the local unit tangent and normal vectors to the free surface, $s$ is the arc-length coordinate along the interface, $\sigma$ is the liquid surface tension and $P_{amb}$ is the ambient pressure.

Like the gas-liquid interfaces, the position of the interlayer is unknown a priori. In order to track its location, three conditions — force balance, no-penetration and velocity continuity — are to be satisfied along the interlayer:

\begin{align*}
\mathbf{n}_I \cdot \mathbf{T}_1 &= \mathbf{n}_I \cdot \mathbf{T}_2 + \sigma_I \frac{d\mathbf{t}_I}{ds}, \\
\mathbf{n}_I \cdot \mathbf{u}_1 &= \mathbf{n}_I \cdot \mathbf{u}_2 = 0, \\
\mathbf{t}_I \cdot \mathbf{u}_1 &= \mathbf{t}_I \cdot \mathbf{u}_2,
\end{align*}

where $\mathbf{n}_I$ and $\mathbf{t}_I$ are the unit normal and tangent vectors to the interlayer. $\mathbf{u}_i$ and $\mathbf{T}_i$ are velocity and stress tensor for phase $i$, and $\sigma_I$ is the interfacial tension.

The singularity of the stress field at a static interlayer separation point is integrable, e.g. the total shear force on the wall is finite (Silliman and Scriven 1980). Therefore, there is no need to relief the stress singularity and the no-slip condition is acceptable. An extra condition is needed to set the location of the separation point.

The interlayer separation point is nothing more than a regular separation point on a solid wall, except that liquids are different in either sides of the interlayer, which is just a “special” dividing or separating streamline. Because of the flow reversal at the separation point, the profile of the tangent velocity presents an inflection point, as sketches in Fig. 4.6 (Schlichting 1968):

\begin{equation}
\frac{\partial u_i}{\partial s_n} = 0,
\end{equation}
4.3. MID-GAP INVASION: NAVIER–STOKES THEORY

Figure 4.6: Flow near interlayer separation point at the solid wall. The local velocity profiles near the wall at different locations around the separation point are shown in the dotted boxes. The inflection point of the velocity profile is the indication of zero shear rate at the interlayer separation point.
where $u_t$ is the tangential velocity component and $s_n$ is the arc-length coordinate normal to the wall.

Because the velocity along the wall is zero, $\partial u_t / \partial s_t = 0$ ($s_t$ is the arc-length coordinate along the wall). Therefore, the normal deformation rates at the separation point vanish. For Newtonian liquids, vanishing deformation rate is equivalent to vanishing viscous stress. The force balance condition, Eq. (4.9), at the separation point reduces to the well-known Young-Laplace equation:

$$\begin{align*}
p_1 = p_2 + \sigma_1 \left( n_1 \cdot \frac{dt_i}{ds} \right) = p_2 + \sigma_1 (2H),
\end{align*}$$

where $H$ is the mean curvature of the interface at the separation point. Furthermore, when the interfacial tension is negligible, one may find that this equation boils down to the equal pressure condition:

$$\begin{align*}
p_1 = p_2
\end{align*}$$

This is the condition used by Scanlan (1990) and Musson (2001) to track the interlayer separation point with zero interfacial tension.

Here we use the more general zero shear rate condition as a boundary condition for the mesh equations in order to track the interlayer separation point along the mid lip. All boundary conditions are summarized in Fig. 4.7.

Flows with free surfaces and interlayer give rise to a free boundary problem. The flow domain is unknown a priori and it is part of the solution.

To solve a free boundary problem by means of standard techniques for boundary value problems, the set of differential equations and boundary conditions posed in the unknown physical domain have to be transformed to an equivalent set defined in a known, fixed computational domain. This transformation is made by a mapping $x = x(\xi)$ that connects the two domains. The physical domain is parameterized by the position vector $x = (x, y)$, and the reference domain, by $\xi = (\xi, \eta)$. The mapping used here is the one described by de Santos (1991). The inverse mapping is governed by a system of elliptic differential equations identical to those encountered in the dilute regime of diffusional transport.

$$\begin{align*}
\nabla \cdot D_\xi(\xi, \eta) \nabla \xi &= 0, \nabla \cdot D_\eta(\xi, \eta) \nabla \eta = 0.
\end{align*}$$
4.3. MID-GAP INVASION: NAVIER–STOKES THEORY

Boundary conditions

\[ \partial u / \partial s \cdot n + \mathbf{t}_n = 0 \]

Zero shear rate condition

\[ u = U_w t_w \]

No-slip condition

Dynamic contact angle

\[ \beta_w = \cos \theta_w \]

Navier slip condition

\[ (u - U_w t_w) \cdot t_w = \mathbf{t}_n : \mathbf{T} \]

Interlayer force balance

\[ n_1 \cdot \mathbf{T} = \sigma d t/ds - n P_{amb} \]

Capillary pressure

\[ n \cdot \mathbf{n}_w = \cos \theta_s \]

No penetration

\[ n \cdot \mathbf{u} = 0 \]

Static contact angle

\[ n \cdot \mathbf{n}_w = \cos \theta_s \]

No-slip condition

\[ u = 0 \]

Impose Poiseuille flow

\[ u = f(x, h_{w,1}, h_{w,2}, U_w) \]

Impose Poiseuille flow

\[ u = f(x) \]

Imposed surface tension

\[ n \cdot \mathbf{T} = \sigma d t/ds - n P_{mb} \]

Figure 4.7: Boundary conditions used for two-layer dual slot coating flow model. Free surface and interlayer configuration are unknown and part of the solution.
4.3. MID-GAP INVASION: NAVIER–STOKES THEORY

$D_\xi$ and $D_\eta$ are mesh diffusivities which control the steepness of gradients in the node-spacing by adjusting the potentials $\xi$ and $\eta$. Curves of constant $\xi$ and $\eta$ define the boundaries of elements used to describe the domain. The cross point of these curves sets the position of a node. Boundary conditions are needed to solve the second-order differential equations, Eqs. (4.15). Solid walls and inflow and outflow planes are described by the function that defines their geometry and nodes were distributed along them by a specified stretching function. The location of the free surfaces and interlayer are implicitly determined by the corresponding kinematic conditions Eqs. (4.8) and (4.10). The discrete version of the mapping equations is generally referred to as mesh generation equations. Detailed procedure and boundary conditions for mesh equation are discussed in de Santos (1991).

4.3.2 Solution of the Navier–Stokes system for free surface flow by G/FEM

Galerkin finite element method is used to solve the Navier–Stokes system, Eq. (4.1), coupled with the mesh generation equation, Eq. (4.15). Each independent variable, velocity, pressure and nodal position, is approximated by a linear combination of a finite number of basis functions, which are the unknowns of the discretized problem. The velocity and nodal position are represented in terms of Lagrangian bi-quadratic function $\phi_i(\xi, \eta)$, and the pressure in terms of linear discontinuous basis function $\psi_k(\xi, \eta)$.

This particular choice of basis functions have two advantages in dealing with the interlayer. The choice of the bi-quadratic function for velocity requires sharing velocity nodal point between both layers along the interlayer. Therefore velocity continuity is redundant and only the no-penetration along the interlayer needs to be imposed. The other merit comes from the discontinuous pressure field. Because pressure jump can occur across the interlayer (Mavridis et al., 1987), even in the absence of interfacial tension. The linear discontinuous basis function can handle the jump naturally without any modification, as needed to be done by Mavridis et al. (1987) and Scanlan (1990).
The weak form of Eqs. (4.1) and (4.15) are obtained by multiplying each equation by weighting functions, integrating over the physical domain, and applying the divergence theorem to the appropriate terms. Essential boundary conditions were imposed by replacing the corresponding weighted residual equation with the desired velocity or node specification. Natural boundary conditions were applied through the boundary integrals that come from the divergence theorem.

In sum, the Galerkin finite element method reduces the Navier–Stokes and mesh generation differential equations to a set of nonlinear algebraic equations on the basis functions coefficients.

\[ \mathbf{R}(\mathbf{z}, \lambda) = \mathbf{0}, \tag{4.16} \]

where \( \mathbf{z} \) is the solution vector which consist of the finite element coefficients for velocity \( \mathbf{u} \), pressure \( P \) and position \( \mathbf{x} \), and \( \lambda \) is a vector that contains the parameters on which the system depends. Eq. (4.16) is solved iteratively by Newton’s method:

\[ \mathbf{J}^{(i)}(\mathbf{z}^{(i)}, \lambda) \delta \mathbf{z}^{(i)} = -\mathbf{R}^{(i)}(\mathbf{z}^{(i)}, \lambda), \tag{4.17} \]

\[ \mathbf{z}^{(i+1)} = \mathbf{z}^{(i)} + \delta \mathbf{z}^{(i)}, \]

the indices \( i \) and \( i+1 \) indicate the current and next Newton’s step. \( \mathbf{J}^{(i)} \equiv \partial \mathbf{R}^{(i)}/\partial \mathbf{z}^{(i)} \) is the Jacobian matrix. The iteration continues until \( \| \mathbf{R}^{(i)} \|_2 < \epsilon \). Here we choose \( \epsilon = 10^{-8} \).

### 4.3.3 Designing a mesh to track the interlayer separation point

In order to study numerically the mid-gap invasion phenomenon with the approach described here and to determine the flow parameters at which the interlayer separation point moves along the mid die lip, special care is required on the design of the free boundary conforming mesh used to discretize the domain. Previous works on two-layer slot coating that used similar approach did not focus on mid-gap invasion phenomena (Scanlan, 1990; Musson, 2001). Taylor and Hrymak (1999) determined the interface zone by using a fixed mesh with convection-diffusion equation for imaginary tracer species, with arbitrarily chosen tracer diffusivity, not requiring special care with the mesh near the separation.
Because the model needs to capture the movement of the separation point, the die edges cannot be described as a mathematical corner, which would lead to an artificial pinning of the interlayer separation point. We describe the lip geometry as a combination of lines and arc of circles. The radius of curvature of each die corner was set to $50 \mu m$. In order to describe the lip geometry accurately, the mesh needs to be concentrated along the sections of the curve that corresponds to the arc of circle, i.e. the die corner. One possibility is to distribute the modes along the die surface using a weighting function \cite{Thompson1985} based on the curvature of the domain boundary:

$$w(s) = 1 + \alpha |\kappa(s)|,$$

(4.18)

where $s$ is arc-length coordinate along the boundary, $\kappa(s)$ is curvature, and $\alpha$ is user-defined coefficient to adjust the concentration of the nodes.

The use of this function was not effective, since the curvature is discontinuous at the point where the straight line and arc of circle meet, leading to large gradients on the elements near that point.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{mesh_topology.png}
\caption{Two mesh topologies for the mid-gap invasion. Mesh topology I provides less distorted mesh when the interlayer separation point is close to downstream corner of the mid lip, i.e. before mid-gap invasion flow state. Mesh topology II is suitable when the point is near upstream corner of the mid lip, i.e. after mid-gap invasion flow state.}
\end{figure}
4.3. MID-GAP INVASION: NAVIER–STOKES THEORY

To prevent the discontinuity on the weighting function \( w(s) \), which may cause discretization error, a modified function \( w_s(s) \), with continuous derivative, was used instead. It was constructed by solving the time evolution of a diffusion-like process using \( w(s) \) as the initial condition.

\[
\begin{align*}
\frac{\partial w^*}{\partial t} &= \frac{\partial^2 w^*}{\partial s^2}, \\
w^*(t = 0, s) &= w(s)
\end{align*}
\]  
(4.19)

The weighting function used to distribute the nodes along the die wall \( w_s(s) \) was the solution of (4.19) at an arbitrary time \( t = t^* \):

\[
w_s(s) = w^*(t^*, s).
\]  
(4.20)

The degree of smoothness increases with \( t^* \). Here, we chose \( t^* = 0.01 \). The weighting functions constructed this way are compatible to the hyperbolic tangent stretching functions used by Vinokur (1983a) and they were adequate to construct finite element meshes that allowed an accurate representation of the die surface, including the rounded corners. See Appendices A and B and for details about mesh constructions for the model.

Because of the large mesh deformation that occurs as the separation point moves upstream along the die lip, we used two different mesh topologies, one designed for flow states before the mid-gap invasion occurred (I), and the other designed for flow states after the mid-gap invasion (II). Figure 4.8 illustrates both mesh topologies. Mesh I had 1392 elements and the resulting non-linear system, 27508 degrees of freedom, whereas Mesh II had 1068 elements and 21128 degrees of freedom. For each mesh configuration, solutions with different discretizations were also obtained at \( \mu_1/\mu_2 = 1 \). The solution paths obtained in the range tested did not change as the number of degree of freedom was raised to 30128 in mesh I and 23864 in mesh II.
### 4.3. MID-GAP INVASION: NAVIER–STOKES THEORY

Table 4.1: Parameters used in two-layer dual slot coating.

<table>
<thead>
<tr>
<th>Operating parameters</th>
<th>Unit</th>
<th>Base case</th>
</tr>
</thead>
<tbody>
<tr>
<td>Top-layer film thickness($h_{w,1}$)</td>
<td>mm</td>
<td>0.0625</td>
</tr>
<tr>
<td>Web speed($U_w$)</td>
<td>m/sec</td>
<td>1</td>
</tr>
<tr>
<td>Ambient pressure ($P_{amb}$)</td>
<td>Pa</td>
<td>0</td>
</tr>
<tr>
<td>Surface tension ($\sigma$)</td>
<td>dyne/cm</td>
<td>61</td>
</tr>
<tr>
<td>Interfacial tension ($\sigma_I$)</td>
<td>dyne/cm</td>
<td>0 or 1</td>
</tr>
<tr>
<td>Density($\rho_1 = \rho_2$)</td>
<td>g/cm$^3$</td>
<td>1.2</td>
</tr>
<tr>
<td>Viscosity($\mu_1$ or $\mu_2$)</td>
<td>cP</td>
<td>23 or 46 or 115</td>
</tr>
<tr>
<td>Dynamic contact angle($\theta_d$)</td>
<td>deg.</td>
<td>128</td>
</tr>
<tr>
<td>Static contact angle($\theta_s$)</td>
<td>deg.</td>
<td>62</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Geometry parameters (See Fig. 4.1)</th>
<th>Unit</th>
<th>Base case</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gap height($H_g$)</td>
<td>mm</td>
<td>0.250</td>
</tr>
<tr>
<td>Upstream lip length($L_u$)</td>
<td>mm</td>
<td>1.75</td>
</tr>
<tr>
<td>Mid lip length($L_u$)</td>
<td>mm</td>
<td>0.500</td>
</tr>
<tr>
<td>Downstream lip length($L_d$)</td>
<td>mm</td>
<td>1.25</td>
</tr>
<tr>
<td>Top-layer feed slot width($L_{f,1}$)</td>
<td>mm</td>
<td>0.250</td>
</tr>
<tr>
<td>Bottom-layer feed slot width($L_{f,2}$)</td>
<td>mm</td>
<td>0.250</td>
</tr>
</tbody>
</table>
4.3. MID-GAP INVASION: NAVIER–STOKES THEORY

4.3.4 Results from numerical model: two mechanisms behind mid-gap invasion

We present the evolution of the flow states as the bottom-layer thickness varies at different viscosity ratios. All the other operating and geometric parameters were kept fixed, their values are presented in Table 4.1. Experiments have shown that mid-gap invasion occurs at \( h_{w,2}^* = h_{w,2}/H_g \approx 0.3 \), so we chose two different solution paths for each viscosity ratio \( m = \mu_1/\mu_2 \) explored:

1. with mesh I, start solution path at \( h_{w,2}^* = 0.4 \). Use arc-length continuation by decreasing \( h_{w,2}^* \).

2. with mesh II, start solution path at \( h_{w,2}^* \lesssim 0.2 \) (the actual value is a function of the viscosity ratio). Use arc-length continuation by raising \( h_{w,2}^* \).

It is important to note that because the separation point location is extremely sensitive to bottom-layer flow rate, the solutions presented here could not be computed by zeroth or first-order continuation. Arc-length continuation (Bolstad and Keller, 1986) proved to be essential.

The location of the interlayer separation point at zero interfacial tension (\( \sigma_I = 0 \)) was recorded at different dimensionless bottom-layer flow rates \( h_{w,2}^* \) at viscosity ratios \( m = 0.2, 0.5, 1, 2, \) and \( 5 \). The results are presented in Figs. 4.9 and 4.10. The position of the separation point is parameterized by an arc-length coordinate system defined along the mid die piece. On this system, the upstream corner of the mid lip is at \( 1.50 < s < 1.58 \), and the downstream corner is at \( 2.08 < s < 2.14 \).

Figure 4.9 shows the results at viscosity ratio \( m = 1 \) (\( \mu_1 = \mu_2 = 23 \text{cP} \)). The top branch corresponds to solutions computed with mesh I. The separation point stays at the downstream corner of the die lip for \( h_{w,2}^* \gtrsim 0.33 \). Below this value, its location becomes extremely sensitive to \( h_{w,2}^* \). The solution path ends at a termination point beyond which solutions could not be computed. The bottom branch on the figure corresponds to solutions obtained with mesh II. At low bottom layer thickness, e.g. \( h_{w,2}^* < 0.3 \), the separation point is located at the upstream die corner. At \( h_{w,2}^* \approx 0.3 \), it
4.3. MID-GAP INVASION: NAVIER–STOKES THEORY

Figure 4.9: Interlayer separation point locations as a function of the dimensionless bottom-layer wet thickness for viscosity ratio $m = \mu_1/\mu_2 = 1$. For measurement of the interlayer separation point location, arc-length along the mid-die block measured from the outflow boundary of bottom-layer (layer 2) inflow boundary. Note that shaded area near $h_{w,2}^* = 0.31$ has both solution branch coexist which implies possible multiple solutions for given flow rate.
4.3. MID-GAP INVASION: NAVIER–STOKES THEORY

**Figure 4.10:** Interlayer separation point location as a function of the dimensionless bottom-layer wet thickness at viscosity ratio $m = 0.5$, $0.2$, $2$, and $5$. For high viscosity ratio, $\mu_2 = 23\text{cP}$ and $\mu_1$ is either $46\text{cP}$ or $115\text{cP}$. For low viscosity ratio, $\mu_1 = 23\text{cP}$ and $\mu_2$ is either $46\text{cP}$ or $115\text{cP}$. 
moves downstream as the bottom-layer thickness rise. Again, the solution path ends at a termination point beyond which solutions could not be obtained. Both solution branches did not connect, but they indicate the existence of multiple solutions at \( h_{w,2}^* \approx 0.31 \). The high sensitivity of the separation point location to the flow rate and moreover the occurrence of multiple solutions may explain the oscillation observed in the experiments near the conditions at which mid-gap invasion occurs.

The solution paths at different viscosity ratios are presented in Fig. 4.10. Clearly, the evolution of the interlayer separation point location as the bottom-layer dimensionless flow rate \( h_{w,2}^* \) changes is different at low and high viscosity ratio. At high viscosity ratios (\( m = 5 \) and \( m = 2 \)), solutions with the separation point located along the mid die lip could not be computed; the solution paths terminate at flow states at which the separation point is at the upstream or downstream corner of the mid lip. At low viscosity ratios (\( m = 0.5 \) and \( m = 0.2 \)), flow states with the separation point at the mid lip could be computed. The curves resemble a typical hysteresis loop. At all viscosity ratios explored, there is a range of bottom-layer thickness at which multiple solutions existed. This range makes the onset of mid-gap invasion. Although the variation is small, the computed critical flow rate falls from \( h_{w,2}^* \approx 0.34 \) to \( h_{w,2}^* \approx 0.2 \) as the viscosity ratio rises from \( m = 0.2 \) to \( m = 5 \). The same behaviour was observed in the experiments shown in Fig. 4.5(b).

Snapshots of streamline plots at different dimensionless bottom-layer (layer 2) wet thickness \( h_{w,2}^* \) along the solution branches at viscosity ratio \( m = 0.2 \) are shown in Fig. 4.11. At this low viscosity ratio, solutions with the interlayer separation point from the downstream corner to the upstream corner of the mid lip could be computed with mesh topology I. At \( h_{w,2}^* = 0.4 \), the separation point is located at the downstream corner of the mid lip; the gap under the mid lip is filled only with liquid phase 2. As the bottom-layer thickness falls, the separation point moves upstream along the mid lip surface, its position is very sensitive to the \( h_{w,2}^* \) in the range of \( 0.33 \lesssim h_{w,2}^* \lesssim 0.4 \). The flow states at \( h_{w,2}^* = 0.27 \) computed with both meshes are virtually the same. The bottom flow branch is constructed by raising the flow with mesh topology II. In this case, the separation point departs from the corner only at \( h_{w,2}^* > 0.35 \) and it is clear that there are two different solutions at \( h_{w,2}^* = 0.34 \). The flow branches show that once a separation point is located on a rounded corner with a small radius of
4.3. MID-GAP INVASION: NAVIER–STOKES THEORY

Figure 4.11: Interlayer separation point location as a function of the dimensionless bottom-layer wet thickness at viscosity ratio \( m = 0.2 \). Shaded area inside the plot indicates possible multiple solutions. Note that both mesh topologies I and II show the same streamline plots at \( h_{w,2}^* = 0.27 \).
curvature, it tends to stay there, leading to a hysteresis loop.

Figure 4.12 presents different flow states along the flow branches at high viscosity ratio, $m = 5$. The solution branch from mesh topology I shows that the interlayer separation point does not move as $h^*_{w,2}$ falls. It is still located at the downstream corner even at $h^*_{w,2} \approx 0.2$. Because of the low bottom layer flow rate, a microvortex attached to the die surface appears around $h^*_{w,2} = 0.33$. Its size grows rapidly as $h^*_{w,2}$ falls. The gap under the mid lip is filled with liquid phase 2 only.

The solution branch obtained with mesh topology II cannot go beyond $h^*_{w,2} = 0.215$ as $h^*_{w,2}$ rises. The cause of failure comes from the mesh distortion in the middle of interlayer near the downstream corner: the interlayer configuration approaches a cusp. There is clearly a region of multiple solution around $h^*_{w,2} = 0.2$. The different flow patterns at $h^*_{w,2} = 0.202$ are shown in the inserts of Fig. 4.12. Both flows show a large recirculation under the mid lip. The difference is that the separation point is located at different extremes of the lip. In the top branch, the vortex structure is filled with the bottom layer liquid. In the bottom branch, the top layer has invaded the mid gap and the recirculation is filled with liquid phase 1 (top layer).

Along the top branch, the recirculation of liquid phase 2 under the mid lip grows as the bottom layer flow rate falls to a point at which the separation bubble almost touches the interlayer, inside the dashed rounded box in the plot. Along the bottom branch, the recirculation of liquid phase 1 shrinks as the bottom layer flow rate rises up to a point at which the interlayer almost touches the saddle point of the streamline, located inside the dashed rounded box.

Since the model does not allow flow though the interlayer, we cannot capture the mixing between two layers. However, these two different flow states at the same set of parameters strongly suggest that a mixing zone will be created after the streamline that defines the vortex touches the interlayer or the saddle point. The evolution of the flow states resembles the birth of saddle point inside a flow which is accompanied by the formation of cusp point on the separating streamline. The interlayer separation point jumps to the upstream corner of the mid lip. Therefore the oscillation of the interlayer observed during the experiments can be explained by the oscillation between the two flow states with the formation of a mixing zone.
4.3. **MID-GAP INVASION: NAVIER–STOKES THEORY**

**Figure 4.12:** Plot of interlayer separation point with respect to the dimensionless bottom-layer wet thickness for viscosity ratio $m = 5$. Note that there are two different flow states at $h_{w,2}^* = 0.202$. For the mesh topology I solution, dashed rounded box shows the area where the interlayer touches the micro vortex. For the mesh topology II solution, dashed rounded box shows the area where saddle point of streamline is located.
4.4. MID-GAP INVASION: A SIMPLE MODEL AND 1/3 RULE

The results show that the evolution of flow states as the bottom-layer flow rate falls and mid-gap invasion occurs at low viscosity ratio is different from that at high viscosity ratio. This difference is summarized in Fig. 4.13. When the top layer is less viscous than the bottom layer (low viscosity ratio), the separation point travels along the mid lip surface. When the top layer is more viscous than the bottom layer, first a recirculation of the bottom layer appears, it grows until it touches the interlayer. The separation point “jumps” from the downstream to the upstream die corner and mixing zone is formed.

Solutions at different density ratios, \( r = \frac{\rho_1}{\rho_2} = 2 \) or 0.5 and non-zero interfacial tension \( \sigma = 1 \text{ dyne/cm} \) where also obtained but there is no significant change on the interlayer separation location and on the evolution of the flow states.

In sum, the predictions obtained with the finite element method explain the observed oscillation of the flow near the critical conditions for the onset of mid-gap invasion. The predicted critical bottom-layer thickness agrees well with the experiments, it is in the same range of the observations, e.g. \( 0.2 \lessapprox h_w^* = \frac{h_{w,2}}{H_g} \lessapprox 0.35 \), and it falls weakly as the viscosity ratio rises. Moreover, the predictions reveal that the evolution of the flow states depends strongly on the viscosity ratio of the two liquid phases.

4.4 Mid-gap invasion: a simple model and 1/3 rule

The evolution of the flow states as mid-gap invasion occurs at low and high viscosity ratios are shown in Fig. 4.13. Even though mid-gap length is not much larger than the gap, e.g. \( \frac{L_m}{H_g} = 2 \), the streamlines strongly suggest that the flow under the mid lip can be effectively represented as a rectilinear flow for both invasion mechanisms. It is also important to notice that in both cases, mid-gap invasion is accompanied by flow reversal. In the low viscosity ratio regime, the flow reversal occurs in liquid phase 1 (top layer); in the high viscosity ratio regime, it occurs first in liquid phase 2 (bottom layer).
4.4. MID-GAP INVASION: A SIMPLE MODEL AND 1/3 RULE

![Diagram showing two mechanisms of mid-gap invasion]

**Figure 4.13:** Two mechanisms of mid-gap invasion. In these plots, low viscosity ratio corresponds to \( m = 0.2 \) and high viscosity ratio to \( m = 5 \). \( h_{w,2}^{*} \) for streamline plots (a), (b), and (c) are 0.40, 0.32, and 0.27, respectively. \( h_{w,2}^{*} \) for (d), (e), and (f) are 0.40, 0.25, and 0.18, respectively.

#### 4.4.1 Simple flow representation near mid lip

Since the flow is almost rectilinear, the velocity profile is approximately a superposition of Couette (drag by the moving substrate) and Poiseuille (pressure gradient along the bead) flows — quadratic velocity profile.

Figure 4.14 shows sketches of parabolic velocity profiles in the mid-gap region at conditions before and after mid-gap invasion. Before mid-gap invasion, flow state (a) of Fig. 4.14, the mid-gap region is filled with bottom-layer (layer 2) coating liquid. The adverse pressure gradient is not strong enough to cause flow reversal. As the bottom layer flow rate falls, the adverse pressure gradient becomes stronger and flow reversal occurs, as shown in Fig. 4.14(c) — low viscosity ratio regime — and Fig. 4.14(d) — high viscosity ratio regime.

The evolution of the velocity profile from (a) to (c) or (d) in Fig. 4.14 as bottom layer (layer 2) flow rate decreases, must pass through flow state (b) which shows an inflection point of the velocity profile at the die lip. This event is the *onset of flow reversal* and it can be related to the mid-gap invasion.
Figure 4.14: Evolution of the rectilinear velocity profile in the mid-gap region as mid-gap invasion occurs. The velocity profile at the mid-gap invasion at low and high viscosity are shown in (c) and (d), respectively. The transition occurs when an inflection point in the velocity profile is located at the die surface (b).
4.4. MID-GAP INVASION: A SIMPLE MODEL AND 1/3 RULE

4.4.2 1/3 Rule, a simple criterion for onset of mid-gap invasion

In order to derive a simple criterion to determine the critical conditions at the onset of mid-gap invasion, we will derive a simple criterion for the onset of flow reversal under the mid lip.

In rectilinear flow, the bottom-layer (layer 2) velocity profile is a combination of Couette and Poiseuille contributions and it is a function of the pressure gradient across the mid-lip $\Delta P/L_m$ and the web speed $U_w$:

$$u(y) = \frac{1}{2\mu_2} \left( \frac{\Delta P}{L_m} \right) (y^2 - H_g y) + \left( 1 - \frac{y}{H_g} \right) U_w,$$

(4.21)

where subscript 2 stands for bottom-layer liquid, $H_g$ is gap height, and $y$ is the coordinate value defined from the moving substrate ($y = 0$) to the die lip ($y = H_g$).

At the onset of flow reversal, the velocity at the die lip has an inflection point, $\left( \frac{du(y)}{dy} \right)_{y=H_g} = 0$. One can extract the critical pressure gradient value at which this occurs:

$$\left( \frac{\Delta P}{L_m} \right)_c = \frac{2\mu_2 U_w}{H_g^2},$$

(4.22)

where subscript $c$ means the value is related to the onset of flow reversal.

The bottom-layer wet thickness can be related to pressure gradient through the bottom-layer flow rate, because the flow rate is the product of wet thickness and substrate speed in slot coating method:

$$q_2 = \int_0^{H_g} u(y) \, dy = -\frac{1}{12\mu_2} \left( \frac{\Delta P}{L_m} \right) H_g^3 + \frac{U_w}{2} H_g = h_{w,2} U_w,$$

(4.23)

where $q_2$ is the bottom-layer flow rate, and $h_{w,2}$ is the bottom-layer wet thickness, as shown in Fig. 4.1.

At the onset of flow reversal, the critical dimensionless bottom-layer wet thickness, or flow rate, can be obtained by combining (4.22) and (4.23):

$$\left( \frac{h_{w,2}^*}{H_g} \right)_c = \left( \frac{h_{w,2}^*}{H_g} \right)_c = -\frac{H_g^2}{12\mu_2 U_w} \left( \frac{\Delta P}{L_m} \right)_c + \frac{1}{2} = \frac{1}{3}. $$

(4.24)
4.5. CONCLUSIONS

This very simple argument shows that flow reversal occurs at \( h_{w,2}^* = 1/3 \), i.e. wet thickness of the bottom layer is 1/3 of the gap. As we have discussed before, flow reversal under the mid lip and mid-gap invasions are related phenomenon. The results from flow visualization and Navier-Stokes theory support this.

Even though the simple criterion cannot predict oscillations of interlayer and the detailed mechanisms of mid-gap invasion and it is based in strong simplifying assumptions, it provides a simple approximate coating window to avoid mid-gap invasion.

4.5 Conclusions

The stability of the separation point of the interlayer in two-layer slot coating is directly associated with the quality of the final product. The location of the interlayer separation point along the mid-die lip as the operating conditions change was examined by both flow visualization and Navier–Stokes theory. From flow visualization, we found that the location of the separation point is a strong function of the bottom-layer flow rate and almost insensitive to the other operating parameters. The mid-gap invasion, the movement of the separation point from the downstream corner to the upstream corner of the mid lip, appears near the critical dimensionless bottom-layer flow rate around 0.3. It is always accompanied by an oscillation of the separation point.

Using Navier–Stokes theory, we construct the G/FEM numerical model with two different mesh topologies to track the variation of the interlayer position as the bottom-layer flow rate changes at different viscosity ratio. From the model, we discovered two different mechanisms — onset of turn-around flow and onset of micro vortex — that depend on the viscosity ratio. They share an apparent similar visual phenomenon, the oscillation of the interlayer due to high sensitivity of the flow state to the flow rate.

Furthermore, a simple flow model in the mid-gap region is proposed based on lubrication theory. The model suggests that the appearance of an inflection point attached to the mid lip on the velocity profile signals the onset of mid-gap invasion. By com-
4.5. CONCLUSIONS

bining the critical pressure gradient at the onset of flow reversal and the relationship between the bottom-layer flow rate and wet thickness, a simple criterion is derived. The onset of mid-gap invasion will occur when the dimensionless bottom-layer wet thickness is $1/3$. This simple criterion is supported by the experimental evidences and the G/FEM model results.
Chapter 5

Linear stability analysis of two-layer fixed-gap slot coating

5.1 Introduction

Continuous liquid coating is the main manufacturing step of different products, such as specialty paper, optical films and flexible electronics. It is also a very strong candidate for mass production of nanoparticle assembly films at lower cost, as discussed by Maenosono et al. (2003a). Most of the time, these products have more than one layer. The most efficient way to manufacture multilayer structures is to coat multiple layers simultaneously before they are solidified.

Slot coating can be adapted for coating two layers in what is called the dual slot coating method, as shown in Fig. 5.1. The coating die has two separate feed slots through which each layer is fed. The flow can be divided into three regions separated by feed slots: upstream gap, mid gap and downstream gap. When both liquids are immiscible, they form an interlayer inside the coating bead. In several coating applications, both layers use solvents that are miscible and the apparent interface is truly an inter-diffusion zone. But it is so thin that it can be considered as a distinct interlayer with zero interfacial tension.
Figure 5.1: Two-layer slot coating flow. $H_g$, $L$, $U_w$, $h_{w,1}$ and $h_{w,2}$ are gap height, downstream die lip length, web speed, layer 1 wet thickness and layer 2 wet thickness, respectively. Note that the flow rate is the product of web speed and wet thickness in slot coating process: $q_1 = h_{w,1} U_w$ and $q_2 = h_{w,2} U_w$. Flow region inside dotted box is called downstream coating gap.

Previous researches about dual slot coating method were focused on controlling interlayer separation point (Taylor and Hrymak, 1999; Nam and Carvalho, 2009b). When the interlayer separation point (See Fig. 5.1) transits from the downstream corner of the mid lip to the upstream corner, the top coating layer can invade the mid-gap region accompanied by a dangerous turn-around flow or sometimes the appearance of microvortex that may lead to coating defects. This event is called mid-gap invasion. Nam and Carvalho (2009b) found that when the bottom layer wet thickness $h_{w,2}$ is greater than one-third of gap height $H_g$, mid-gap invasion can be prevented.

Even if the separation point is located at the downstream corner of the mid lip, coating defects associated with instabilities on the interlayer may occur. A waviness in the interlayer can cause quality degradation even when the gas/liquid interface is uniform, especially for optical products. The interlayer stability can be determined at the downstream coating gap (see Fig. 5.1), where flow under the downstream lip is almost rectilinear. In this region, the distance between the downstream die lip and moving web is about $100 \mu m$ and speed of web is about $1 m/s$. Therefore the interlayer suffers from enormous shear that may lead to the unstable configurations.

Historically, Yih (1967) shows that viscosity difference, even in low Reynolds num-
5.1. INTRODUCTION

ber flow, can generate instabilities along the interlayer, so called the interfacial mode in a plane Couette-Poiseuille two-layer channel flow. Extensive studies about the interfacial mode on parallel channel flow has been carried out by many researchers, as Yiantsios and Higgins (1988a) for a pure Poiseuille flow; and Hooper and Boyd (1983) and Renardy (1985) for a pure Couette flow. Beside the interfacial mode, shear mode instability can be manifested by Tollmien-Schlichting waves near the wall of the channel. However, the shear mode appears only at high Reynolds number (Yiantsios and Higgins, 1988a). In a liquid/liquid two phase system, the viscosity difference is not the only mechanism that drives interfacial instability. Density difference can also create Rayleigh-Taylor type instability that is driven by the gravity action perpendicular to the flow direction (Yiantsios and Higgins, 1989), and also a gravity-induced instability driven by the gravity action parallel to the flow direction (Renardy, 1987). One can find an extensive summary about two-layer flow stability in Joseph and Renardy (1993). They pointed out that long wave disturbance at the interlayer can be suppressed by thin-layer effect (Hooper, 1985) and short wave disturbance can be damped out by interfacial tension.

Typically, the flow under the downstream die lip in a two-layer slot coating has small channel height and high web speed, and the two-layer products usually use same or similar solvents for both layers. The flow can be characterized as a low Reynolds number with high shear flow with negligible gravitational and density difference effects. Therefore the interlayer instability is mainly driven by viscosity difference combined with the high shear rate, not by density discrepancy.

The available analysis of two-layer rectilinear flow stability use the interlayer position as the independent parameter. In coating flows, what is fixed is the flow rate of each layer; the interface position varies as the other operating parameters are changed. Therefore, the classical results in the literature cannot be applied directly to determine the interlayer stability in two-layer slot coating.

In this study, we use linear stability theory to analyze the stability of the interlayer inside the high shear zone of a dual slot coating system: the downstream coating gap. The eigenspectrum for the flow system is computed using an efficient numerical method based on Valerio et al. (2007). In order to find the stable region of operating
5.2. LINEAR STABILITY ANALYSIS OF VISCOUS COATING FLOW

conditions, we choose flow rate ratio as the flow parameter that can be controlled directly, instead of thickness ratio. Combined with viscosity ratio, the parameter ranges that guarantee linearly stable coating flow are found. Furthermore, the critical mid-gap invasion condition (Nam and Carvalho 2009b) is also considered together with the stability criteria to define the operating window of the process.

5.2 Linear stability analysis of viscous coating flow

5.2.1 Formulation of equations

We consider a parallel channel to represent the two-layer flow inside the downstream coating gap as shown in Fig. 5.2. The flow system consists of a moving substrate and a stationary wall where the no-slip condition applies. The pressure gradient along the

Figure 5.2: Base flow configuration for the downstream coating gap flow. The system consist of two fluids with the interlayer, moving substrate and stationary wall. \( P_d \) and \( P_u \) are the downstream and upstream pressure, \( \Delta P = P_d - P_u \) is the pressure difference across the flow domain, and \( L \) is the channel. With the pressure gradient across the system, the base flow profile is the combination of Couette and Poisuille flow.
channel \( G \equiv \Delta P/L \), where \( L \) is the channel length, is a function of liquid properties and operating conditions. The base flow configuration have a flat interlayer at \( z = 0 \) with interfacial tension \( \sigma_I \). Velocity and shear stress should be continuous across the interlayer, and normal stresses are balanced at it. The layer 1, the top layer of the coating, occupies \(-H_1 \leq z \leq 0\) and the layer 2, the bottom layer, occupies \(0 \leq z \leq H_2\), respectively. Fluid in layer \( l \) has density \( \rho_l \) and viscosity \( \mu_l \), and each of them is governed by the Navier-Stokes and the continuity equation.

When we use dimensionless numbers as defined in Table 5.1, the base flow velocity profile for layer \( l \) is

\[
U^b_l(z) = \frac{1}{2\mu_l} G z^2 + A_l z + B_l \quad l = 1, 2, \tag{5.1}
\]

where \( A_l \) and \( B_l \) are the coefficient,

\[
A_1 = \frac{U_w}{(m+n)H_2} + \frac{1}{2} \frac{G H_2^2 (n^2 - m)}{\mu_1 (m+n)H_2} = m A_2
\]

\[
B_1 = \frac{U_w n}{(m+n)} - \frac{1}{2} \frac{G H_2^2 (n^2 + n)}{\mu_2 (m+n)} = B_2.
\]

The goal of linear stability analysis is to determine whether the flow is stable or not with respect to infinitesimal disturbances. The perturbed fields and interlayer are

**Table 5.1:** Dimensionless variables used in stability analysis

<table>
<thead>
<tr>
<th>Name</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reynolds number</td>
<td>( N_{Re} = \frac{\rho_2 U_w H_2}{\mu_2} )</td>
</tr>
<tr>
<td>Viscosity ratio</td>
<td>( m = \frac{\mu_2}{\mu_1} )</td>
</tr>
<tr>
<td>Density ratio</td>
<td>( r = \frac{\rho_2}{\rho_1} )</td>
</tr>
<tr>
<td>Thickness ratio</td>
<td>( n = \frac{H_1}{H_2} )</td>
</tr>
<tr>
<td>Dimensionless pressure gradient</td>
<td>( N_G = \frac{(\Delta P/L) H_2^2}{\mu_2 U_w} )</td>
</tr>
<tr>
<td>Interfacial tension number</td>
<td>( N_T = \frac{\sigma_I}{\mu_2 U_w} )</td>
</tr>
</tbody>
</table>
written as the sum of the base states and infinitesimal disturbances:

\[ u^p(x, t) = i U^p_l(x_0) + \varepsilon \hat{u}_l \]
\[ P^p_l(x, t) = P^0_l(x_0) + \varepsilon \hat{P}_l \]
\[ h^p(x_{\Sigma}, t) = h^0(x_{0,\Sigma}, t) + \varepsilon \hat{h} = \varepsilon \hat{h} \]  \hspace{1cm} (5.2)

where the superscript \( p \) stands for perturbed flow, \( i \) is the basis vector for \( x \) direction, \( P^0_l \) is the base flow pressure field, hat(\( ^\wedge \)) stands for the magnitude of disturbance in the unit of \( \varepsilon \) from the base flow, subscript \( l \) can be either 1 or 2 depending on liquid, and subscript \( \Sigma \) stands for the interlayer, i.e. \( x_{0,\Sigma} \) is the position of the interlayer in the base flow configuration.

The perturbed flow is governed by the time-dependent Navier-Stokes/continuity equations with appropriate boundary conditions.

\[ \rho_l \frac{Du^p_l}{Dt} = -\nabla P^p_l + \mu_l \nabla^2 u^p_l, \]  \hspace{1cm} (5.3)
\[ \nabla \cdot u^p_l = 0 \]  \hspace{1cm} (5.4)

where \( D/Dt \) is the material derivative.

No slip condition applies to both moving and stationary walls:

\[ u^p_l(z = -H_1) = 0, \]
\[ u^p_l(z = H_2) = i U_w. \]  \hspace{1cm} (5.5)

At the perturbed interlayer, velocity and shear stress are continuous:

\[ u^p_1 - u^p_2 \bigg|_{x = x_{\Sigma}} = 0, \]  \hspace{1cm} (5.6)
\[ t \cdot [ T(u^p_1) - T(u^p_2) ] \cdot n \bigg|_{x = x_{\Sigma}} = t \cdot \nabla \sigma_l \bigg|_{x = x_{\Sigma}}, \]  \hspace{1cm} (5.7)

where \( T \) is the state of stress. Note that Eq. (5.7) is set to zero, when the interfacial tension is uniform along the interlayer.

The normal stress is also balanced at the interlayer,

\[ n \cdot [ T(u^p_1) - T(u^p_2) ] \cdot n \bigg|_{x = x_{\Sigma}} = 2H \sigma_l, \]  \hspace{1cm} (5.8)
where the mean curvature at the interlayer $2H$ is given by

$$2H = \nabla_{II} \cdot n = \frac{\nabla_{II}^2 h^p - (\mathbf{k} \times \nabla_{III} h^p)(\mathbf{k} \times \nabla_{II} h^p) : \nabla_{II} \nabla_{II} h^p}{(1 + (\nabla_{II} h^p)^2)^{3/2}}. \tag{5.9}$$

The motion of the interlayer is described by

$$w^p = \frac{D h^p}{D t} = \frac{\partial h^p}{\partial t} + u^p \frac{\partial h^p}{\partial x} + v^p \frac{\partial h^p}{\partial y}. \tag{5.10}$$

The linear differential equations that describes the disturbances are obtained after substituting the perturbed field onto the transient Navier-Stokes / continuity equations and boundary conditions, and neglecting terms of order higher than $O(\epsilon^2)$. For example, the linear term of the perturbed Navier-Stokes / continuity equations are

$$\nabla \cdot \hat{\mathbf{u}}_l = 0 \tag{5.11}$$

$$\rho_l \frac{\partial \hat{\mathbf{u}}_l}{\partial t} + \rho_l \left[ (\mathbf{i} U^p_l) \cdot \nabla \hat{\mathbf{u}}_l + \hat{\mathbf{u}}_l \cdot \nabla (\mathbf{i} U^p_l) \right] = -\nabla \hat{P}_l + \mu_l \nabla^2 \hat{\mathbf{u}}_l. \tag{5.12}$$

It follows that we can separate variables, so that the general solution of the initial-value problem is a linear superposition of normal modes, each of the form

$$\hat{\mathbf{u}}(x, t) = u(z) \exp(i \alpha x + i \beta y + \omega t),$$

$$\hat{P}(x, t) = P(z) \exp(i \alpha x + i \beta y + \omega t),$$

$$\hat{h}(x, t) = h \exp(i \alpha x + i \beta y + \omega t), \tag{5.13}$$

where $\alpha$ and $\beta$ are wavenumbers of an imposed spatial periodic disturbances, and $\omega$ is the growth rate. When the real part of $\omega$ is less than zero, the flow system is stable with respect to the specified periodic disturbances.

Further simplification is possible due to Squire’s theorem for a two-layer flow system (Hesla et al., 1986). According to the theorem, critical parameter values for the stability limit are completely determined by a two-dimensional perturbation. The flow system for two-dimensional disturbances can be easily obtained by setting $\beta = 0$. The governing equations and boundary conditions for the downstream coating gap
5.2. LINEAR STABILITY ANALYSIS OF VISCOUS COATING FLOW

flow system are summarized below. The momentum equation in the flow direction becomes Equation of motion becomes

\[
\omega \rho u_i + \rho (i \alpha U_i^b) u_i + \rho \frac{dU_i^b}{dz} \omega_i = -i \alpha P_i + \mu \left( \frac{d^2 u_i}{dz^2} - \alpha^2 u_i \right),
\]

(5.14)

and the continuity equation becomes

\[
i \alpha u_i + \frac{d\omega_i}{dz} = 0.
\]

(5.15)

The no-slip condition at the stationary wall becomes

\[
u_i(z = -H_1) = \omega_i(z = -H_1) = 0,
\]

(5.16)

the no-slip at the moving web becomes

\[
u_i(z = H_2) = \omega_i(z = H_2) = 0,
\]

(5.17)

the velocity continuity at the interlayer becomes

\[
u_i(z = 0) + h \frac{dU_i}{dz} \bigg|_{z=0} = \omega_i(z = 0) + h \frac{dU_2}{dz} \bigg|_{z=0}, \quad \omega_i(z = 0) = \omega_2(z = 0),
\]

(5.18)

the normal stress balance at the interlayer becomes

\[
\left[ -(P_1 - P_2) + \left( 2 \mu_1 \frac{d\omega_1}{dz} - 2 \mu_2 \frac{d\omega_2}{dz} \right) \right] \bigg|_{z=0} + \left( \alpha^2 \sigma_1 \right) h = 0,
\]

(5.19)

the shear stress continuity at the interlayer becomes

\[
\left[ \mu_1 \left( h \frac{d^2 U_1}{dz^2} + \frac{dU_1}{dz} + i \alpha \omega_1 \right) - \mu_2 \left( h \frac{d^2 U_2}{dz^2} + \frac{dU_2}{dz} + i \alpha \omega_2 \right) \right] \bigg|_{z=0} = 0,
\]

(5.20)

and kinematic condition at the interlayer becomes

\[
\omega h + i \alpha U_i^b(z = 0) h - \omega_i(z = 0) = 0.
\]

(5.21)

5.2.2 Discretization by Galerkin’s method and finite element basis functions

Given a periodic disturbance \( \alpha \), the perturbed fields \( \mathbf{u}, P, h \) and the growth rate \( \omega \) can be computed by applying Galerkin’s weighted residual method to equations
5.2. LINEAR STABILITY ANALYSIS OF VISCOUS COATING FLOW

Eqs. (5.14) — (5.21). The weighting functions for the momentum equation $\phi_j$ and the continuity equation $\psi_j$ are piecewise Lagrangian quadratic basis function and linear discontinuous basis function, respectively.

However, in this particular linearized system, the velocity continuity condition at the interlayer requires a jump of the velocity field disturbance across the interlayer, as indicated in Eq. (5.18). In order to handle it, we use two nodes at the interlayer: two degrees of freedom for the interfacial velocity component are assigned to the point, located at the interlayer. The pressure jump across the interlayer, a consequence of the normal stress balance, is fulfilled naturally by the linear discontinuous basis function. Therefore, the field variables are approximated by

$$u_l = \sum_{j=1}^{2N+2} U_j \phi_i,$$
$$p_l = \sum_{j=1}^{2N} P_j \psi_i,$$  \hspace{1cm} (5.22)

where $N$ is the number of elements used to discretize the domain, $U_j = iU_j + kW_j$ and $P_j$ are the coefficients for velocity vector and pressure, respectively. $l = 1$ is for $-H_1 \leq z \leq 0$, and $l = 2$ for $0 \leq z \leq H_2$. The interlayer location $h$ is assigned to single degree of freedom in this system. Note that, at the interlayer, the basis functions associated to the layer 1 and the layer 2 do not span across the other layer.

In order to obtain accurate predictions of growth rates and corresponding disturbed fields in cases at which the most dangerous modes are associated with disturbances at the interlayer, a fine mesh is necessary. To minimize the number of degree of freedom of the discrete system, we use a non-uniform mesh. We increase node concentration around the interlayer using stretching function (Vinokur, 1983b). However, as pointed out by Yiantsios and Higgins (1987), the mesh refinement requires special care in order to avoid erroneous conclusions. We compare the results from a fine uniform mesh (400 elements) with those obtained with a graded mesh (100 elements) to confirm that the graded mesh can give an accurate stability prediction with less computational time.
Using integral by parts, the weighted residual equation can be expressed as

\[ R_{mx}^j = \omega \left[ \int_{-H_1}^{0} (\rho_1 \phi_j u_1) \, dz + \int_{0}^{H_2} (\rho_2 \phi_j u_2) \, dz \right] = R_{t, mx}^j \]

\[ + \left[ \int_{-H_1}^{0} \left( \rho_1 i \alpha U_1^b \phi_j u_1 + \rho_1 \frac{d U_1^b}{dz} \phi_j w_1 \right) \, dz + i \alpha \phi_j P_1 + \mu_1 \frac{d \phi_j}{dz} w_1 \right] \]

\[ + \int_{0}^{H_2} \left( \rho_2 i \alpha U_2^b \phi_j u_2 + \rho_2 \frac{d U_2^b}{dz} \phi_j w_2 \right) \, dz + i \alpha \phi_j P_2 + \mu_2 \frac{d \phi_j}{dz} w_2 \right] \]

\[ = R_{s, mx}^j, \quad (5.23) \]

\[ R_{mz}^j = \omega \left[ \int_{-H_1}^{0} (\rho_1 \phi_j w_1) \, dz + \int_{0}^{H_2} (\rho_2 \phi_j u_2) \, dz \right] = R_{t, mz}^j \]

\[ + \left[ \int_{-H_1}^{0} \left( \rho_1 i \alpha U_1^b \phi_j w_1 - \frac{d \phi_j}{dz} P_1 + \mu_1 \frac{d \phi_j}{dz} w_1 + \mu_1 \alpha^2 \phi_j u_1 \right) \, dz \right. \]

\[ + \int_{0}^{H_2} \left( \rho_2 i \alpha U_2^b \phi_j w_2 - \frac{d \phi_j}{dz} P_2 + \mu_2 \frac{d \phi_j}{dz} w_2 + \mu_2 \alpha^2 \phi_j u_2 \right) \, dz \]

\[ = R_{s, mz}^j, \quad (5.24) \]
### 5.2. LINEAR STABILITY ANALYSIS OF VISCOUS COATING FLOW

<table>
<thead>
<tr>
<th>Block # in Jacobian and mass matrix (Size of partition)</th>
<th>Sequence of entries for residual and solution vector</th>
<th>The weighted residual</th>
<th>Unknown coefficient for GFEM system</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 (2N)</td>
<td>Internal $x$-momentum conservation</td>
<td>1</td>
<td>Internal $x$-velocity d.o.f</td>
</tr>
<tr>
<td></td>
<td>Normal stress balance</td>
<td>2N-2</td>
<td>Interfacial $z$-velocity d.o.f for layer 1</td>
</tr>
<tr>
<td>2 (2N-2)</td>
<td>Internal $z$-momentum conservation</td>
<td>2N-1</td>
<td>Interfacial $z$-velocity d.o.f for layer 1</td>
</tr>
<tr>
<td></td>
<td>Kinematic condition</td>
<td>4N-3</td>
<td>Interfacial height d.o.f</td>
</tr>
<tr>
<td>3 (2N)</td>
<td>Mass conservation</td>
<td>4N-1</td>
<td>Pressure d.o.f</td>
</tr>
<tr>
<td>4 (3)</td>
<td>Shear stress continuity</td>
<td>6N-1</td>
<td>Interfacial $x$-velocity d.o.f for layer 1</td>
</tr>
<tr>
<td></td>
<td>$x$-velocity continuity</td>
<td>6N</td>
<td>Interfacial $x$-velocity d.o.f for layer 2</td>
</tr>
<tr>
<td></td>
<td>$z$-velocity continuity</td>
<td>6N+1</td>
<td>Interfacial $z$-velocity d.o.f for layer 2</td>
</tr>
<tr>
<td>5 (4)</td>
<td>No-slip conditions</td>
<td>6N+2</td>
<td>Boundary velocity d.o.f</td>
</tr>
</tbody>
</table>

![Figure 5.3](image_url): The sequence of entries in weighted residual equations inside residual vector and unknown coefficient for finite element expansion inside solution vector.

\[
R_f^i = \int_{-H_1}^{0} \left[ i \alpha \psi_j u_1 + \psi_j \frac{dw_1}{dz} \right] dz \\
+ \int_{0}^{H_2} \left[ i \alpha \psi_j u_2 + \psi_j \frac{dw_2}{dz} \right] dz.
\]  

Note that the momentum residual can be divided into time-dependent terms that are multiplied by the growth rate $\omega$.

The number of algebraic equations is $6N+5$, where $N$ is the number of finite elements: $4N-4$ momentum equations, $2N$ continuity equations, $4$ no-slip conditions, and $5$
interfacial conditions. In vector form, the set of equations is represented by \( \mathbf{R}(\mathbf{c}) = 0 \), where \( \mathbf{R} \) is the vector of weighted residual equations and \( \mathbf{c} \) consist of the coefficients of the finite element basis functions. For the purpose of the method we used to solve the problem, the sequential ordering of equations and degree of freedoms is chosen as in Fig. 5.3.

When the perturbation is infinitesimal, one can truncate Taylor series expansion of the set of equations at order \( \mathcal{O}(c^2) \):

\[
\frac{\partial \mathbf{R}}{\partial \mathbf{c}} \mathbf{c} = \left( \frac{\partial \mathbf{R}_t}{\partial \mathbf{c}} + \frac{\partial \mathbf{R}_s}{\partial \mathbf{c}} \right) \mathbf{c} = (-\omega \mathbf{M} + \mathbf{J}) \mathbf{c} = 0,
\]

(5.26)

where \( \frac{\partial \mathbf{R}}{\partial \mathbf{c}} \) is the sensitivities of the weighted residual with respect to the unknown coefficient of the perturbation, \( \mathbf{R}_t \) and \( \mathbf{R}_s \) stand for time-dependent and time-independent parts of residuals, \( \mathbf{M} \) and \( \mathbf{J} \) are called the mass matrix and the Jacobian matrix. Thus the set of differential equations leads to the generalized eigenvalue problem (GEVP)

\[
\mathbf{J} \mathbf{c} = \omega \mathbf{M} \mathbf{c}.
\]

(5.27)

The Jacobian and mass matrix entries are summarized in Tables 5.2 and 5.3.

### 5.2.3 Filtering eigenvalues at infinity

The mass matrix \( \mathbf{M} \) is singular, because the continuity equation for incompressible fluid, the no-slip boundary conditions and the interfacial conditions, except the kinematic condition, have no time derivative. Thus, the number of finite eigenvalues of the generalized eigenvalue problem, Eq. (5.27), is smaller than the dimension of the problem \( 6N + 5 \). The missing eigenvalues are commonly referred to as *eigenvalues at infinity*, because if the mass matrix is slightly perturbed to remove the singularity, e.g. \( \mathbf{M}^* = \mathbf{M} + \epsilon \mathbf{I} \), large eigenvalues appear in the spectrum, and they grow unbounded as \( \epsilon \to 0 \) (Valerio et al., 2007). During numerical computation of the eigenspectrum, truncation errors and round-off errors may cause perturbations of the mass matrix and lead to the eigenvalues at infinity — unrealistically large eigenvalues — of the GEVP. The method of filtering eigenvalues at infinity for the single-layer channel flow was discussed in Valerio et al. (2007). The original GEVP, Eq. (5.27), can be
5.2. LINEAR STABILITY ANALYSIS OF VISCOUS COATING FLOW

Table 5.2: Mass and Jacobian matrix entries for the two-layer parallel flow system. $l = 1$ is for the first layer and $l = 2$ for the second layer. $\Omega_1$ and $\Omega_2$ span $-H_1 \leq z \leq 0$ and $0 \leq z \leq H_2$, respectively.

<table>
<thead>
<tr>
<th>Name</th>
<th>Equations</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mass matrix</td>
<td>$-\frac{\partial R_{i,mx}^l}{\partial U_k} = -\int_{\Omega_l} \rho_l \phi_j \phi_k , dz$</td>
</tr>
<tr>
<td></td>
<td>$-\frac{\partial R_{i,mz}^l}{\partial W_k} = -\int_{\Omega_l} \rho_l \phi_j \phi_k , dz$</td>
</tr>
<tr>
<td>Jacobian matrix</td>
<td>$\frac{\partial R_{s,mx}^l}{\partial U_k} = \int_{\Omega_l} \left[ \rho_l i \alpha U_i^b \phi_j \phi_k + \mu_l \frac{d\phi_j}{dz} \frac{d\phi_k}{dz} + \mu_l \alpha^2 \phi_j \phi_k \right] , dz$</td>
</tr>
<tr>
<td></td>
<td>$\frac{\partial R_{s,mx}^l}{\partial W_k} = \int_{\Omega_l} \left[ \rho_l \frac{dU_i^b}{dz} \phi_j \phi_k \right] , dz$</td>
</tr>
<tr>
<td></td>
<td>$\frac{\partial R_{s,mx}^l}{\partial P_k} = \int_{\Omega_l} \left[ i \alpha \phi_j \psi_k \right] , dz$</td>
</tr>
<tr>
<td></td>
<td>$\frac{\partial R_{s,mz}^l}{\partial W_k} = \int_{\Omega_l} \left[ \rho_l i \alpha U_i^b \phi_j \phi_k + \mu_l \frac{d\phi_j}{dz} \frac{d\phi_k}{dz} + \mu_l \alpha^2 \phi_j \phi_k \right] , dz$</td>
</tr>
<tr>
<td></td>
<td>$\frac{\partial R_{s,mz}^l}{\partial P_k} = \int_{\Omega_l} \left[ -\frac{d\phi_j}{dz} \psi_k \right] , dz$</td>
</tr>
<tr>
<td></td>
<td>$\frac{\partial R_{c}^l}{\partial U_k} = \int_{\Omega_l} \left[ i \alpha \psi_j \phi_k \right] , dz$</td>
</tr>
<tr>
<td></td>
<td>$\frac{\partial R_{c}^l}{\partial W_k} = \int_{\Omega_l} \left[ \psi_j \frac{d\phi_k}{dz} \right] , dz$</td>
</tr>
</tbody>
</table>
5.2. LINEAR STABILITY ANALYSIS OF VISCOSOUS COATING FLOW

**Table 5.3:** Mass and Jacobian matrix entries for interfacial and boundary conditions in the two-layer parallel flow system. Interfacial node for velocity is $N_i$ and pressure node near the interlayer is $N_{p,i}$ and $N_{p,i+1}$. Note that the position where interfacial condition are evaluated is $z = 0$.

<table>
<thead>
<tr>
<th>Name</th>
<th>Equations</th>
</tr>
</thead>
</table>
| **Shear stress continuity**  | $\frac{\partial R_{s,m,x}^{N_i}}{\partial U_k} = \begin{cases} 
\mu_1 \frac{d\phi_k}{dz} & (k = N_i, N_{i-1}, N_{i-2}) \\
\mu_1 \frac{d\phi_k}{dz} - \mu_2 \frac{d\phi_k}{dz} & (k = N_i) \\
-\mu_2 \frac{d\phi_k}{dz} & (k = N_{i+1}, N_{i+2}, N_{i+3})
\end{cases}$ |
|                           | $\frac{\partial R_{s,m,x}^{N_i}}{\partial W_k} = \begin{cases} 
i \mu_1 a & (k = N_i) \\
-i \mu_2 a & (k = N_{i+1})
\end{cases}$ |
|                           | $\frac{\partial R_{s,m,x}}{\partial h} = \mu_1 \frac{d^2 U_1}{dz^2} - \mu_2 \frac{d^2 U_2}{dz^2}$ |
| **Normal stress balance**  | $\frac{\partial R_{s,m,z}^{N_i}}{\partial U_k} = \begin{cases} 
2 \mu_1 \frac{d\phi_k}{dz} & (k = N_i, N_{i-1}, N_{i-2}) \\
-2 \mu_2 \frac{d\phi_k}{dz} & (k = N_{i+1}, N_{i+2}, N_{i+3})
\end{cases}$ |
|                           | $\frac{\partial R_{s,m,z}}{\partial W_k} = \begin{cases} 
-\psi_k & (k = N_{p,i-1}, N_{p,i}) \\
\psi_k & (k = N_{p,i+1}, N_{p,i+2})
\end{cases}$ |
|                           | $\frac{\partial R_{s,m,z}}{\partial h} = \alpha^2 \sigma_i$ |
| **Velocity continuity**    | $\frac{\partial R_{s,m,x}^{N_i}}{\partial U_k} = \begin{cases} 
1 & (k = N_i) \\
-1 & (k = N_{i+1})
\end{cases}$ |
|                           | $\frac{\partial R_{s,m,x}}{\partial W_k} = \frac{dU_1}{dz} \bigg|_{z=0} - \frac{dU_2}{dz} \bigg|_{z=0}$ |
|                           | $\frac{\partial R_{s,m,z}^{N_i}}{\partial U_k} = \begin{cases} 
1 & (k = N_i) \\
-1 & (k = N_{i+1})
\end{cases}$ |
|                           | $\frac{\partial R_{s,m,z}}{\partial W_k} = \begin{cases} 
i \alpha U_1^h(z = 0)
\end{cases}$ |
| **Kinematic condition**    | $-\frac{\partial R_{s,K}}{\partial h} = -1$ |
|                           | $\frac{\partial R_{s,K}}{\partial W_k} = -1 & (k = N_i)$ |
|                           | $\frac{\partial R_{s,K}}{\partial h} = i \alpha U_1^h(z = 0)$ |
| **No-slip condition**      | $\frac{\partial R_{s,m,x}}{\partial U_k} = 1 & (k = 1, N_{v1} + N_{v2})$ |
|                           | $\frac{\partial R_{s,m,z}}{\partial W_k} = 1 & (k = 1, N_{v1} + N_{v2})$ |
transformed into the smaller eigenvalue problem that do not contain eigenvalues at infinity. The basis of the method, that was extended here for the two-layer flow, is briefly summarized here.

First, entries of the matrix system need to be ordered based on the numbering scheme presented in Fig. [5.3]. Then both the mass and Jacobian matrices can be partitioned in $5 \times 5$ block structure with square blocks along the diagonal. The configuration of the matrices is shown below.

$$M = \begin{pmatrix}
M_{11} & M_{12} & 0 & M_{14} & M_{15} \\
M_{22} & M_{22} & 0 & M_{24} & M_{25} \\
0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0
\end{pmatrix} \quad 2N \quad 2N - 2 \quad 2N \quad 3 \quad 4$$

$$J = \begin{pmatrix}
J_{11} & J_{12} & J_{13} & J_{14} & J_{15} \\
J_{21} & J_{22} & J_{23} & J_{24} & J_{25} \\
J_{31} & J_{32} & 0 & J_{34} & J_{35} \\
J_{41} & J_{42} & 0 & J_{44} & J_{45} \\
0 & 0 & 0 & 0 & I_{[4]}
\end{pmatrix} \quad 2N \quad 2N - 2 \quad 2N \quad 3 \quad 4$$

Note that most of the subblocks are zero, especially in the mass matrix, and the last block of Jacobian $I_{[4]}$ is the identity matrix of dimension four.

After elimination of the rows and columns related to no-slip boundary conditions, a $4 \times 4$ sub block structure of the mass matrix $M^b$ and the Jacobian matrix $J^b$ remains. It is convenient to define the block structure of $B = J^b - \omega M^b$ as:

$$B = \begin{pmatrix}
B_{11}(\omega) & B_{12}(\omega) & B_{13} & B_{14}(\omega) \\
B_{21}(\omega) & B_{22}(\omega) & B_{23} & B_{24}(\omega) \\
B_{31} & B_{32} & 0 & B_{34} \\
B_{41} & B_{42} & 0 & B_{44}
\end{pmatrix}$$

$$= B T_r^{(1)}$$

Note that $B_{44}$ is always invertible when $J^b$ is not singular. Because the $4 \times 6N + 1$ size sub block $[J_{41}, J_{42}, 0, J_{44}]$ should have rank 3, but $J_{41}$ and $J_{42}$ are always rank deficient matrix. Exploiting this fact, one can construct a transformation matrix $T_r^{(1)}$ to eliminate block matrices $B_{14}$ and $B_{41}$.
5.2. LINEAR STABILITY ANALYSIS OF VISCOSOUS COATING FLOW

where

\[
T^{(1)}_r = \begin{pmatrix}
I_{[2N]} & 0 & 0 & 0 \\
0 & I_{[2N-2]} & 0 & 0 \\
0 & 0 & I_{[2N]} & 0 \\
-B_{44}^{-1}B_{41} & -B_{44}^{-1}B_{42} & 0 & I_{[3]}
\end{pmatrix}
\] (5.30)

We define \( A \) to be the first \( 3 \times 3 \) blocks of \( \tilde{B} \),

\[
A = \begin{pmatrix}
\tilde{B}_{11}(\omega) & \tilde{B}_{12}(\omega) & \tilde{B}_{13} \\
\tilde{B}_{21}(\omega) & \tilde{B}_{22}(\omega) & \tilde{B}_{23} \\
\tilde{B}_{31} & \tilde{B}_{32} & 0
\end{pmatrix}
\] (5.31)

Now, the matrix structure become similar to a plane single-layered flow with Newtonian fluid \(^{[\text{Valerio et al., 2007}]\). As mentioned in their work, \( \tilde{B}_{31} \) and \( B_{13} \) are invertible, when matrix \( \tilde{B} \) is non-singular. Therefore, one can construct matrices \( T^{(2)}_l \) and \( T^{(2)}_r \) to eliminate \( \tilde{B}_{32} \) and \( B_{23} \) from \( A \), when the Jacobian is not singular.

\[
\tilde{A} = \begin{pmatrix}
\tilde{B}_{11}(\omega) & \tilde{A}_{12}(\omega) & \tilde{B}_{13} \\
\tilde{A}_{21}(\omega) & \tilde{A}_{22}(\omega) & 0 \\
\tilde{B}_{31} & 0 & 0
\end{pmatrix} = T^{(2)}_l A T^{(2)}_r
\] (5.32)

where

\[
T^{(2)}_l = \begin{pmatrix}
I_{[2N]} & 0 & 0 \\
0 & I_{[2N-2]} & 0 \\
0 & 0 & I_{[2N]}
\end{pmatrix}
\quad T^{(2)}_r = \begin{pmatrix}
I_{[2N]} & -\tilde{B}_{31}^{-1}\tilde{B}_{32} & 0 \\
0 & I_{[2N-2]} & 0 \\
0 & 0 & I_{[2N]}
\end{pmatrix}
\] (5.33)

One can prove that the eigenspectrum of \( \tilde{A}_{22} = \tilde{J}_{22} - \omega\tilde{M}_{22} \) is the same as the original generalize eigenvalue problem, Eq. (5.27), when the original Jacobian matrix is non-singular, as in \(^{[\text{Valerio et al., 2007}]\), by comparing characteristic polynomials for both equations. The smaller GEVP which contain the finite portion of the spectrum of the original can be written as

\[
\tilde{J}_{22} c_2 = \omega\tilde{M}_{22} c_2.
\] (5.34)

Note that the generalized eigenvectors \( c \) of the original GEVP, Eq. (5.27), can be recovered from the eigenvector \( c_2 \) by the reverse process of the transformation.
5.2. LINEAR STABILITY ANALYSIS OF VISCOUS COATING FLOW

Most numerical methods to compute eigenvalues are based on an iterative scheme, like QR iteration based on Schur decomposition. The convergence property and accuracy of the computed eigenvalues depend on the separation between eigenvalues (Golub and Loan [1996]). The separation between eigenvalues can be adjusted easily by using the shift-and-invert method:

\[
\left( \tilde{J}_{22} - \delta \tilde{M}_{22} \right)^{-1} \tilde{M}_{22} c_2 = \frac{1}{\omega - \delta} c_2,
\]

(5.35)

where \( \delta \) is shift factor. In this study, we choose \( \delta = 0 \). Because the dimension of the reduced eigenvalue problem is about one third of that of original one, the method on the reduced problem can save significant amount of computational times. In this study, we use LAPACK subroutine ZGEEV to solve Eq. (5.35). Detailed discussions about the algorithm of filtering eigenvalues at infinity and the validations of the linear stability model are discussed at Appendix C.

5.2.4 Flow rate ratio continuation

In two-layer slot coating, the independent flow variable is the flow rate ratio \( f = q_1/q_2 \) not the thickness ratio that naturally appears in the formulation. Therefore, we have to solve the eigenproblem using the flow rate as the independent parameters. However, it is extremely difficult to express a close form of the base flow velocity profile as a function of the flow rates and their ratio. However, at a fixed set of parameters, the thickness of each layer can be founded by solving the flow rate equations:

\[
q_1 = \frac{(\Delta P/L)}{6\mu_1} H_1^2 + \frac{1}{2} \left( \frac{U_w}{(m+n)H_2} H_1^2 + \frac{1}{2} \frac{(\Delta P/L)H_2^2}{\mu_1} \frac{n^2 - m}{(m+n)H_2} \right) H_1^2
\]

\[
+ \left( \frac{U_w n}{n+m} - \frac{1}{2} \frac{(\Delta P/L)H_2^2}{\mu_2} \frac{n^2 + n}{m+n} \right) H_1,
\]

(5.36)

\[
q_2 = \frac{(\Delta P/L)}{6\mu_2} H_2^2 + \frac{1}{2} \left( \frac{U_w}{(m+n)H_2} H_2^2 + \frac{1}{2} \frac{(\Delta P/L)H_2^2}{\mu_1} \frac{n^2 - m}{(m+n)H_2} \right) m H_2^2
\]

\[
+ \left( \frac{U_w n}{n+m} - \frac{1}{2} \frac{(\Delta P/L)H_2^2}{\mu_2} \frac{n^2 + n}{m+n} \right) H_2.
\]

(5.37)

Note that the interlayer location is not only a function of flow rate ratio but also a function of other parameters, like viscosity ratio. We use Newton’s method to solve these equations to find \( H_2 \) and \( \Delta P/L \) at a given set of flow parameters.
The flow rate equations, Eqs. (5.36) and (5.37), allow multiple solutions. Each solution with respect to the flow rate ratio constructs a trajectory in a parameter map, so called a solution branch. Each branch shows different stability behavior. However, not all of them are physically acceptable, for example, complex values of pressure or interface location, or negative interface location.

In order to track a single branch, we deploy a method of continuation. One simple way to do this is to use the solution for the previous continuation step as a initial guess for the next step, the so called zeroth order continuation. Therefore, once we pick a correct initial guess for the first continuation step, the stability of the flow can be analyzed along a specific solution branch. In this study, we perform the flow rate ratio continuation keeping the total flow rate constant: the change of flow rate ratio was carried out by adjusting $q_1$ and $q_2$ simultaneously.

Here, we propose a two step method to perform the stability analysis on the flow:

1. Solving flow rate equations for each layer to find the interlayer location,
2. Converting the interlayer location to thickness ratio and use it with the conventional base flow profile to perform the linear stability analysis.

5.3 Results & Discussion

We use linear stability analysis to find the desirable operating condition ranges for the dual slot coating method. One way to visualize the stable region in the parameter space is to construct the neutral stability curve or neutral curve, by collecting neutrally stable points with respect to different flow parameters and wavenumbers. Furthermore, since our target is to find the ranges for defect-free products, the stability results obtained here are combined with the critical mid-gap invasion conditions presented by [Nam and Carvalho (2009b)].
5.3. RESULTS & DISCUSSION

Figure 5.4: Results from two-layer slot coating flow stability model without interfacial tension for density ratio $r = 1$, viscosity ratio $m = 2$, flow rate ratio $f = 1$ and dimensionless wavenumber $\alpha^* = 3.80$. (a) shows ten loading eigenvalues from eigenspectrum. (b), (c) and (d) plot show eigenvectors corresponding to the solid dot in (a).
5.3. RESULTS & DISCUSSION

5.3.1 Construction of the neutral curve

The solution of the GEVP, Eq. (5.27), at a given set of flow parameters and wavenumber is a set of eigenpairs. Here, we use dimensionless variables based on the characteristic length unit $H_g$ and time unit $H_g/U_w$ to display the results. Flow parameters and operating conditions of the base are summarized in table 5.4. The most dangerous eigenvalue that can lead to flow instability are those with the largest real part, or growth rate. The solid dot in Fig. 5.4 (a) is the most dangerous eigenvalue at dimensionless wave number $\alpha^* = 3.80$ with the largest real part $\omega_{\alpha, MD}^* = 5.43 \times 10^{-3}$. The corresponding eigenvectors are presented in Fig. 5.4 (b), (c) and (d). The plots show the modulus of eigenvector components with respect to vertical coordinate $z^*$ for horizontal velocity $u$, vertical velocity $w$, and pressure $p$, respectively. Note that the moduli are normalized with respect to the maximum value for each variables. The eigenvector plots show that the disturbances are concentrated near the interlayer — interfacial mode. We found that the interfacial mode is always the most dangerous disturbance for the parameter ranges considered in this study.

When we collect the most dangerous growth rate at different wavenumbers and flow parameters, two types of growth rate plots are obtained, as shown in Fig. 5.5. Plot

<table>
<thead>
<tr>
<th>Name</th>
<th>Base case</th>
</tr>
</thead>
<tbody>
<tr>
<td>Web speed ($U_w$)</td>
<td>1 m/s</td>
</tr>
<tr>
<td>Die lip length ($L$)</td>
<td>1 mm</td>
</tr>
<tr>
<td>Gap height ($H_g$)</td>
<td>250 µm</td>
</tr>
<tr>
<td>Layer 1 Viscosity ($\mu_1$)</td>
<td>46 cP</td>
</tr>
<tr>
<td>Layer 2 Viscosity ($\mu_2$)</td>
<td>23 cP</td>
</tr>
<tr>
<td>Layer 1 density ($\rho_1$)</td>
<td>1.2 g/ml</td>
</tr>
<tr>
<td>Layer 2 density ($\rho_2$)</td>
<td>1.2 g/ml</td>
</tr>
<tr>
<td>Layer 1 wet thickness ($h_{w,1}$)</td>
<td>50 µm</td>
</tr>
<tr>
<td>Layer 2 wet thickness ($h_{w,2}$)</td>
<td>50 µm</td>
</tr>
</tbody>
</table>
5.3. RESULTS & DISCUSSION

-0.4
0.0
1
1\cdot10^{-2}
1\cdot10^{-1}
1\cdot10^{0}
1\cdot10^{1}
1\cdot10^{2}

-4.0\cdot10^{-3}
-1.0\cdot10^{-3}
0.0\cdot10^{0}
1.0\cdot10^{-3}

Figure 5.5: Most dangerous growth rate versus wavenumber plot at fixed flow rate ratio \( f = 1 \) (a) and Most dangerous growth rate versus flow rate ratio plot at fixed wavenumber \( \alpha^* = 3.80 \) (b). Small plot inside (a) shows the base flow velocity profile for given parameters.

(a) was constructed by finding the most dangerous eigenvalue from a wavenumber range scanned over at the fixed flow rate ratio \( f = 1 \). We will call it wavenumber continuation. Plot (b) was constructed using flow rate ratio continuation, as discussed in section 5.2.4 at wavenumber \( \alpha^* = 3.8 \). In both plots, the density ratio and viscosity ratio are fixed to \( r = 1 \) and \( m = 2 \). The solid dot in both plots represents to most dangerous eigenvalue of Fig. 5.4 (a). The open triangle point and square point in figures 5.5(a) and (b) are neutrally stable points at which the growth rate vanishes. The corresponding wavenumber and flow rate ratio are \( \alpha_N^* = 6.75 \) and \( f_N = 0.535 \), respectively. In plot (a), the flow is linearly stable only when the wavenumber is lower than \( \alpha_N^* \); and, in plot (b), the flow is stable only when flow rate ratio is higher than \( f_N \).

Neutral curves can be constructed by collecting neutrally stable points with respect to wavenumbers and flow parameters. Fig. 5.6 shows a representative neutral curve that depicts stable and unstable region inside the parameter space of wavenumber and flow rate ratio. Neutral curve with solid line was obtained without interfacial tension, and dotted line was obtained with interfacial tension, e.g. \( \sigma_I = 1 \text{ dyne/cm} \).
5.3. RESULTS & DISCUSSION

Figure 5.6: Neutral curve for density ratio $r = 1$ and viscosity ratio $m = 2$ with or without interfacial tension. Interfacial tension used in this study is $\sigma_I = 1$ dyne/cm. Note that triangular point and square point are neutrally stable points obtained from Fig. 5.5 (a) and (b), respectively. “U” and “S” stand for unstable interlayer and stable interlayer, respectively.
Since there is no method to track neutrally stable point, the point should be collected from scanning over ranges of wavenumber and a flow parameter. When the neutral curve is perpendicular to the flow rate ratio axis, a wavenumber continuation is the proper method to collect neutrally stable point. Flow rate continuation is used otherwise.

When the interfacial tension vanishes ($\sigma_I = 0$), there is always a range of flow rate at which the flow is unstable. However the results at high wavenumber should be analyzed with care. At $\alpha^* \gtrsim 10^2$, the wave length of the disturbance is very small, less than a micron, in the cases analyzed here. This length may be comparable to the characteristic length unit of interlayer diffusion that was neglected in our model. Therefore, without including diffusion, it is hard to determine whether the interlayer is stable or not.

When interfacial tension does not vanish, e.g. $\sigma_I = 1$ dyne/cm ($N_T = 0.0435$), the tension damps out the high wavenumber disturbance: perturbations at wavenumber $\alpha^*$ above 4.28 are always stable. In this case, flow rate ratio above $f > 0.205$ guarantees linearly stable interlayer.

In this study, we determined neutral curves as a function of viscosity ratio and flow rate ratio to determine the desirable operating condition range.

5.3.2 Finding stable flow rate ratio range

Figure 5.7 shows the neutral curves for high viscosity ratio $m = 2$ with or without interfacial tension at different total wet thickness $h_{w,i}$. As shown in Fig. 5.7(b), the interfacial tension damped-out high wavenumber disturbances. However, stability characteristics of both flows, with and without interfacial tension, are the same from low to medium wavenumber range.

According to the predictions, large flow rate ratio leads to a stable interlayer at $m = 2$. The reason is that the less viscous layer 2 becomes thin at large flow rate ratio $f = q_1/q_2$, and the thin-layer effect stabilizes interlayer disturbance as discussed in the literature. The range of flow rate ratio for stable interlayer increases as total
Figure 5.7: Neutral curve for density ratio $r = 1$ and viscosity ratio $m = 2$ with interfacial tension (a) or without interfacial tension (b) for different total wet thickness, $h_{w,t} = 0.34$, 0.40 and 0.50.
wet thickness $h_{w,t}^* = h_{w,t}/H_g$ decrease, i.e. thinner coating is favorable in terms of the interlayer stability.

Figure 5.8 shows the neutral curve at viscosity ratio $m = 0.5$ at different total wet thickness. Now, layer 1 is less viscous than layer 2. At low and intermediate wavenumbers, the stable region is now restricted to low flow rate ratio. This is easily explained by the fact that the flow is stable when the low viscosity layer is thin (Joseph and Renardy, 1993). The stable region shrinks as the total wet thickness $h_{w,t}^*$ falls. Therefore, in this case, thick coating favorable for interlayer stability.

### 5.3.3 Critical flow rate ratio condition for mid-gap invasion

Interlayer instability is not the only source of coating defects. As discussed by Nam and Carvalho (2009b), if the wet thickness of the bottom layer (layer 2) is less than approximately one third of the coating gap the separation point travels through the mid lip leading to coating defects. Since dual slot coating method is known as a pre-metered method, the thickness of the deposited liquid layer is set by the flow rate and the web speed: $h_{w,l} = q_l/U_w$, where $l = 1$ or 2. The total wet thickness $h_{w,t} = h_{w,1} + h_{w,2}$ is usually set by product specifications, therefore the critical flow rate ratio for the mid-gap invasion can be computed from the results presented by Nam and Carvalho (2009b):

$$f_m = \frac{q_1}{q_2} = \frac{h_{w,1}}{h_{w,2}} = \frac{3}{H_g} \frac{h_{w,t} - H_g}{H_g} = \frac{3}{H_g} \frac{q_t - H_g U_w}{H_g U_w} = 3 h_{w,t}^* - 1.$$  \hspace{1cm} (5.38)

Therefore, coating defects can be present when the flow rate ratio $f$ is greater than the critical value $f_m$. Note that in this simple criterion, the critical condition does not depend on the material property like viscosity and density. This condition should be considered together with the linear stability analysis results for finding desirable operating conditions.

The predictions at high viscosity ratio ($m = 2$), Fig. 5.7, show that the stable range of flow rate coincides with the range of flow rate at which mid-gap invasion occurs. Therefore, operation at high viscosity is not recommended when both interlayer stability and mid-gap invasion are considered simultaneously. At low viscosity ratio
Figure 5.8: Neutral curve for density ratio $r = 1$ and viscosity ratio $m = 0.5$ with interfacial tension (a) or without interfacial tension (b) for different total wet thickness, $h_{w,t} = 0.34$, 0.40 and 0.50.
(\(m = 0.5\)), Fig. 5.8, the stable range of flow rate is free of mid-gap invasion. Consequently, operations with the top layer less viscous than the bottom layer is recommended to prevent coating defects coming from interlayer instability and mid-gap invasion.

### 5.3.4 Finding desirable viscosity ratio range

According to the results from section 5.3.2, low viscosity ratio, i.e. a less viscous top layer, is favorable for interlayer stability. However, there are limits on the range of viscosity ratio that stabilizes the flow. Figure 5.9(a) shows the neutral stability curve in the plane of wavenumber and viscosity ratio at a fixed flow rate. Note that, for the chosen flow rate ratio, \(f = 0.04\), there is no mid-lip vortex and turn-around flow inside mid-gap region: no mid-gap invasion. Considering low and intermediate wavenumbers, the interlayer is linearly stable only from \(m = 0.321\) to 1.00. The flow is unstable if top layer is more viscous (\(m > 1\)) or if its viscosity is much smaller than the bottom layer (\(m < 0.321\)). Again, stability of high wavenumber disturbance may not be predicted without diffusion for vanishing interfacial tension case, and the small wavelength disturbance are always faded away by the action of interfacial tension (region S*).

The existence of a range of viscosity ratio at which the flow is stable can be explained by the fact that although the flow rate ratio and total flow rate are fixed, the location of interlayer is not constant, but it is a function of viscosity ratio. Figure 5.9(b) displays the layer 2 thickness inside the downstream coating gap region as the viscosity ratio increases. The results are based on solving Eqs. (5.36) and (5.37) at given flow rates. The less viscous layer is thin inside the range of \(m = 0.321\) to 1.00. Note that, in this range, the less viscous layer is layer 1 (the top layer). When the viscosity ratio is less than 0.321, the less viscous layer 1 is not thin, because of turn-around flow, as shown in Fig. 5.9(c). When the viscosity ratio is greater than one, the less viscous layer 2 is always thicker than layer 1, as shown in Fig. 5.9(e). The results clearly show that only a limited viscosity ratio range enables the thin-layer effect that leads to stable flow.
5.3. RESULTS & DISCUSSION

Figure 5.9: Neutral curve for density ratio \( r = 1 \), flow rate ratio \( f = 0.04 \), and total wet thickness \( h^*_{w,t} = 0.4 \) with or without interfacial tension. “S” and “U” stand for stable and unstable interlayer, respectively. “S*” inside plot (a) stands for stable interlayer only when interfacial tension is presented. Plot (b) shows the interlayer location. Plots (c), (d), and (e) show the base flow profiles at \( m = 0.1, 0.6 \) and 1, respectively.
The viscosity range for stable flow is a function of flow rate ratio. Figure 5.10 shows the neutral curves for different flow rate ratios at total wet thickness $h_{w,t}^* = 0.40$ and immiscible liquid ($\sigma_l = 0$). Neutral stability curves in plot (a) of Fig. 5.10 shows that the stable regimes appear at moderately low viscosity ratio $m$ except at $f = 0.2$. As the flow rate ratio increases, the neutral stability curves at low and moderate wavenumber, $\alpha^* = 10^{-2} \sim 10^1$, shift toward the high viscosity ratio. At high flow rate ratios, plot (b), the stable window occurs at a viscosity ratio range above $m = 1$.

Not all stable regimes in Fig. 5.10 are desirable in a point of view of coating operations. When the location of the interlayer separation point is not located at the downstream corner of the mid lip (mid-gap invasion), either micro-vortex under the mid lip or periodic oscillation of the interlayer can cause coating defects or degrade product quality (Nam and Carvalho, 2009b). At the total thickness $h_{w,t}^* = 0.40$, the critical flow rate ratio for mid-gap invasion is $f_m = 3 \times 0.40 - 1 = 0.20$. Therefore, plot (a) shows neutral curves for low flow rate ratio and they are free from mid-gap invasion. On the contrary, flow rate ratios for plot (b) implies that, even though the coating flow are linearly stable at high viscosity ratio, mid-gap invasion can cause problems in production.

5.4 Conclusion

Linear stability analysis was carried out on the two-layer flow in the downstream coating gap of a dual slot coating system. The problem is written in terms of the flow rate ratio, instead of the usual thickness ratio. The original generalized eigenvalue problem was effectively reduced to a smaller size simple eigenproblem by extending the method proposed by Valerio et al. (2007). The most dangerous disturbance related to the largest real part of eigenvalue or growth rate was found to be concentrated at the interlayer. The waviness of the interlayer caused by the disturbance can spoil the quality of two-layer coating product. Therefore it is important to find parameter ranges at which the flow is a stable to prevent coating defects.

When the neutrally stable points, where the growth rate changes sign, are computed as a function of wavenumber and flow parameters, a neutral curve can be constructed
5.4. CONCLUSION

![Figure 5.10:](image)

Figure 5.10: Neutral curve for density ratio \( r = 1 \) and total wet thickness \( h_{w,t}^* = 0.40 \) for different flow rate ratio with or without interfacial tension. Note that when the viscosity for both layers are the same \( (m = 1) \), interlayer is neutrally stable because there are no distinction between layer 1 and layer 2. Plot (a) displays \( f = 0.20, 0.04 \) and \( 0.02 \), and plot (b) displays \( f = 0.50, 1.00 \) and \( 2.00 \). For plot (a), “\( S \)” is stable for \( f = 0.02 \) and \( 0.04 \), “\( S^0 \)” is stable for \( f = 0.04 \), and “\( S^+ \)” is stable for \( f = 0.20 \). For plot (b), “\( S \)” is stable for \( f = 0.50, 1.00 \) and \( 2.00 \), “\( S^+ \)” is stable for \( f = 1.00 \) and \( 2.00 \), and “\( S^0 \)” is stable for \( f = 2.00 \). “\( S^* \)” inside plots (a) and (b) stands for stable interlayer only when interfacial tension is presented.
5.4. CONCLUSION

In order to mark stable and unstable regions in the parameter space. As pointed out by Joseph and Renardy (1993), the long wave (large wavenumber) disturbance can be suppressed by utilizing thin-layer effect, i.e. making the less viscous liquid layer thin, while the short wave (small wavenumber) disturbance is damped out by the action of interfacial tension. When miscible coating liquids are considered (vanishing interfacial tension), the stability of short wave disturbance is hard to determine without including diffusion in the model.

Besides the interlayer instability, the location of the interlayer separation point can cause coating defects (Nam and Carvalho, 2009b). The critical mid-gap invasion condition can be re-written in terms of flow rate ratio at the fixed total flow rate, or total wet thickness.

Combining the range of operating parameters for stable two-layer flow and pinned separation point (no mid-gap invasion), a defect free region can be created. The predictions show that the recommended conditions are at low flow rate ratio $f = q_1/q_2$ and low viscosity ratio $m = \mu_1/\mu_2$, i.e. a thin top layer of a low viscosity liquid. These condition exploit the thin layer effect without invoking mid-gap invasion. Because the interlayer location in the downstream coating gap is a function of the viscosity ratio at a fixed flow rate, only a small window of viscosity ratio leads to stable flows. The defect free region is a function of the total flow rate; it shrinks as the wet thickness falls.
Chapter 6

Computational analysis of single-layer tensioned-web-over-slot die coating flow

6.1 Introduction

Liquid coating process is the main step in the manufacturing of adhesive and magnetic tapes, specialty papers, optical films and many other products. When a precisely controlled thin layer at relatively high speed is required, one of the preferred coating method is the conventional fixed-gap slot die coating, in which the substrate is supported by a rigid back-up roll in order to control the distance between the die surface and the substrate (Sartor 1990).

However, the range of attainable coating thickness is limited in conventional slot coating method. The competition among viscous, capillary and pressure forces sets the range of operating parameters (coating window) in which the viscous free surface flow can be two dimensional and steady, which is the desired state. At a given set of operating conditions, there is a critical wet thickness below which the deposited layer is not uniform. Several patents, as for example Tanaka and Noda (1984), claim that
6.1. INTRODUCTION

at a coating rate of \(100 \text{ m/min}\) or more, it is difficult to produce an uniform coating layer thinner than \(20 \mu\text{m}\) cannot be obtained with conventional slot coating. The coating window of fixed gap slot coating is bounded by different modes of failures, as discussed by Romero et al. (2004). The minimum possible wet thickness can be related to the invasion of the upstream meniscus towards the feed slot, if the pressure difference under the upstream die lip is not strong enough to balance the viscous drag force, or to the invasion of the downstream meniscus towards the coating bead, if the meniscus needs to curve so much, to create an adverse pressure gradient, that it cannot bridge the gap. Both failure modes lead to three dimensional flow states and the coating bead takes a form that deliver separate rivulets to the moving web. The minimum wet thickness is a direct function of the coating gap. Therefore, extremely thin films can only be obtained by adjusting the gap to be extremely small, which cannot be used in practice. The minimum gap used in a real industrial application is in the order of \(100 \mu\text{m}\).

In order to operate at extremely small gaps without the risk of clashing the roll and the coating die, a flexible support of the substrate proves to be an useful idea. One way of having a self regulating gap is to wrap the web under tension around

\[\text{Figure 6.1: A schematic diagram of tensioned-web-over-slot die coating system.}\]
the coating die, as sketch in Fig. 6.1. Such system is known as tensioned-web-over-slot die (TWOSD) coating method. The competition between the hydrodynamic force due to the liquid flow and the normal force resultant from the curved substrate under tension is the essential ingredient of the elastohydrodynamic interaction that regulates the gap height between the die surface and the moving substrate. A major advantage over conventional slot coating is the simplicity of the overall setup: there is no precision back-up roll, nor is there a need for a vacuum system to stabilize the coating bead, since the pressure in the coating bead is above atmosphere. In general, the method can maintain extremely thin gap height (down to the order of a micron) that is crucial to coat a ultra-thin layer of liquid, without scratching die surfaces. However, two-dimensional flow can only be obtained if the tension along the width of the web is uniform. Substrates that are not perfectly flat are usually not suitable for this coating method. Moreover, as we discuss here, the geometry of the die has a strong effect on the window of the process.

As in any other coating method, it is important also to map the region in the operating parameter space at which the flow is two dimensional, except near the edges, and steady. This region is generally called the operating window of the process. Because the presence of recirculations inside the coating bead can be a source of coating defects, it is important to map the region inside the operating window at which vortices are not present in the flow, called here the vortex-free operating window.

The fundamental understanding of the flow in TWOSD coating is limited. Feng (1998) presented flow states at different conditions by solving two-dimensional Navier–Stokes equation for free surface flow by means of Galerkin finite element method. The web was described using membrane theory (Flügge 1973) and the tension variation along the length of the substrate was neglected. These approximations are valid only at a limited parameter range. It cannot capture the accurate liquid/web interaction when the curvature of web changes rapidly, that occurs when complex die lip geometry is used. The analysis did not include the construction of coating windows nor the effect of the geometry on the process limits.

Park (2008) used an elasto-visco-capillary model to describe the flow. He showed that the die geometry has a strong effect on the operating window. This $1-D$ model is
not able to describe the presence of micro recirculation in the coating bead, that lead to several coating effects.

Lin et al. (2008) took a different approach. They used lubrication approximation to describe the liquid flow and a simplified membrane theory for the web. The web configuration is only computed at the exit of the feed slot by balancing the normal force of the curved web to the liquid traction. Under the die lips, the web location is approximated simply by using geometric arguments. The downstream meniscus location is based on the Landau and Levich (1942) equation, which is only valid at low capillary number. Even with all the approximations made on the development of the model, the predictions show the same trends observed in the experiments by Lin et al. (2007). All these analysis were based on straight die lip design, as proposed by Tanaka and Noda (1984).

Here, we present a single-layer TWOSD flow model by coupling two-dimensional Navier–Stokes theory and thin inextensible shell theory with linear elastic models (Flügge 1973), based on the method proposed by Carvalho (2003). With aid of a powerful direct tracking of flow feature technique (Nam et al. 2009), we constructed the operating window and the vortex-free operating window automatically. We use the model to determine the effect of die lip design on the critical parameters of the process. Because several patents (Takahashi and Shibata, 1995; Shibata and Chino, 1993; Okuno and Kawabe, 1994) have shown that rounded lips present some advantages, we focus our analysis on rounded die lips.

6.2 Elastohydrodynamic model and solution method

6.2.1 Governing equations and boundary conditions

The flexible substrate is assumed to be infinitely wide, and therefore the flow in the transverse direction is neglected. The motion of the coating liquid is described by the Navier–Stokes equation and continuity equation for incompressible Newtonian fluid:

\[ \rho \mathbf{u} \cdot \nabla \mathbf{u} = \nabla \cdot \mathbf{T}, \quad \nabla \cdot \mathbf{u} = 0, \quad (6.1) \]
6.2. ELASTOHYDRODYNAMIC MODEL AND SOLUTION METHOD

\[ n \cdot \nabla u = 0 \]
\[ n \cdot u = 0 \]
\[ n \cdot n_w = \cos \theta_s \]
\[ n \cdot n_w = \cos \theta_d \]
\[ u = f(x, h_w, U_w) \]
\[ u = t_w U_w \]
\[ u = 0 \]

**Figure 6.2:** Tensioned-web-over-slot die coating model boundary conditions and mesh scheme. Mesh used to solve the governing equations by G/FEM 558 elements and 10318 unknown coefficients of the finite element basis functions are used to discretize the system.

\[ p \] is the liquid density and \( T \) is stress tensor. For Newtonian liquid, it is given by \( T = -p I + \mu \left[ \nabla u + (\nabla u)^T \right] \), where \( p \) is the pressure and \( \mu \) is the liquid viscosity.

Boundary conditions are need to solve the Navier–Stokes system. In tensioned-web-over-slot die coating flow, the domain is bounded by inflow and outflow planes, solid walls, gas-liquid interfaces and the flexible web that deforms according to elastohydrodynamic interaction, as shown in Fig. 6.2. At the feed slot inlet, we imposed

\[ u = U_0(x), \quad (6.2) \]
6.2. ELASTOHYDRODYNAMIC MODEL AND SOLUTION METHOD

where $U_b(x)$ is the imposed parabolic velocity profiles. At the outflow, we impose

$$n_b \cdot \nabla u = 0, \quad (6.3)$$
$$p = p_{amb}. \quad (6.4)$$

The wet thickness $h_w$ is determined by the flow rate and the web velocity $U_w$. $p_{amb}$ is the imposed outflow pressure (ambient pressure), and $n_b$ is the unit normal vector to the boundary.

At rigid solid walls, the no-slip and no-penetration conditions were imposed. Along the die surface,

$$t_w \cdot u = 0, \quad n_w \cdot u = 0. \quad (6.5)$$

Along the web,

$$t_w \cdot u = U_w, \quad n_w \cdot u = 0, \quad (6.6)$$

where $U_w$ is the solid wall velocity, $n_w$ and $t_w$ are the unit normal and tangent vectors to the wall. At the dynamic contact line, where the upstream meniscus meets the moving web, Navier slip condition was used instead of the no slip condition,

$$\frac{1}{\beta} t_w \cdot (u - U_w t_w) = t_w n_w : T, \quad (6.7)$$

where $\beta$ is the slip coefficient. Here we choose $\beta = 0.1 g^{-1} sec^{-1}$, based on the numerical tests reported by Sartor (1990).

Along the gas-liquid interfaces, a force balance and the no-penetration condition (kinematic condition) were imposed:

$$n_f \cdot T_i = \sigma \frac{dt_f}{ds} - n_f P_{amb}, \quad (6.8)$$
$$n_f \cdot u = 0, \quad (6.9)$$

where $t_f$ and $n_f$ are the local unit tangent and unit normal to the free surface, $s$ is the arc-length coordinate along the interface, $\sigma$ is the liquid surface tension and $P_{amb}$ is the ambient pressure.

Like the gas-liquid interfaces, the position of the flexible web is unknown a priori. Here, we model the deformation and location of the moving substrate using the equations of cylindrical shells [Flügge 1973]. In sum, linear and angular momentum
balances for the thin cylindrical shell boils down to tangential and normal force balance equations, and one more equation to define the curvature in terms of the web configuration.

\[
\begin{align*}
\frac{dT}{d\xi} + \kappa \frac{d}{d\xi} (\kappa D) + P_t &= 0 \\
-\frac{d^2}{d\xi^2} (\kappa D) + \kappa T + P_n &= 0 \\
\frac{d^2 x}{d\xi^2} + \kappa \frac{dy}{d\xi} &= 0 \text{ or } \frac{d^2 y}{d\xi^2} - \kappa \frac{dx}{d\xi} = 0
\end{align*}
\]

\[
(6.10)
\]

\(\xi\) is the coordinate along the web. \(T\) and \(\kappa\) are the web tension and curvature at each position, and \(x\) and \(y\) are the Cartesian coordinates of points on the web. The web stiffness \(D \equiv \frac{E t^3}{12(1-\nu^2)}\) is a function of the elastic modulus \(E\), Poisson ratio \(\nu\), and thickness of the web \(t\). \(P_t\) and \(P_n\) are the loading forces on the web in the tangential and normal direction. Those forces are responsible for the web deformation and can be obtained from the traction exerted by the fluid neighboring the web:

\[
P_t = -t_w \cdot (n_w \cdot T) \quad \text{and} \quad P_n = -n_w \cdot (n_w \cdot T),
\]

\[
(6.11)
\]

where \(t_w\) and \(n_w\) are tangent and normal vectors on the flexible moving web.

The shell equations require two corner conditions for position and curvature, and one condition for tension. In our model, corner conditions for the shell equations are

\[
\begin{align*}
\mathbf{x} &= \mathbf{x}_d, \quad \kappa = 0 \quad &\text{for downstream web end,} \\
\mathbf{x} &= \mathbf{x}_u, \quad \kappa = 0, \quad T = T_s \quad &\text{for upstream web end}
\end{align*}
\]

\[
(6.12)
\]

Figure 6.3 shows a graphical representation of the important parameters of the model, including liquid properties, die lip geometry (lip lengths and radius of curvature), and operating parameters, wrap angles, web tension and web speed. The operating parameters and geometric parameters and their values at the base case analyzed here are summarized in Table 6.1. The relevant variables can be combined into the following dimensionless groups:

- Tension number \(N_T \equiv \frac{\mu U_w}{T}\),
Table 6.1: Operating and geometric parameters for base case die lip (BC) configuration.

<table>
<thead>
<tr>
<th>Operating parameters</th>
<th>Name</th>
<th>Unit</th>
<th>Base case</th>
</tr>
</thead>
<tbody>
<tr>
<td>Film thickness($h_w$)</td>
<td>$\mu$m</td>
<td></td>
<td>40</td>
</tr>
<tr>
<td>Web speed($U_w$)</td>
<td>m/sec</td>
<td></td>
<td>1</td>
</tr>
<tr>
<td>Ambient pressure ($P_{amb}$)</td>
<td>Pa</td>
<td></td>
<td>0</td>
</tr>
<tr>
<td>Surface tension ($\sigma$)</td>
<td>dyne/cm</td>
<td></td>
<td>61</td>
</tr>
<tr>
<td>Web tension ($T$)</td>
<td>N/m</td>
<td></td>
<td>175.1</td>
</tr>
<tr>
<td>Density($\rho_1 = \rho_2$)</td>
<td>g/cm$^3$</td>
<td></td>
<td>1.2</td>
</tr>
<tr>
<td>Viscosity($\mu_1$ or $\mu_2$)</td>
<td>cP</td>
<td></td>
<td>23</td>
</tr>
<tr>
<td>Young’s modulus ($E$)</td>
<td>MPa</td>
<td></td>
<td>8</td>
</tr>
<tr>
<td>Web thickness ($t$)</td>
<td>$\mu$m</td>
<td></td>
<td>6</td>
</tr>
<tr>
<td>Dynamic contact angle($\theta_d$)</td>
<td>deg.</td>
<td></td>
<td>140</td>
</tr>
<tr>
<td>Static contact angle($\theta_s$)</td>
<td>deg.</td>
<td></td>
<td>60</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Geometric parameters (See Fig. 6.3)</th>
<th>Name</th>
<th>Unit</th>
<th>Base case</th>
</tr>
</thead>
<tbody>
<tr>
<td>Upstream lip length($L_u$)</td>
<td>mm</td>
<td></td>
<td>1.1</td>
</tr>
<tr>
<td>Downstream lip length($L_d$)</td>
<td>mm</td>
<td></td>
<td>1.1</td>
</tr>
<tr>
<td>Feed slot height($H_f$)</td>
<td>$\mu$m</td>
<td></td>
<td>100</td>
</tr>
<tr>
<td>Upstream lip radius of curvature ($R_u$)</td>
<td>in</td>
<td></td>
<td>0.250</td>
</tr>
<tr>
<td>Downstream lip radius of curvature ($R_d$)</td>
<td>in</td>
<td></td>
<td>0.250</td>
</tr>
<tr>
<td>Upstream die lip approaching angle ($\theta_{L,u}$)</td>
<td>deg.</td>
<td></td>
<td>9.97</td>
</tr>
<tr>
<td>Downstream die lip departure angle ($\theta_{L,d}$)</td>
<td>deg.</td>
<td></td>
<td>9.97</td>
</tr>
<tr>
<td>Upstream web wrap angle ($\theta_{w,u}$)</td>
<td>deg.</td>
<td></td>
<td>7.40</td>
</tr>
<tr>
<td>Downstream web wrap angle ($\theta_{w,d}$)</td>
<td>deg.</td>
<td></td>
<td>8.97</td>
</tr>
</tbody>
</table>
6.2. ELASTOHYDRODYNAMIC MODEL AND SOLUTION METHOD

Figure 6.3: Tensioned-web-over-slot-die coating model parameters. \( L_d \) and \( R_d \) are downstream die lip length and radius of curvature. \( L_u \) and \( R_u \) are upstream die lip length and radius of curvature. \( H_f \) is feed slot height. \( \theta_{w,d} \) and \( \theta_{w,u} \) are downstream and upstream web wrap angle. \( \theta_{L,u} \) and \( \theta_{L,d} \) are upstream lip approaching angle and downstream lip departure angle. \( \theta_s \) and \( \theta_d \) are static contact angle and dynamic contact angle of coating liquid. \( T \) and \( U_w \) are tension and web speed, respectively.
6.2. ELASTOHYDRODYNAMIC MODEL AND SOLUTION METHOD

- Dimensionless wet thickness $h^*_w = \frac{h_w}{R_d}$
- Reynolds number $N_{Re} \equiv \frac{\rho h_w U_w}{\mu}$
- Capillary number $N_{Ca} \equiv \frac{\mu U_w}{T}$
- Elasticity number $N_{Es} \equiv \frac{D}{T R_d^2}$

plus the parameters associated with the geometry of the die (dimensionless radius of curvature of the lip and wrapping angles) Note that $D = E t^3 / 12 (1 - \nu^2)$ is flexural rigidity. We assume that web is incompressible, $\nu = 1/2$, has $6 \mu\text{i}n$. thickness; and is made out of polyethylene naphthalate (PEN), i.e. Young’s modulus is $E = 8\text{MPa}$.

The governing equations, Eq. (6.1) to (6.12), give rise to a free boundary problem. The locations of the flexible substrate and the gas/liquid interfaces are unknown a priori and they are parts of the solution. Kistler and Scriven (1984) explained the basis of solving viscous free surface problems. The method of coupling viscous free surface flow and flexible moving substrate used here was in detail discussed by Carvalho (2003). The basis of the solution method in Galerkin finite element framework is recounted briefly here, but the computational details on the liquid/web interaction will be emphasized.

To solve a free boundary problem by means of standard techniques for boundary value problems, the set of differential equations and boundary conditions posed in the unknown physical domain have to be transformed to an equivalent set defined in a known, fixed computational domain. This transformation is made by mapping $x = x(\xi)$ that connects two domains. The physical domain is parameterized by the position vector $x = (x, y)$, and the reference domain, by $\xi = (\xi, \eta)$. The mapping used here is the one described by de Santos (1991). The inverse mapping is governed by a system of elliptic differential equations identical to those encountered in the dilute regime of diffusional transport.

$$\nabla \cdot D_\xi(\xi, \eta) \nabla \xi = 0, \nabla \cdot D_\eta(\xi, \eta) \nabla \eta = 0. \quad (6.13)$$
6.2. ELASTOHYDRODYNAMIC MODEL AND SOLUTION METHOD

$D_{\xi}$ and $D_{\eta}$ are mesh diffusivities which control the steepness of gradients in the node-spacing by adjusting the potentials $\xi$ and $\eta$. Curves of constant $\xi$ and $\eta$ define the boundaries of elements used to describe the domain. The cross point of these curves sets the position of a node. Boundary conditions are needed to solve the second-order differential equations (6.13). Solid walls and inflow and outflow planes are described by the function that defines their geometry and nodes were distributed along them by a specified hyperbolic tangent type stretching function $[\text{Vinokur} 1983a]$. The location of gas-liquid interface is implicitly determined by the kinematic condition, Eq. (6.9), and the location of the flexible substrate is implicitly determined by the thin cylindrical shell equations, Eqs. (6.10). The discrete version of the mapping equations is generally referred to as mesh generation equations. Detailed procedure and boundary conditions used for mesh equation are discussed in $[\text{de Santos} 1991]$.

6.2.2 Solution of the Navier-Stokes / thin inextensible cylindrical shell system for free surface flow by G/FEM

The Navier–Stokes equation (6.1), the substrate deformation (6.10) and the mesh generation equations (6.13) together with corresponding boundary conditions were solved by the Galerkin’s method with quadrilateral and one-dimensional finite elements. For the two-dimensional liquid domain, velocity and position were approximated by continuous biquadratic basis functions $\phi^i$, and pressure field was represented using piecewise linear basis function $\psi^k$. For the one dimensional shell elements, velocity and nodal position values are shared with the neighboring liquid element, and tension and curvature are approximated by continuous quadratic basis functions $\varphi^j$. Therefore, the unknown fields were written in terms of their basis functions as:

\[
\begin{align*}
\mathbf{u} &= \sum_i U_i \phi^i, \quad p = \sum_k P_k \psi^k, \quad \mathbf{x} = \sum_i X_i \phi^i, \\
T &= \sum_j T_j \varphi^j, \quad \kappa = \sum_j \kappa_j \varphi^j,
\end{align*}
\]

(6.14)

where $U_i$, $P_k$, $X_i$, $T_j$, and $\kappa_j$ are the basis function coefficients, the unknowns of the discretized problem.

The weak form of Eqs. (6.1) and (6.13) were obtained by multiplying each equation
by weighting functions, integrating over the physical domain, and applying the divergence theorem to the appropriate terms. In Galerkin’s method, the weighting and basis functions are the same. The weighted residuals are:

\[
R^i_m = \int_A \phi^i \rho \mathbf{u} \cdot \nabla \mathbf{u} + \mathbf{T} \cdot \nabla \phi^i \, dA - \int_{\Gamma} \phi^i \mathbf{n} \cdot \mathbf{T} \, ds, \tag{6.15}
\]

\[
R^i_c = \int_A \psi^k \nabla \cdot \mathbf{u} \, dA, \tag{6.16}
\]

\[
R^i_x = \int_A D_{\xi} \nabla \xi \cdot \nabla \phi^i \, dA + \int_{\Gamma} \phi^i D_{\xi} \mathbf{n} \cdot \nabla \xi \, ds, \tag{6.17}
\]

\[
R^i_y = \int_A D_{\eta} \nabla \eta \cdot \nabla \phi^i \, dA + \int_{\Gamma} \phi^i D_{\eta} \mathbf{n} \cdot \nabla \eta \, ds,
\]

where \( A \) is the flow domain, and \( \Gamma \) is the domain boundary and \( s \) is arc-length coordinate along the boundary. The superscripts, \( m \), \( c \) and \( x \), denote momentum, continuity and mesh residuals, respectively. Essential boundary conditions were imposed by replacing the corresponding weighted residual equation with the desired velocity or node specification. Natural boundary conditions were applied through the boundary integrals in (6.15) and (6.17).

Along the gas-liquid interface, one of the mesh equation residual was replaced by the kinematic condition. The corresponding weighted residual is

\[
R^i_x \text{ or } R^i_y = \int_{\Gamma} (\mathbf{n} \cdot \mathbf{u}) \phi^i \, ds \tag{6.18}
\]

Also, the boundary integrals in the weighted residual of the momentum equation that comes from the divergence theorem become

\[
\int_{\Gamma} \phi^i \mathbf{n} \cdot \mathbf{T} \, ds = - \int_{\Gamma} \phi^i P_{\alpha} \mathbf{n} + \sigma \mathbf{t} \frac{d \phi^i}{ds} \, ds + \sigma \phi^i \mathbf{t} \bigg|^{s_e}_{s_s}, \tag{6.19}
\]

where \( s_s \) and \( s_e \) are the arc-length coordinate values for both extremes of the gas-liquid interface.
Along the liquid/web boundary, the mesh equations were replaced by the thin cylindrical shell equations (6.10). After applying integral by parts on the second derivative terms, the residual equations become:

\[ R^i_T = \int_{\Gamma_w} \varphi^i \left( \frac{dT}{ds} + \kappa \frac{d}{ds}(\kappa D) \right) ds + \int_{\Gamma_w} \varphi^i P_t ds, \]  

\[ R^i_\kappa = \int_{\Gamma_w} \left( D \frac{d \varphi^i}{ds} \frac{d \kappa}{ds} + \varphi^i \kappa T \right) ds + \int_{\Gamma_w} \varphi^i P_n ds, \]  

\[ R^i_x \text{ or } R^i_y = \int_{\Gamma_w} \left( -\frac{d \varphi^i}{ds} \frac{dx}{ds} + \kappa \varphi^i \frac{dy}{ds} \right) ds \]  

or \[ \int_{\Gamma_w} \left( -\frac{d \varphi^i}{ds} \frac{dy}{ds} - \kappa \varphi^i \frac{dx}{ds} \right) ds. \]

The boundary terms of \( R^i_T \) and \( R^i_\kappa \) that come from the integral by parts need not to be evaluated, because positions, curvature, and tension at both extremes of the web are specified as essential boundary conditions. Note that \( R^i_T \) of Eq. (6.22) is used when the moving web is mostly aligned to \( y \) coordinate direction, and \( R^i_\kappa \) for \( x \) direction. The other residual associated with nodal position along the web specifies the node distribution — hyperbolic stretching function, in this study.

The loading forces in Eqs. (6.20) and (6.21) are computed from the traction vector \( n_w \cdot T \) at the liquid/web boundary \( \Gamma_w \). Therefore, it is computationally convenient to construct part of residual equations \( R^i_{T,web} \) and \( R^i_{k,web} \) at the shell elements; and the remaining part \( R^i_{T,liq} \) and \( R^i_{k,liq} \) at the two-dimensional liquid elements.

Furthermore, biquadratic basis function \( \varphi^i \) becomes identical to quadratic basis func-
tion \( \varphi^i \) at the boundary \( \Gamma_w \). Therefore \( R_{T,liq}^i \) and \( R_{k,liq}^i \) can be expressed as

\[
R_{T,liq}^i = \int_{\Gamma_w} \varphi^i \left[ t_w \cdot (n_w \cdot T) \right] ds
\]

\[
= \int_{\Gamma_w} \varphi^i \left\{ t_{w,x} \left[ -p n_{w,x} + 2 n_{w,x} \frac{\partial u}{\partial x} + \left( n_{w,y} \frac{\partial u}{\partial y} + n_{w,x} \frac{\partial v}{\partial x} \right) \right] 
+ t_{w,y} \left[ -p n_{w,y} + 2 n_{w,y} \frac{\partial v}{\partial y} + \left( n_{w,y} \frac{\partial u}{\partial y} + n_{w,x} \frac{\partial v}{\partial x} \right) \right] \right\} ds
\]

\[
R_{k,liq}^i = \int_{\Gamma_w} \varphi^i \left[ n_w \cdot (n_w \cdot T) \right] ds
\]

\[
= \int_{\Gamma_w} \varphi^i \left\{ n_{w,x} \left[ -p n_{w,x} + 2 n_{w,x} \frac{\partial u}{\partial x} + \left( n_{w,y} \frac{\partial u}{\partial y} + n_{w,x} \frac{\partial v}{\partial x} \right) \right] 
+ n_{w,y} \left[ -p n_{w,y} + 2 n_{w,y} \frac{\partial v}{\partial y} + \left( n_{w,y} \frac{\partial u}{\partial y} + n_{w,x} \frac{\partial v}{\partial x} \right) \right] \right\} ds,
\] (6.23)

where normal and tangent vectors at the web are \( n_w = i n_{w,x} + j n_{w,y} \) and \( t_w = i t_{w,x} + j t_{w,y} \).

Along the shell elements not neighboring a two-dimensional liquid element, i.e. upstream of the contact line, the tension, curvature and position of the substrate are determined in the same way, without the terms corresponding to the liquid traction.

The \( x \) and \( y \) momentum weighted residuals along the substrate are replaced by the no-slip/no-penetration condition:

\[
R_{m_x}^i = \int_{\Gamma_w} \varphi^i n \cdot v ds
\]

\[
R_{m_y}^i = \int_{\Gamma_w} \varphi^i (t \cdot v - U_w) ds.
\] (6.24)

In sum, the Galerkin finite element method (G/FEM) reduces the Navier-Stokes, mesh generation and thin cylindrical shell differential equations to a set of nonlinear algebraic equations on the basis function coefficients.

\[
R(z, \lambda) = 0,
\] (6.25)
where \( \mathbf{z} \) is the solution vector, that consists of the finite element coefficients for velocity \( \mathbf{u} \), pressure \( P \), position \( \mathbf{x} \), tension \( T \) and curvature \( \kappa \), and \( \lambda \) is a vector that contains the \( M \) parameters on which the system depends. The non-linear system of algebraic equations (6.25) is solved iteratively by Newton’s method:

\[
\mathbf{J}^{(i)}(\mathbf{z}^{(i)}, \lambda) \delta \mathbf{z}^{(i)} = -\mathbf{R}^{(i)}(\mathbf{z}^{(i)}, \lambda),
\]

\[
\mathbf{z}^{(i+1)} = \mathbf{z}^{(i)} + \delta \mathbf{z}^{(i)},
\]

the indices \( i \) and \( i+1 \) indicate the current and next Newton’s step. \( \mathbf{J}^{(i)} \equiv \partial \mathbf{R}^{(i)}/\partial \mathbf{z}^{(i)} \) is the Jacobian matrix. The iteration continues until \( ||\mathbf{R}^{(i)}||_2 < 10^{-8} \).

### 6.2.3 Direct tracking of flow feature

In this study, we identify four dangerous flow features that (possibly) lead to coating defects. The critical parameters at which these feature appear were automatically computed by a direct tracking method in order to construct operating windows in a very efficient way.

#### Limit flow states in tensioned-web-over-slot die coating flow

For a given die geometry and set up (wrapping angles) and at a fixed tension number \( N_T \equiv \mu U_w/T \), the flow in the coating bead is a function of the wet thickness. Figure 6.4 presents the evolution of the flow states as the wet thickness rises. The range of possible wet thickness \( h^*_w \) is bounded by two extremes: at low \( h^*_w \), the contact line invades the feed slot leading to cross web non-uniformities and ultimately to bead break-up. Moreover, a large recirculation is present at the exit of the feed slot, as illustrated in Fig. 6.4(a). As the wet thickness rises, the contact line moves upstream and the recirculation inside the feed slot disappears(Fig. 6.4(b)). On the other extreme, at high wet thickness, the contact line reaches the end of the die lip surface and liquid may drip, losing the pre-metered action of the method, a phenomenon referred as weeping. Depending on the die lip geometry, vortices may also appear under the upstream and downstream die lip, as shown in Fig. 6.4(c).
Figure 6.4: Change of flow states as wet thickness increase for base case (BC) configuration. See Fig. 6.7 and Table 6.2 for BC configuration details. Tension number is set to $N_T = 4.8 \times 10^{-4}$. Dimensionless wet thickness is defined to be $h^*_w = h_w / R_d$, where $h_w$ and $R_d$ are wet thickness and downstream lip radius, respectively. Note that the coating flow in (b) has desirable flow state.
6.2. ELASTOHYDRODYNAMIC MODEL AND SOLUTION METHOD

As mentioned before, it is important to map not only the parameters at which the contact line reaches both extremes of the upstream die lip, but also the conditions at which recirculations appear inside the flow. Vortices can lead to undesired effects on the final coated product, such as cross-web non-uniformities, due to a nodular structure of the vortex, or point defects, due to unwanted polymerization or particle aggregation because of the long residence time.

In order to find the critical parameters at which these undesired flow states occur computationally, we need to define mathematical conditions associated with the onset of these flow states. Weeping and bead breakup can be detected by stability analysis. However, they are also closely related to the location of meniscus. Since the flow is mostly aligned to the y-coordinate in our model (Fig. 6.2), the onset of conditions can be written as

\[ y_{SCL} = y_{uc,u} \] for weeping, \hspace{1cm} (6.27)
\[ y_{DCL} = y_{uc,f} \] for bead breakup, \hspace{1cm} (6.28)

where \(y_{SCL}, y_{DCL}, y_{uc,u}\) and \(y_{uc,f}\) are the y-coordinates of the upstream static contact line, dynamic contact line, the upstream corner of upstream die lip and the upstream corner of feed slot exit, respectively.

Both vortices in this coating flow detach from a solid wall — feed slot wall and die lip. This type of vortex birth occurs when the shear stress and its derivative with respect to the arc length coordinate along the wall vanish (for Newtonian liquid) as discussed by Nam et al. (2009):

\[ \tau_{tn}(s_t^*) = 0 \] and \[ \frac{\partial \tau_{tn}}{\partial s_t}(s_t^*) = 0, \] (6.29)

where \(\tau_{tn}\) is shear stress and \(s_t^*\) is the arc-length coordinate along the solid surface at the vortex birth point.

**Automatic tracking of limit flow states**

In order to construct the coating window in an efficient manner, a multiparameter continuation method can be used to track the flow state limits. Nam et al. (2009)
used an automatic tracking of vortex birth to create a vortex-free window by solving the Navier–Stokes system by G/FEM only at the conditions at which vortex appear in the flow. The savings compared to the determination of the window by post-processing a large number of solutions is enormous. In this study, we use the same method algorithm to track operating limits and vortex birth inside the flow in TWOSD coating.

The method of direct tracking discussed by Nam et al. (2009) is outlined here. The central point of the method is to solve an augmented system consisting of the original Navier–Stokes system for free surface flow (and tensioned web) plus the conditions that define the flow feature being tracked Eqs. (6.27), (6.28) and Eq. (6.29).

The augmented system of equation can be represented as

$$R^A(z^A) = R(z, \lambda, p) + A(z, \lambda, p) = 0,$$

where $z$ and $\lambda$ are the vectors that contain the finite element coefficients and the fixed flow parameters, as defined in Eq. (6.26), and $p$ is a vector that contains the extra set of unknowns of the augmented problem, i.e. the flow parameters that are not fixed. $A$ is the set of algebraic equations that defines the flow states being tracked.

Weeping or bead breakup requires the corner of the flow domain to be aligned at a certain position. Therefore Eqs. (6.27) and (6.28) can be easily re-written as an augmented equation in the finite element context:

$$A = Y_{i(SCL)} - y_{uc,u} = 0 \quad \text{for weeping,} \quad (6.30)$$

$$A = Y_{i(DCL)} - y_{uc,f} = 0 \quad \text{for bead breakup,} \quad (6.31)$$

where $i(SCL)$ and $i(DCL)$ are the node numbers of the finite element node assigned to the upstream static contact line and to the dynamic contact line, and $Y_{i(SCL)}$ and $Y_{i(DCL)}$ are the corresponding y-coordinate coefficients.

For vortex birth, it is difficult to use Eq. (6.29) as augment equations (Nam et al., 2009). This condition was replaced by an approximated condition is used:

$$A = \begin{bmatrix} \tau_{in}(s^*_t) \\ \tau_{in}(s^*_t + \delta s_t) \end{bmatrix} = 0, \quad (6.32)$$
where \( \delta s_t \) is the distance between the two distinct, but close, separation points.

Newton’s method is used to solve the augmented set of non-linear algebraic equations:

\[
\begin{bmatrix}
\frac{\partial R}{\partial z} & \frac{\partial R}{\partial p} \\
\frac{\partial A}{\partial z} & \frac{\partial A}{\partial p}
\end{bmatrix}
\begin{bmatrix}
\delta z \\
\delta p
\end{bmatrix}
= -\begin{bmatrix}
R_A \\
A
\end{bmatrix}
\]  

(6.33)

where \( \partial R / \partial p \) are the sensitivity of the residual equations to the extra unknowns, and \( \partial A / \partial z \) and \( \partial A / \partial p \) are the sensitivity of the limit flow state conditions to the flow field (finite element coefficients) and extra unknowns, respectively. \( \partial R / \partial z \) is the Jacobian matrix of the original problem. The iteration to solve the augmented Navier–Stoke system proceeds until the \( L_2 \) norm of the augmented residual \( ||R^A(z^A)||_2 \) is less than \( 10^{-8} \).

By solving the augmented Navier–Stokes system, one can perform direct tracking of limit flow states based on a multi-parameter continuation. The basic scheme is shown in Fig. 6.5. \( \alpha \) and \( \beta \) are chosen operating or design parameters from the parameter space: \( \alpha \) is part of augmented Navier–Stokes system, and \( \beta \) is controlled by an user-defined rule for continuation. Here we use a simple rule \( \beta_{(i+1)} = \beta_{(i)} + \delta \beta \), the subscript stands for the number of the continuation step and \( \delta \beta \) is a given step size that depends on the flow. Steps (I) and (II) are used to find the first critical flow state and the parameter \( \alpha \) at which it occurs at a given value \( \beta \). Steps (III) through (V) represents the multi-parameter continuation used to track the critical parameter curve as the continuation parameter \( \beta \) changes. In this study, we use a tangent predictor [Seydel 1994] for step (IV).

6.2.4 Construction of operating window and vortex-free operating window

Figure 6.6 shows an example of an operating window as a function of the dimensionless wet thickness \( h_w^* \) and tension number \( N_T \). The path of the continuation procedures used to construct the lines that define the bead breakup, weeping, birth of vortex inside the feed slot and attached to the downstream die lip are also shown in the plot.
6.2. ELASTOHYDRODYNAMIC MODEL AND SOLUTION METHOD

(I) Solve Navier–Stokes (NS) System with $\alpha$ and $\beta$ fixed

(II) Bordering algorithm to find $\alpha$ ($\beta = \beta_i$): Newton method for augment NS system

Start multi-parameter continuation (MPC)

(III) Change $\beta$

(IV) Bordering algorithm to find $\partial\alpha/\partial\beta$ ($\beta$ fixed): Predictor step for augment NS system

(V) Bordering algorithm to find $\alpha$ ($\beta$ fixed): Corrector step for augment NS system

Is $\beta = \beta_f$ ?

End MPC

Figure 6.5: Procedure of direct tracking algorithm. $\alpha$ and $\beta$ are chosen operating or design parameters.
Figure 6.6: Construction of vortex-free operating window (vortex-free and operating window) for BC configuration using direct tracking of flow features.
6.2. ELASTOHYDRODYNAMIC MODEL AND SOLUTION METHOD

The detailed procedure to construct the operating window and the vortex-free window are described as follows:

Step 1. Perform arc-length continuation (Bolstad and Keller, 1986) with increasing wet thickness at a constant web speed, $U_w = 1 \text{ m/s}$ in this example: path (A) in Fig. 6.6.

Step 2. Select the flow states along path (A) that correspond to weeping, feed slot vortex birth and bead breakup.

Step 3. Track critical wet thicknesses for weeping, feed slot vortex birth, and bead breakup as web speed rises: paths (B), (C), and (D) in Fig. 6.6.

Step 4. At a given web speed ($U_w = 4.8 \text{ m/s}$ in this example), perform arc-length continuation with decreasing wet thickness from the flow state along path (B) (onset of weeping): path (E) in Fig. 6.6.

Step 5. Select flow state at birth of die lip vortex along path (E).

Step 6. Track critical wet thickness for die lip vortex birth as web speed falls: path (F) in Fig. 6.6.

The area bounded by curves (A), (B), (F), (E) and (C) — shaded in the plot — corresponds to the set of parameters at which the coating flow is feasible (without weeping and bead breakup) and free of vortices.

6.2.5 Die lip configurations

The effect of the downstream die lip geometry was analyzed by varying four different parameters: the die lip radius of curvature $R_d$, its length $L_d$, the apex point location $Y_{\text{Apex}}$ and the patch point location $Y_{\text{Patch}}$ in the case of a compound die with a rounded and a straight sections.

By adjusting these four parameters, six different die lip configurations were studied. They are sketched in Fig. 6.7 and their geometric parameters are listed in Tab. 6.2.
Figure 6.7: Die lip configurations considered in this study. BC, LR, SR, Sh, Comp and DA stand for base case die lip, large downstream die lip radius, small downstream die lip radius, short die lip length, compound die lip with a rounded and a straight section, and downstream die lip with apex point in the middle configurations, respectively. $L_d$ and $R_d$ are downstream lip length and downstream lip radius of curvature, respectively. $Y_{patch}$ is the location of patch point between straight die lip section and rounded die lip section. $Y_{apex}$ is downstream apex point for the downstream die lip. Big dots on the downstream die lip stand for the downstream apex point. All die lip configuration has the same upstream die lip. See Table 6.2 for geometric parameters details.
6.3. RESULTS & DISCUSSION

Table 6.2: Geometric parameters for die lip configurations. See Fig. 6.7. Upstream die lip for all die lip configurations are the same: die lip length is 1.1 \( mm \), radius of curvature is 0.250\(^\circ\), and the apex point for the upstream die lip is located at the upstream corner.

<table>
<thead>
<tr>
<th>Name</th>
<th>( R_d )</th>
<th>( L_d )</th>
<th>( Y_{patch} )</th>
<th>( Y_{Apex} )</th>
<th>( \theta_{w,d} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>BC</td>
<td>0.250&quot;</td>
<td>1.1 ( mm )</td>
<td>0.0 ( mm )</td>
<td>0.0 ( mm )</td>
<td>9.97(^\circ)</td>
</tr>
<tr>
<td>LR</td>
<td>0.500&quot;</td>
<td>1.1 ( mm )</td>
<td>0.0 ( mm )</td>
<td>0.0 ( mm )</td>
<td>4.97(^\circ)</td>
</tr>
<tr>
<td>SR</td>
<td>0.125&quot;</td>
<td>1.1 ( mm )</td>
<td>0.0 ( mm )</td>
<td>0.0 ( mm )</td>
<td>20.2(^\circ)</td>
</tr>
<tr>
<td>Sh</td>
<td>0.250&quot;</td>
<td>0.55 ( mm )</td>
<td>0.0 ( mm )</td>
<td>0.0 ( mm )</td>
<td>4.97(^\circ)</td>
</tr>
<tr>
<td>Comp</td>
<td>0.250&quot;</td>
<td>1.1 ( mm )</td>
<td>0.55 ( mm )</td>
<td>0.0 ( mm )</td>
<td>4.97(^\circ)</td>
</tr>
<tr>
<td>DA</td>
<td>0.250&quot;</td>
<td>1.1 ( mm )</td>
<td>0.0 ( mm )</td>
<td>0.55 ( mm )</td>
<td>4.83(^\circ)</td>
</tr>
</tbody>
</table>

An important parameter to be considered is the downstream wrap angle \( \theta_{w,d} \), indicated in Fig. 6.3. When \( \theta_{w,d} \) is too large for a given lip configuration, the downstream static contact line will wet the die shoulder leading to a non-uniform downstream meniscus. The opposite case, too small \( \theta_{w,d} \), will create a diverging channel between the die lip and the moving web near the meniscus that may cause a large adverse pressure gradient. This pressure gradient can cause a ribbing instability that destroys cross-web coating uniformity. Since the downstream die departure angle \( \theta_{L,d} \) is a function of die lip geometry, the wrapping angle, \( \theta_{w,d} \) need to be adjusted by changing the \( x \)-coordinate of the downstream web extreme point at each die lip configuration. Here, we set \( \theta_{w,d} = \theta_{L,d} - 1^\circ \) at the downstream die corner in order to create an almost parallel channel near the meniscus.
Figure 6.8: Effect of lip length on vortex-free operating window by comparing two die lip length configurations: 1.1 mm for BC and 0.55 mm for Sh. $h_{w,M}(N_T)$ and $h_{w,m}(N_T)$ stand for maximum and minimum wet thicknesses, respectively. Note that V.B. and O.L. stands for vortex birth and operating limit, respectively.
6.3 Results & Discussion

6.3.1 Effect of die lip geometry on operating window

Effect of die lip length

Two different configurations — BC, with downstream lip length equal to 1.1 mm and Sh, with lip length equal to 0.55 mm — are used to show the effect of the lip length on the operating window of the process (Figure 6.8). The radius of curvature of the die is the same $R_d = 0.250"$. The results show significant differences at the weeping limit curve and die lip vortex birth curve, but virtually no effect on the onset of feed slot vortex birth and bead breakup limit.

As the die lip length decreases, the length of web over the die lip, or footprint, shrinks as well. In order to support the loading force from the curved web under tension, the short lip configuration lead to a higher pressure near the feed slot exit, as shown in Fig. 6.9. Because the pressure at either the upstream or the downstream menisci is close to ambient pressure, for both configurations. Therefore, the Sh configuration has steeper pressure gradients along the upstream and downstream die lip. Accordingly, the distance between the web and die lip, i.e. the gap height, is smaller in order to accommodate the same flow rate.

The upstream length of the bead, measured from the feed slot exit to the upstream meniscus, for the Sh configuration is shorter than for the BC configuration, even though they have the same upstream die lip geometry, at the same flow rate. This tendency will delay weeping phenomena: weeping curve on Fig. 6.8 is shifted upwards (higher wet thickness) for the Sh configuration.

As it is clear in Fig. 6.9, downstream die lip length affects the pressure gradient along the die downstream lip, that is the source of die lip vortex. Figure 6.10 shows the pressure profile under the die lip at a set of conditions near the onset of the die lip vortex for both configurations. At this condition (high $N_f$ and $h^*_w$) the adverse pressure gradient from the exit of the feed slot to the local maximum pressure is steeper with the BC configuration, which promotes the onset of lip vortex at lower
6.3. RESULTS & DISCUSSION

Figure 6.9: Pressure profile along moving web for BC configuration and Sh configuration for \( h_w^* = 1.1 \times 10^{-2} \) and \( N_T = 1.4 \times 10^{-4} \).
6.3. 

RESULTS & DISCUSSION

Figure 6.10: Pressure profile along moving web for BC configuration and Sh configuration for $h_w^* = 8.1 \times 10^{-2}$ and $N_T = 6.3 \times 10^{-4}$.

wet thickness. This explains the reason why the curve that marks the onset of die lip vortex is shifted toward higher thickness with the Sh configuration.

The microvortex at the feed slot is caused by the adverse pressure gradient near the exit of the slot. The pressure along the feed slot for both configurations at a set of conditions near the onset of feed slot vortex is presented in Fig. 6.11. Because the pressure along the coating bead changes due to the different geometry, the pressure at the exit of the feed slot is not the same. However, the pressure gradient at every point along the wall is virtually the same. The pressure profile with the Sh is simply translated upwards. This behavior explains why the lip length has no effect on the vortex birth inside the feed slot.

According to elasto-hydrodynamic theory, pressure is proportional to the curvature and tension of the web. At low flow rates, i.e. small wet thickness, the curvature of the moving web is close to the radius of curvature of the die lip, and the pressure at the feed slot are similar for both configurations. Since the upstream meniscus location
is strongly affected by the pressure field inside the coating bead, the bead breakup limit curves should be close to each other, as shown in Fig. [6.8]

**Effect of die lip curvature**

As discussed before, the menisci positions and consequently the operating limits are a function of the pressure profile along the coating bead, which is a direct function of the web curvature. Therefore the radius of curvature of the die lip should have a strong effect on the operability limits of the process. Three different downstream die lip configurations are used to study the effect, BC, SR, and LR. The operability and vortex birth limits obtained with each configuration is shown in Fig. [6.12]. For comparison purpose, we choose the downstream lip radius of the BC configuration, \( R_{d,base} \), as the characteristic length unit. The higher pressure obtained with the die with smaller radius of curvature shifts the minimum and maximum thickness at a fixed tensioned number to lower values, when compared to the BC configuration.
Figure 6.12: Effect of lip curvature on vortex-free operating windows by comparing three radius of curvature: 0.125” for SR, 0.250” for BC, and 0.500” for LR. $h_{w,M}(N_T)$ and $h_{w,m}(N_T)$ stand for maximum and minimum wet thicknesses, respectively. Note that V.B. and O.L. stands for vortex birth and operating limit, respectively.
6.3. RESULTS & DISCUSSION

Figure 6.13: Pressure profile along moving web for three different die lip radius of curvatures: 0.125” for SR, 0.250” for BC, and 0.500” for LR. $R_{d,\text{base}}$ is the downstream die lip radius of curvature of BC configuration.

Thinner wet films can be obtained with die lips with a small radius of curvature. The effect of the downstream die lip radius on the onset of the die lip vortex and feed slot vortex is small, as shown in the plot.

Figure 6.13 shows the pressure profiles for all configurations in units of web tension over downstream lip radius of the BC configuration. As the curvature of downstream die lip rises, i.e. as the radius of curvature of the die lip falls, the pressure inside the coating bead rises significantly and it will push the upstream meniscus away from the feed slot. As a consequence, the bead breakup will be delayed and the weeping will be accelerated, as shown in Fig. 6.12.

The vortex birth curve for all configurations does not change significantly, especially for vortex birth inside the feed slot. Similar to the lip length, the pressure profile along the feed slot wall for all configurations were virtually the same when they are
shifted properly: the reversal flow that comes from the adverse pressure gradient is not affected by the die lip curvature. However, the die lip curvature slightly influences the vortex birth under die lip, especially for small radius of curvature die lip.

Effect of downstream apex point

Takahashi and Shibata (1995) claimed that coating defects that comes from foreign particles (streak line) can be prevented when the apex point of the downstream die lip is shifted from the corner of the feed slot exit to the middle of the downstream die lip. In order to study the effect of the apex point on the operating window, we

Figure 6.14: Effect of downstream apex point on vortex-free operating windows by comparing two different configurations: apex point located at the upstream corner of the downstream die lip for BC and 0.567 mm away from the feed slot for DA. \( h_{w,M}^*(N_T) \) and \( h_{w,m}^*(N_T) \) stand for maximum and minimum wet thicknesses, respectively. Note that V.B. and O.L. stands for vortex birth and operating limit, respectively.
6.3. RESULTS & DISCUSSION

Figure 6.15: Pressure profile along moving web for two different downstream
die lip apex point location: 0.0 mm for BC, and 0.567 mm for DA.

compared the predictions obtained with the DA configuration to the BC; the results
are shown in Fig. 6.14.

Shifting the apex point to the middle of the downstream lip pushes both the weeping
and bead break-up limits to higher wet thickness. This occurs because the local
maximum pressure under the downstream lip is shifted upstream, located near the
apex point, and consequently the pressure near the feed slot is reduced, as presented
in Fig. 6.15. Therefore, at the same operating parameters, the upstream meniscus
will be located closer to the feed slot with the DA configuration.

Since the DA configuration has a converging channel between the web and the die lip
spanned from the upstream corner to the apex point of the downstream die lip, there
is always an adverse pressure gradient along the web in this portion of the coating
bead. This adverse pressure gradient promotes vortex birth under the die lip. The
change in the pressure field also affects the feed slot vortex birth. Consequently, the
curve that mark the onset of vortex formation under the die lip and on the feed slot are shifted to lower wet thickness when the DA configuration is used, as shown in Fig. 6.14.

Effect of straight die lip

A downstream die lip that has two sections with different curvature has been suggested by several patents (Shibata and Chino 1993; Okuno and Kawabe 1994) in order to control the pressure inside coating bead. Here, to enhance any effect of this configuration, we analyze a die lip with two sections, a rounded portion followed by a straight section (infinite radius of curvature) as sketched in Fig. 6.7. The operating window predicted with this configuration (Comp) is compared to the one obtained

Figure 6.16: Effect of additional straight die lip section on vortex-free operating windows by comparing Sh configuration and Comp configuration. $h_{w,M}^*(N_T)$ and $h_{w,m}^*(N_T)$ stand for maximum and minimum wet thicknesses, respectively. Note that V.B. and O.L. stands for vortex birth and operating limit, respectively.
when the short die lip (Sh) is used, which corresponds to the first section of the compound die lip design. The results will clearly show the effect of adding a straight portion on an existing die lip on the operability limits and vortex formation.

The effect of the straight section is minor, as shown in Fig. 6.16. The operability limits and vortex-free window are almost identical, with the exception that the weeping limit is shifted to larger wet thickness. The reason is that the pressure profile near the feed slot and under the rounded portion of the die lip is very close to that of the short die lip, as presented in Fig. 6.17.

6.3.2 Feed slot vortex birth condition

Figure 6.18 shows pressure profiles along the feed slot for all the die lip configurations analyzed at the conditions of vortex birth at a tension number $N_T = 2.7 \times 10^{-4}$. All
6.3. RESULTS & DISCUSSION

Figure 6.18: Pressure profile along the downstream wall of feed slot for different die lip configurations.
cases show an adverse pressure gradient near the feed slot exit that leads to flow detachment and consequently to vortex birth. Even though the values of pressure are not the same, the pressure profiles for all the configurations can be overlapped when properly shifted. This similarity may imply a simple condition for feed slot vortex birth.

The presence of micro recirculation inside the feed slot in a fixed-gap slot coating process was studied by [Sartor (1990)]. For the range of parameters in his analysis, he found that a vortex will appear at the end of the feed slot when the feed slot height reaches the critical value of five times the wet thickness.

The critical wet thickness, in units of feed slot height \( h_w \), at the condition of feed slot vortex birth at different values of tension number \( N_T \) and feed slot height \( H_f \) for all the geometries analyzed in this work is presented in Fig. 6.19. The simple criteria suggested by [Sartor (1990)], e.g. vortex birth occurs at \( h_w^\dagger = h_w/H_f = 1/5 \), approximates well most of the predictions.

We observed that the departure from the critical wet thickness (flow rate) of \( h_w^\dagger = 0.2 \) is a function of the Reynolds number of the flow. The critical wet thickness at feed slot vortex birth was automatically tracked by continuation in different dimensional parameters: feed slot height \( H_f \), viscosity \( \mu \), density \( \rho \), and web speed \( U_w \). The four curves are plotted in Fig. 6.20 as a function of the Reynolds number, \( N_{Re} \equiv \rho h_w U_w / \mu \). The curves clearly lay on top of each other. As Reynolds number increases, the critical thickness decreases from 0.2 to 0.1. In other words, lower flow rate, or smaller wet thickness \( h_w \), at a given web speed \( U_w \), is required to cause vortex at the feed slot. High inertia helps to prevent flow reversal near the exit of the feed slot. However, the Reynolds number for typical tensioned-web-over-slot die coating operation conditions are typically less than 5, and in this range \( h_w^\dagger \sim 0.2 \) is still an reasonable critical condition for feed slot vortex birth.

### 6.3.3 Minimum wet thickness

The minimum wet thickness that can be coated without bead break up at a given operating condition is one of the most important output of the model. The result can
6.3. RESULTS & DISCUSSION

**Figure 6.19:** Critical wet thickness $h_w$ for feed slot vortex birth in unit of feed slot height $H_f$, $h_{w,m}^+$. (a) shows $h_{w,m}^+$ for different tension number $N_T$. (b) shows $h_{w,m}^+$ for different feed slot height $H_f$, expressed in units of upstream radius of curvature $R_u$. Here, $N_T$ is fixed to $1.31 \times 10^{-4}$. 

---

- Wet thickness in the unit of $H_f$ ($h+w = hw / H_f$)
- Tension number ($N_T = \mu U_w / T$)
- Feed slot height in unit of $R_u$ ($H_f^* = H_f / R_u$)
Figure 6.20: Effect of inertia on critical wet thickness $h_w$ for feed slot vortex birth in unit of feed slot height $H_f$, $h_{w,m}$, for BC configuration.
also be interpreted as the maximum web speed possible at a given wet thickness.

**Effect of die lip geometric parameters on minimum wet thickness**

The dependence of the minimum wet thickness on die lip geometry can be determined by using direct tracking of bead break-up flow states as a given die lip geometric parameter is changed. Figure 6.21 shows the effect of four different die lip geometric parameters on the minimum wet thickness $h_{w,m}^*$. Plot (a) in Fig. 6.21 reveals that the minimum thickness $h_{w,m}^*$ falls as the radius of curvature decreases as discussed before. The downstream apex point location remarkably alters $h_{w,m}^*$ as shown in plot (b). As the apex point moves from the upstream corner of the downstream die lip ($Y_{Apex} = 0$) to the downstream corner ($Y_{Apex} = 1$), $h_{w,m}^*$ increases by almost one order of magnitude. Figure 6.21 (c) shows that as long as the downstream die lip is long enough, it has only small influence on $h_{w,m}^*$. When the die is too short, due to small wrap angle, it cannot create enough pressure to push the upstream meniscus to maintain the coating bead at small flow rates. Fig. 6.21 (d) shows that the downstream straight lip section does not significant impact $h_{w,m}^*$.

**Minimum wet thickness correlation**

The dependence of the minimum wet thickness in unit of the base case downstream die lip radius $R_{d,base}$, $h_{w,m}^* = h_{w,m}/R_{d,base}$, on tension number, $N_T$, is well described by power-law correlations, i.e. $h_{w,m}^* = C N_T^m$, for each die lip configuration, as shown in Fig. 6.22. According to the correlations in the figure, the coefficient $C$ depends strongly on die lip geometry, but the exponent $m$ depends weakly on the geometry.

When the downstream apex point is located at the exit of feed slot, the exponent $m$ does not depend on the downstream die lip shape: $m \approx 0.5$ for all cases analyzed. The coefficient $C$ is a strong function of the downstream radius of curvature $R_d$. However, when the apex point is located at the middle of die lip, both constant $C$ and exponent $m$ are a function of downstream radius of curvature $R_d$. Overall, the downstream apex point location and downstream lip curvature are the key die lip geometric parameters to control the minimum wet thickness.
Figure 6.21: Direct tracking of minimum wet thickness $h_{w,m}^*$ with respect to a given die lip geometric parameter. (a) effect of downstream die lip radius of curvature $R_d$ on $h_{w,m}^*$. (b) effect of downstream apex point location $Y_{Apex}$ on $h_{w,m}^*$. (c) effect of downstream die lip length $L_d$ on $h_{w,m}^*$. Here $L_{d,base}$ is the downstream die lip length of BC configuration. (d) effect of the patch point of composite downstream die lip $Y_{patch}$ on $h_{w,m}^*$. 
Figure 6.22: Correlation between minimum wet thickness and tension number for different die lip configurations. Solid lines are results from power-law relations $h_{w,m}^* = C N_T^{m}$ for each configuration.
6.3. RESULTS & DISCUSSION

Figure 6.23: Dependence of coefficient $C$ of the minimum thickness correlation, $h_w^* = C N_T^{0.5}$, on the downstream die lip curvature $\kappa_d^*$, when the downstream apex point is located at the feed slot. $R_{d,base}$ is 0.250". Coefficient $C$ is computed based on data shown in Fig 6.21(a).
6.4. Final remarks

When the apex point is located at the feed slot exit, an analytical expression for the minimum wet thickness can be derived by curve fitting the computer-aided predictions. It turns out that the coefficient $C$ is a linear function of die lip curvature, as shown in Fig. 6.23. These results were obtained at $NT = 1.31 \times 10^{-4}$. Since the exponent $m$ is 0.5, the minimum wet thickness can be expressed as

$$h_{w,m,b}^* = \left[ 6.5 \times 10^{-2} - \frac{1.3 \times 10^{-2}}{R_d^*} \right] N_T^{0.5}, \quad (6.34)$$

where $h_{w,m,b}^* = h_{w,m}/R_{d,base}$, $R_d^* = R_d/R_{d,base}$, $N_T = \mu U_w/T$ and $R_{d,base} = 0.250"$. The equation can be used as a guideline for determining minimum wet thickness for a given die lip configuration and tension number, or designing a suitable die lip curvature necessary for a desired operating condition.

6.4 Final remarks

A computational analysis of two-dimensional tensioned-web-over-slot die coating flow was carried out by means of Galerkin finite element method. In order to obtain an accurate description of the web, thin inextensible shell theory was used instead of simple membrane theory. Deploying direct tracking method of flow features, such as weeping, bead breakup and vortex birth, operating windows and vortex-free windows for various die lip configurations were computed automatically. In this study, we used die lip geometry based on rounded surfaces, as proposed by several patents.

The results show that the downstream die lip geometry is one of key parameters to adjust the pressure inside the coating bead. Here we analyzed seven lip configurations (Fig. 6.7) based on four geometric parameters — downstream lip radius of curvature, downstream die lip length, downstream lip apex point location and location of patch point between a straight lip section and a rounded lip section. Operating limits and vortex births conditions are strong function of some geometric parameters of the die lip. It turns out that the lip curvature and the apex point are the most influential parameters to regulate the windows, especially for weeping and bead breakup limits.

All four parameters do not change the vortex-free window considerably, especially for feed slot vortex birth. The results show that vortex birth at the feed slot occurred.
6.4. FINAL REMARKS

when the feed slot height is approximately five times the wet thickness. That is the same critical value for a conventional slot coating reported by Sartor (1990).

Since the purpose of using TWOSD is to produce ultra-thin layer, finding the minimum wet thickness for a given die lip configurations is extremely important. In this study, we considered that the minimum wet thickness occurs when the upstream meniscus reaches the feed slot, leading to bead break up. The prediction are well described by a power-law correlation between minimum wet thickness, in units of $R_{d,\text{base}}$, and tension number: $h_{w,m}^* = C N_T^m$. When the apex point is located at the upstream corner of downstream die lip, the coefficient $C$ is only a function of the lip curvature, and the exponent $m$ is virtually unaffected by geometric parameters. When the apex point is located in the middle of die lip, both $C$ and $m$ are both functions of the lip curvature.

For thinner coating, smaller downstream die lip radius of curvature and downstream apex point closer to the feed slot are favorable. However, in practice, too small radius of curvature requires large wrap angle to prevent ribbing instability. In this condition, substantial amount of normal stress resultant from large web curvature may push the web too much against the die surface, with the possibility of scratching. Also it may cause lateral or cross-web flow from the center to the edge of the die lip due to the extreme pressure difference between the coating bead and the ambient air. Also when the apex point is at the corner of the feed slot, particles in the coating solution may be trapped near the corner and create streak lines along the flow direction. These practical limitations, not considered in the analysis, should also be considered for a successful die lip design.
Chapter 7

Flow visualization of single-layer tensioned-web-over-slot die coating

7.1 Introduction

When a fluid is in contact with a solid flexible material, the interaction between the two phases plays an important role in controlling the location of the interface between the fluid and the solid material. This is called elastohydrodynamic interaction. It occurs in many different situations such as biological flows in joints and coating flows.

Several coating flows exploit the elastohydrodynamic interaction to deposit an ultrathin layer of liquid on top of a flexible moving substrate. Tensioned-web-over-slot die (TWOSD) coating is one of them. Unlike conventional slot coating, the TWOSD coating method does not have a rigid backup roll to support the substrate, and hence the gap height, the distance between the coating die and the substrate, is regulated by the elastohydrodynamic interaction between the coating liquid and the curved substrate under tension. In general, the method can maintain an extremely small gap height, up to few microns, without scratching the die lip surface. Hence, the minimum possible wet thickness which is a function of the gap height can be significantly reduced by the method [Nam and Carvalho 2009a].
Since the method was developed from industrial needs, only few scientific literature are available. Most of them were computational or simple mathematical analysis of the method (Feng, 1998; Lin et al., 2008; Nam and Carvalho, 2009a). There was an attempt to visualize the TWOSD flow (Lin et al., 2007) but the scope of the analysis was limited focusing only on the coating, bead breakup that occurs when the flow rate is smaller than a critical minimum value.

As discussed in Ref. Romero et al. (2004), the range of operating conditions of the tensioned web over slot coating method surrounded by different modes of failure, which are closely related to downstream meniscus configuration (ribbing), upstream meniscus location (bead breakup and weeping), and microvortex inside the coating flow (feed slot vortex). These different failure modes are summarized in Fig. 7.1 and a detail descriptions of each of them can be found in Ref. Nam and Carvalho (2009a).
7.2. FLOW VISUALIZATION — SETUP

The goal of the present work is to explore and examine the limit flow states that define the operating limits of TWOSD coating process by visualizing the flow. The critical parameter values for coating failure obtained from the visualizations are compared to the predictions from the computational model presented by Ref. Nam and Carvalho (2009a). Flow state evolution beyond the critical parameter values are explored. These flow states cannot be predicted by the two-dimensional computational model of Ref. Nam and Carvalho (2009a).

The visualization was done by installing a camera on top of the transparent moving substrate to record the configuration of the downstream meniscus and the upstream meniscus location. The presence of microvortex in the feed slot was verified by injecting tracer particles inside the coating flow.

As suggested by patents, as Ref. Shibata and Chino (1993), and recent computational analysis (Nam and Carvalho 2009a), we use rounded die lips with a single radius of curvature for both upstream and downstream die lips $R_d = R_u = 0.250 \text{in}$, the same as the BC configuration in Ref. Nam and Carvalho (2009a). The details of the flow features that lead to coating defects are visualized and the critical flow rates at the onset of the features are recorded to serve as basis of comparison to validate the theoretical predictions of Ref. Nam and Carvalho (2009a). The visualizations not only support the predictions from the computational model but also show detail mechanisms behind defect-causing flow features.

7.2 Flow visualization — setup

Flow visualization was done in a laboratory-scale coating apparatus with built-in tension controllers. The coating die was designed by Wieslaw Suszynski at the University of Minnesota and manufactured by PREMIER die Co., Chippewa Falls, WI. The web transport machine was designed and made by IMATION Co., St. Paul, MN. Schematic details of the apparatus setup is shown in Fig. 7.2.

The coating solution is stored in a tank that was connected to a calibrated precision gear pump (Parker-Nichols Zenith, West Newton, WA). The pump is driven by a
Figure 7.2: Tensioned-web-over-slot die coating apparatus setup
DC motor (EP3640 Graham company, Menomonee falls, WI). Flow rate is controlled by the pump speed and measured with a Coriolis flow meter (model MFC 100/MFS 3000, Krophne America Inc. Peabody, MA). Downstream of the pump, the liquid flows through a custom-made transparent polycarbonate debubbler in order to damp pulsations from the pump and check for the presence of gas bubbles in the coating solution. At small flow rates, close to bead breakup, even tiny air bubbles can break up the coating bead before the meniscus invasion that induce breakup. Therefore, removing all possible bubbles is important to capture the critical flow rate for the bead breakup accurately.

The web or the substrate is released from the unwinding roll and is wrapped around a rubber roll that is nipped against an idler roll. It reaches the liquid application zone between two aluminum idlers where the coating die is located. The radius of the idler is \( \frac{1}{2} \) in and its width is 7.63 in: they are wider than the 4 in-wide coating die and the 6 in-wide web. The upstream and downstream wrap angle, \( \theta_{d,w} \) and \( \theta_{u,w} \), were controlled by the position of the idlers, which can be adjusted by a microslide, as indicated in Fig. 7.2.

The coating liquid is applied over the moving substrate at the coating bead, and is almost completely removed from the surface of the substrate by a rubber squeeze, (C) in Fig. 7.2. If the liquid is not properly removed, it may cause slip in the rewinding roll that spoils a precise tension control.

Tension isolation occurs either by pushing an idler against a rubber roll, (A) in Fig. 7.2 or wrapping the substrate about 180° degree, (C). Due to the tension isolation, there are three tension zones in the coating station: the section of the web from unwinding roll to (A), from (A) to (C), and from (C) to rewinding roll. For the process, the important tension is the one acting along the second section, where the coating bead is located. It is measured by a tension transducer (model PS1010T, Vishay Transducers Americas, Norwood, MA) located at (B). The tension inside this zone is controlled by two servo motors (Kollmorgen Goldline XT, MT302A1-T2C1, Danaher motion Co.) attached to the rubber rolls, (A) and (C).

Figure 7.3 shows the coating die used in the experiments. The die is 4.0 in-wide and has a center-fed tear-drop cross section shaped cavity for distributing liquid across
the width of the die. The feed slot height, i.e. the distance between downstream and upstream die lip blocks, is adjusted by the thickness of the shim used. In these experiments, it was fixed at $H_f = 130\mu m$. Unfortunately, the surface of the coating die is not perfectly rounded due to machining imprecision, especially the upstream die lip, as shown in Fig. 7.3(b). This mechanical defects may affect the detail mechanism of weeping which will be discussed later.

We exploit the transparent polyethylene terephthalate film, which is 3.5 in wide, that enables visualization of the coating bead. A high speed CCD camera (DFK31BF03H, Image source Co., Germany) with a microscope lens (Micro-NIKKOR, 105mm, 1:28 aperture, Japan) and extension rings (PN-11 and PK-13, Nikon, Japan) was used in the experiments.

To quantify the size of the feed slot vortex, one has to visualize the separating streamline that encloses the vortex, however this is extremely hard to do in this micro-scale coating flow. Instead, we inject silvered sphere particles (conduct-o-fil S5000-S3 spherical shperiglass, Potter industries, Carlstadt, NJ) with diameter range from $5\mu m \sim 14\mu m$ through the inlet of die cavity. They deflect light emitted from a fiber optic lamp marking the pathline pattern. Note that the use of a fiber optic lamp reduces heating of the coating bead from radiation that helps to prevent fluid property changes due to temperature. Since the size of the vortex is small, we use a digital microscope (KH-7700, Hirox-USA, River Edge, NJ) with high magnification.

Figure 7.3: The coating die used for the flow visualization
7.3. FLOW VISUALIZATION RESULTS

**Figure 7.4:** Camera setup and visualization area in coating bead.

**Table 7.1:** Operating conditions and parameters for flow visualization.

<table>
<thead>
<tr>
<th>Physical parameter</th>
<th>value</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density ($\rho$)</td>
<td>1.206</td>
<td>g/L</td>
</tr>
<tr>
<td>Viscosity ($\mu$)</td>
<td>63</td>
<td>cP</td>
</tr>
<tr>
<td>Surface tension ($\sigma$)</td>
<td>64.6</td>
<td>dyne/cm</td>
</tr>
<tr>
<td>Web speed ($U_w$)</td>
<td>100</td>
<td>fpm</td>
</tr>
<tr>
<td>Tension ($T$)</td>
<td>2</td>
<td>PLI</td>
</tr>
<tr>
<td>Coat width ($W_d$)</td>
<td>4</td>
<td>in</td>
</tr>
</tbody>
</table>

instead of the high speed camera in order to be able to visualize the recirculated flow.

Figure 7.4 shows the set up of the camera or digital microscope and the visualization area, and Tab. 7.1 summarize the base case operating conditions.

### 7.3 Flow visualization results

Ribbing is a well-known flow instability in coating flows. It appears as a result of the competition between surface tension force that stabilizes the flow and the adverse pressure gradient that comes from a diverging channel that destabilizes the
7.3. FLOW VISUALIZATION RESULTS

Table 1: Operating and geometric parameters for base case die lip (BC) configuration.

<table>
<thead>
<tr>
<th>Operating parameters</th>
<th>Unit</th>
<th>Base case</th>
</tr>
</thead>
<tbody>
<tr>
<td>Top-layer film thickness</td>
<td>µm</td>
<td>30</td>
</tr>
<tr>
<td>Bottom-layer film thickness</td>
<td>µm</td>
<td>30</td>
</tr>
<tr>
<td>Web speed</td>
<td>m/sec</td>
<td>1</td>
</tr>
<tr>
<td>Web tension</td>
<td>lb f/in</td>
<td>1</td>
</tr>
<tr>
<td>Ambient pressure</td>
<td>Pa</td>
<td>0</td>
</tr>
<tr>
<td>Surface tension</td>
<td>dyne/cm</td>
<td>61</td>
</tr>
<tr>
<td>Interfacial tension</td>
<td>dyne/cm</td>
<td>0</td>
</tr>
<tr>
<td>Density for both layers</td>
<td>g/cm³</td>
<td>1</td>
</tr>
<tr>
<td>Top-layer viscosity</td>
<td>cP</td>
<td>23</td>
</tr>
<tr>
<td>Bottom-layer viscosity</td>
<td>cP</td>
<td>23</td>
</tr>
<tr>
<td>Dynamic contact angle</td>
<td>deg.</td>
<td>140</td>
</tr>
<tr>
<td>Static contact angle</td>
<td>deg.</td>
<td>60</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Geometric parameters</th>
<th>Unit</th>
<th>Base case</th>
</tr>
</thead>
<tbody>
<tr>
<td>Downstream lip length</td>
<td>mm</td>
<td>1.1</td>
</tr>
<tr>
<td>Mid lip length</td>
<td>mm</td>
<td>0.5</td>
</tr>
<tr>
<td>Upstream lip length</td>
<td>mm</td>
<td>1.1</td>
</tr>
<tr>
<td>Feed slot heights</td>
<td>µm</td>
<td>100</td>
</tr>
<tr>
<td>Downstream lip radius of curvature</td>
<td>in</td>
<td>0.250</td>
</tr>
<tr>
<td>Mid lip radius of curvature</td>
<td>in</td>
<td>0.250</td>
</tr>
<tr>
<td>Upstream lip radius of curvature</td>
<td>in</td>
<td>0.250</td>
</tr>
<tr>
<td>Downstream die lip departure angle</td>
<td>deg.</td>
<td>9.97</td>
</tr>
<tr>
<td>Upstream die lip approaching angle</td>
<td>deg.</td>
<td>16.0</td>
</tr>
<tr>
<td>Downstream web wrap angle</td>
<td>deg.</td>
<td>9.97</td>
</tr>
<tr>
<td>Upstream web wrap angle</td>
<td>deg.</td>
<td>15.9</td>
</tr>
</tbody>
</table>

**Figure 7.5:** Flow visualization of ribbing instability appeared when the small downstream wrap angle $\theta_{d,w} = 6^\circ$ is smaller than the downstream die lip departure angle $\theta_{L,d} = 9.97^\circ$.

Flow [Pearson (1960)] may form a diverging channel upstream of the free surface. When the downstream wrap angle $\theta_{d,w}$ is too small, we found that the downstream meniscus invades the coating bead and the meniscus surface shows a periodic wave pattern across the width of the substrate, as shown in Fig. 7.5. The cause of the ribbing instability, the diverging channel at the downstream die lip, can be removed by increasing the downstream wrap angle $\theta_{d,w}$ to a value close to the downstream die lip departure angle $\theta_{L,d}$ defined in Fig. 7.1. In TWOSD coating, ribbing instability is almost only a function of the wrapping angle $\theta_{d,w}$: it always appeared at all ranges of flow rate or web speed we explored in this study, if the wrapping angle $\theta_{d,w}$ was small enough. Therefore, we fix the wrapping angle $\theta_{d,w}$ to about $9.5^\circ$ which is close to $\theta_{L,d} = 9.97^\circ$ for excluding the possibility of onset of ribbing instability during the rest of the experiments.

When the upstream meniscus invades the feed slot, the shape of the meniscus become irregular along the cross flow direction and, eventually, the coating bead will break. This event is called bead breakup. As shown in Ref. Nam and Carvalho (2009a) at a
constant web speed $U_w$ and line tension $T$, the location of the upstream meniscus is a function of the wet thickness $h_w$: wet thickness $h_w$ below a critical value leads to bead breakup. Note that in pre-metered coating flows, the wet thickness is a direct function of the flow rate, at a fixed web speed.

Figure 7.6 shows the evaluation of the upstream meniscus location as $h_w$ decreases at $U_w = 200\, \text{fpm}$. At this web speed, the tension number is $N_T = \mu U_w / T = 1.82 \times 10^{-4}$ where $T$ is web tension and $\mu$ is the coating liquid viscosity. The onset of bead breakup in this study adopted the same criterion suggested by Ref. [Nam and Carvalho, 2009a]: the meniscus located at the feed slot corner, as shown in Fig. 7.6 (b). At this web speed, this event occurs at $h_{w,b} = 4.40\, \mu\text{m}$. Note that the computational model predicts $h_{w,b} = 4.48\, \mu\text{m}$. Higher than this critical wet thickness, the meniscus locates over the upstream die lip surface, as shown in Fig. 7.6 (a). Below this critical value $h_{w,b}$, the meniscus invades the coating bead towards the feed slot and it becomes jagged, like a sawtooth, as sketched in Fig. 7.6 (c). Air entrainment is observed at the sharp corners of the irregular-shaped meniscus and it creates dry lanes or rivulets. In general, the flow is highly unstable: these rivulets appear and disappear alternatively at high frequency.

If the flow rate is very high, the upstream meniscus reaches the end of the die lip surface, the meniscus surface becomes bulged and the liquid may drip. The process loses its pre-metered action, i.e. controlling wet thickness by flow rate. This undesired flow state is called *weeping* and occurs at high flow rate or wet thickness, at a given web speed.

Since high flow rate builds enormous pressure inside the liquid feeding system, the coating system suffered from serious leakage problems, especially for the coating die. We pick relatively slow speed, $U_w = 100\, \text{fpm}$ for the visualization of weeping, which is corresponding to $N_T = 9.14 \times 10^{-4}$.

The computational model predicts the onset of weeping at $h_{w,w} = 63.5\, \mu\text{m}$ using the criterion adopted in Ref. [Nam and Carvalho, 2009a], i.e. meniscus location at the upstream corner of the die lip surface. But, as mentioned before, the die lips used had mechanical defects, sharp corners in the middle of its surface, as sketched by dotted lines in Fig. 7.7. Therefore, the upstream meniscus lodges at the corner of
7.3. FLOW VISUALIZATION RESULTS

Figure 7.6: Flow visualization of bead breakup as decreasing wet thickness $h_w$ at the given web speed $U_w = 200$ fpm, from (a) to (c).
7.3. FLOW VISUALIZATION RESULTS

Figure 7.7: Flow visualization of weeping as increasing wet thickness $h_w$ at the given web speed $U_w = 100\text{fpm}$, from (a) to (c).
7.3. FLOW VISUALIZATION RESULTS

Figure 7.8: Flow visualization of feed slot vortex at wet thickness $h_w = 10 \mu m$ and web speed $U_w = 100 fpm$. Images are process by cutting off some color ranges.

the upstream die lip surface for a wide range of wet thicknesses, $h_w = 35 \sim 69 \mu m$, as shown in Fig. 7.7 (a): the contact line of the meniscus is virtually pinned at the corner. Also a relatively large curvature of the meniscus, due to narrow gap between the corner and the moving substrate, counteracts the high pressure inside the coating bead, which also delays weeping as well.

Eventually, at slightly larger wet thickness ($h_{w,w} = 71.2 \mu m$) than the computational prediction, the liquid menisci invades from both sides, Fig. 7.7 (b), and liquid drop(s) were observed thereafter, Fig. 7.7 (c). Note that the flooding of the liquid was not from the top to bottom, as predicted from the two-dimensional computational mode due to the mechanical defect. We declare this flooding phenomena as an experimental onset of weeping.

As mentioned before, the evidence of the feed slot micro vortex are visualized by injecting tracer particles. The feed slot area was magnified 280 times and a fiber optics was used to shine a light on the area. Since the particles are randomly distributed inside the cavity, we took a video for more than $5 \sim 10$ minutes and used post processing of the image to find the evidence of vortical motions of the particles. Figure 7.8 shows two consecutive frames, (a) and (b), from the digital microscope (15 fpm). Clearly, particle traces depict a swirling motion: it is an evidence of microvortex inside the feed slot. Due to relative larger exposure time than the high speed camera, the spiral passage of particles was captured.
Figure 7.8 (b) shows the nodular structure of the vortex: the cross sections of the vortical tube are not the same across its length. This structure may spoil the cross-web uniformity of the coating surface which is usually a product requirement. Note that the vortex was observed at a wet thickness of \( h_w = 10.8 \mu m \), which is approximately 1/12 of the gap height \( H_f = 130 \mu m \), i.e. below the critical vortex birth dimensionless wet thickness \( h_w^+ = h_w / H_f \sim 1/5 \) proposed by Ref. [Nam and Carvalho (2009a)].

### 7.4 Comparison: computational model vs. experiment

Throughout the visualizations of the TWOSD coating flow, we collected data points for onset of bead breakup, weeping and feed slot vortex in the parameter space of dimensionless wet thickness \( h_w^* = h_w / R_d \) (\( R_d \) is the radius of curvature of the downstream die lip) and tension number \( N_T \). All data points were found by fixing the web speed \( U_w \) and increasing or decreasing \( h_w \) through controlling the flow rate. When the flow rate was adjusted, we waited at least 2 minutes to ensure a steady-state flow.

For the onset of bead breakup, we slowly decrease the flow rate and found the critical wet thickness when the upstream meniscus reached the feed slot. For the onset of weeping, we slowly increased flow rate and found the wet thickness when the upstream die lip was flooded with the coating solution. However, the onset of feed slot vortex could not be captured during the visualization. In this study, we started from the flow with the vortex (closed circles in Fig. 7.9) and increased the flow rate, and hence the wet thickness, until the evidence of the vortex disappeared (open circles in Fig. 7.9). Note that the method we used is not to capture the onset of the microvortex, because the particles may disturb the flow so that the flow could be different without the particles.

The range of the web speed was based on the machine and control limits. The flow at the onset of weeping and the flow without feed slot vortex requires high flow rate that may lead to high pressure inside the coating system and cause serious leakages through the coating die. On the other hand, the flows at the onset of bead breakup
Figure 7.9: Comparison between predictions from the computational model and results from the flow visualization experiment.
7.5 Conclusion

Flow visualization is powerful tool to examine the different flow states in a coating flow. The images and experimental data should be used to validate computational predictions, to explore the effect of flow parameters beyond the limits at which the model is valid and to ultimately help the fundamental understanding of the physical mechanisms. However, capturing different flow states in micro-sized coating flows is extremely challenging.

According to Ref. Nam and Carvalho (2009a), the critical wet thickness for the onset of the vortex is nearly a function of the gap height when Reynolds number is moderately small $N_{Re} = \rho \alpha q/\mu \sim O(1)$, which is the case for our study. Open and closed circle data points in Fig. 7.9 support the computational predictions. This microvortex is usually linked to defect coating (Nam et al., 2009). In TWOSD coating flows, the existence of feed slot vortex was only shown theoretically or computationally (Nam and Carvalho, 2009a), because, unlike roll coating or slide coating (Schweizer, 1988) that have relative large vortices. In the TWOSD coating, the size of the vortex is in the order of ten microns.
In this study, the ribbing instability, the rivulet, and the dripping phenomena were visualized, and the three-dimensional nodular structure of the feed slot vortex was examined. Those flow states could not be described by the two-dimensional computational model proposed by Ref. (Nam and Carvalho, 2009a).

Not only qualitative, but also quantitative comparisons between the flow visualizations and the computational predictions were performed. The critical wet thickness data were collected with properly chosen limit flow states during the visualizations. The data were plotted in terms of two chosen dimensionless parameters, namely dimensionless wet thickness and tension number, and compared with the computational predictions. The agreement was excellent.
Chapter 8

Computational analysis of two-layer tensioned-web-over-slot die coating

8.1 Introduction

In continuous liquid coating, one or several liquid layers are deposited on a moving substrate and dried to form a solid film that serves a specific function, like enhancing optical property or improving scratch resistance. Liquid coating is also a very strong candidate for mass production of nanoparticle assembly films, as discussed by Maenosono et al. (2003b). Slot coating is one of the most versatile method used in the coating industry to obtain a precisely controlled thin uniform liquid layer over a substrate moving at high speed. In conventional slot coating, the substrate is supported by a rigid back-up roll to control the distance between the die surface and the substrate, so called the gap height. The range of operating parameters (coating window), in which the coating flow is two dimensional and steady, for the conventional fixed-gap slot coating, is bounded by different modes of failure that are usually accompanied by flow instabilities (Romero et al. 2004). In general, the minimum thickness is proportional to the gap height and it is difficult to achieve the thickness
8.1. INTRODUCTION

![Diagram of two-layer tensioned-web-over-slot die coating]

**Figure 8.1:** Two types of two-layer tensioned-web-over-slot die coating. Tandem coating method consists of two consecutive single-slot coaters in series to coat two different layer sequentially. Dual slot coating method uses a coating die with two feed slots to coat two layer simultaneously.

lower than 10\(\mu\)m at relatively high speed. The major difficulty comes from the practical limitation of using an extremely small gap height that may lead to clashing the roll and the coating die.

One way to maintain small gap height, down to the order of a micron, is to use unsupported flexible substrate; the method is called tensioned-web-over-slot die (TWOSD) coating. It exploits the competition between the hydrodynamic force due to the liquid flow and the normal force resultant from the curved substrate under tension. [Nam and Carvalho (2009a)] studied single-layer TWOSD coating flow using a computational model based on two-dimensional Navier—Stokes theory for the liquid and thin inextensible shell theory for the web under tension.

However, several products require more than one layer for optimal performance, for example one layer to enhance mechanical strength and the other to improve optical property. The most efficient way to manufacture these product is by applying all the
layers at once before they are solidified. In TWOSD coating, there are two possible ways to apply liquids on the moving substrate, as shown in Fig. 8.1. The tandem coating (a) consists of two consecutive single slot coaters in series to coat liquid layers sequentially. On the other hand, the dual slot coating (b) has a single liquid applicator with two feed slots to coat the two layers simultaneously. Both methods have their own strengths. For example, the tandem coating can be easily constructed with existing single slot coaters, while the dual slot coater may shift, or perhaps increase, operating window by getting rid of additional liquid/gas interface for the bottom layer — a potential source of flow instabilities.

In this study, we focused on dual slot TWOSD coating method. When both coating liquids are immiscible, they form an interlayer inside the coating bead. But, in several coating applications, both layers uses solvents that are miscible and the apparent interface is truly an inter-diffusion zone. However, as discussed in [Musson (2001)], it can be treated as a distinct interlayer with zero interfacial tension in high speed coating process.

[Nam and Carvalho (2009b)] studied the movement of the interlayer separation point along the mid lip as a function of the operating conditions in the fixed-gap dual slot coating. They found that the bottom-layer flow rate is the primary parameter for controlling the interlayer separation point. When the bottom-layer wet thickness in unit of the gap height is less than about one third, the point moves or jumps from the downstream corner of the mid lip to the other corner. This event is called mid-gap invasion and the detailed mechanism depends on the viscosity ratio between the coating liquids.

Like in the fixed gap dual slot coating, controlling of the location of the interlayer separation point is important in the dual slot TWOSD coating. [Nakama and Chin (1999)] claimed that there are two mechanisms of interlayer separation point movements that can destroy uniformity of the interlayer. One of them is the same as the mid-gap invasion: the point travels along the mid lip surface. The other is the feed slot invasion: the point moves into the downstream feed slot. According to them, the movements are directly related to the bottom-layer wet thickness in unit of the gap height measured at the downstream corner of the mid lip. However, unlike the
fixed-gap slot coating, the gap height is not known *a priori*.

Geometrically, the dual slot TWOSD coating flow is similar to the fixed-gap dual slot coating one. But, like the single-layer TWOSD coating process, the significance of viscous force and capillary force, that are important in the fixed gap coating method, are usually overshadowed by pressure force and normal stress resultant from the curved web under tension. Therefore the flow states that signal coating defects are similar in both single and dual layer processes. Mid-gap invasion, feed slot vortices, bead breakup and weeping limit the size of vortex-free operating window for the dual slot TWOSD coating. Determining the film thickness of both layers at which these limiting flow states occur and the effect of the operating parameters on them is crucial in the design of the process.

The scientific literature about the dual slot TWOSD coating method is scarce, but there are several patents related to this method. Chino et al. (1989) emphasized the importance of downstream and mid die lip radii in dual slot TWOSD coating. They disclosed the guideline for the choice lip radii based on the flow rates in order to prevent streak line along the flow direction. Tomaru (1995) claimed that the offset of the downstream die lip with respect to the other lips with proper wrap angles is important to control the pressure inside the coating bead. Takahashi and Shibata (1995) claimed that the properly chosen underbite downstream die lip can prevent the micro vortex inside the downstream feed slot. Tomaru et al. (1997) disclosed the method using a pre-wetted web with upstream die lip overbite configuration, i.e. pushing the upstream die lip further toward the web than the other lips in order to scrape the excessive pre-wetted solution. According to them, the upstream meniscus can be eliminated by the pre-wetted web and the overbite configuration can decrease pressure near the mid lip.

Park (2008) performed computational studies on two-layer tensioned-web-over-slot die coating using two different models: elasto-visco-capillary model and 1-D/2-D hybrid model. Both models assume that the moving web behaves like a thin membrane — ignoring tension variations along the length direction. However, these approximations are valid only at a limited parameter range where the curvature of web does not change rapidly (Nam and Carvalho 2009a). In the elasto-visco-capillary model,
8.1. INTRODUCTION

he used lubrication approximation to describe the liquid flow and the downstream meniscus is treated by the equation proposed by Landau and Levich (1942). Using this model, he showed the location of upstream meniscus and the interlayer separation point as a function of the operating conditions and die lip curvatures. However, the flow near the interlayer separation point is highly two-dimensional, because the flow separates at the point from the die lip. Therefore the model may not be suitable to predict the interlayer separation point location. In the 1-D/2-D hybrid model, Park used two-dimensional Navier-Stokes equation for the flow near the mid lip and both the upstream and the downstream of the coating bead were described by the same equations used in the elasto-visco-capillary model. Using the model, he focused on the effect of operating conditions on size of the downstream feed slot vortex. The analysis did not explain the critical conditions that define the boundaries of the operating window of the process.

Here, we predict the operating window of two-layer dual slot TWOSD coating method. The coating flow is analyzed by the fully coupled Navier—Stokes theory and thin inextensible shell theory in Galerkin/finite element framework. Details of this elastohydrodynamic model were discussed by Nam and Carvalho (2009a). In order to handle the interlayer and the interlayer separation point inside the flow, we use the method described by Nam and Carvalho (2009b). In order to determine the vortex-free operating window, the critical parameters for feed slot vortex births, bead breakup and weeping need to be tracked. Here, we use the direct tracking of flow feature method proposed by Nam et al. (2009). The results show how different operating conditions may be adjusted in order to coat thinner layers at higher speeds.

Furthermore, we also analyze the effect of die geometric parameters. Based on the die design used in Nam and Carvalho (2009c), we focused on four different parameters, namely downstream lip radius, mid lip radius, downstream die lip offset and mid lip apex point, on the coating window of the process. With aid of a powerful direct tracking of flow feature technique Nam et al. (2009), the boundaries of the vortex-free operating window are obtained automatically for each die lip configuration. The coating windows obtained for each die lip configuration are compared to determine the most critical lip design parameters for die shape optimization.
8.2 Elastohydrodynamic model and solution method

8.2.1 Governing equations and boundary conditions

Compared to the distance between the moving web and the die lips, which is in the order of a micron, the web can be considered infinitely wide. When the coating solution is dilute, the flow can be described by the Navier–Stokes equation and continuity equation for incompressible Newtonian fluid:

\[ \nabla \cdot u = 0, \quad \rho_i u \cdot \nabla u = \nabla \cdot \mathbf{T}_i, \]  

(8.1)

where \( \rho_i \) is the liquid density and \( \mathbf{T}_i \) is stress tensor. For Newtonian liquid, it is given by \( \mathbf{T}_i = -p \mathbf{I} + \mu_i \left[ \nabla u + (\nabla u)^T \right] \), where \( p \) is the pressure and \( \mu_i \) is the liquid viscosity. Here, subscript \( i \) defines the two liquid phases, \( i = t \) for top layer and \( b \) for bottom layer.

Proper boundary conditions are need to solve the Navier-Stoke system. In dual slot TWOSD coating flow, the domain is bounded by inflow and outflow planes, solid walls, gas-liquid interfaces and the flexible web that deforms according to elastohydrodynamic interaction, as shown in Fig. 8.2.

At both feed slot inlets, we imposed,

\[ u = U_b(x), \]  

(8.2)

where \( U_b(x) \) is the imposed parabolic velocity profiles. At outflow, we imposed

\[ n_b \cdot \nabla u = 0, \quad p = p_{amb}, \]  

(8.3)

where \( n_b \) and \( p_{amb} \) stand for the unit normal vector to the boundary and ambient pressure. The total wet thickness \( h_{tot,w} = h_{t,w} + h_{b,w} \) is determined by the flow rate of each layer and the web speed \( U_w \).

At rigid solid walls, the no-slip and no-penetration boundary conditions were imposed. Along the die surfaces and the feed slot walls,

\[ t_w \cdot u = 0, \quad n_w \cdot u = 0. \]  

(8.4)
Figure 8.2: Dual slot tensioned-web-over-slot-die coating model boundary conditions and parameters. \(L_d\) and \(R_d\) are downstream die lip length and radius of curvature. \(L_u\) and \(R_u\) are upstream die lip length and radius of curvature. \(H_f\) is feed slot height. \(\theta_{w,d}\) and \(\theta_{w,u}\) are downstream and upstream web wrap angle. \(\theta_{L_u}\) and \(\theta_{L,d}\) are upstream lip approaching angle and downstream lip departure angle. \(\theta_s\) and \(\theta_d\) are static contact angle and dynamic contact angle of coating liquid. \(T\) and \(U_w\) are tension and web speed, respectively.
Along the moving web,
\[ \mathbf{t}_w \cdot \mathbf{u} = U_w, \quad \mathbf{n}_w \cdot \mathbf{u} = 0, \tag{8.5} \]
where \( \mathbf{n}_w \) and \( \mathbf{t}_w \) are the unit normal and tangent vectors at the wall. At the dynamic contact point, where the upstream meniscus meets the moving web, Navier slip condition was used instead of the no slip in order to relieve the stress singularity,
\[ \frac{1}{\beta} \mathbf{t}_w \cdot (\mathbf{u} - U_w \mathbf{t}_w) = \mathbf{n}_w \mathbf{t}_w : \mathbf{T}_b, \tag{8.6} \]
where \( \beta \) is the slip coefficient. Here we choose \( \beta = 0.1 \, \text{g}^{-1} \text{sec}^{-1} \), based on the numerical tests reported by Sartor (1990).

Along the gas-liquid interfaces, a force balance and the no-penetration condition were imposed:
\[ \mathbf{n}_f \cdot \mathbf{T}_i = \frac{\partial \mathbf{u}_i}{\partial s} = 0, \tag{8.7} \]
\[ \mathbf{n}_f \cdot \mathbf{u} = 0, \tag{8.8} \]
where \( \mathbf{t}_f \) and \( \mathbf{n}_f \) are the local unit tangent and unit normal to the free surface, \( s \) is the arc-length coordinate along the interface, \( \sigma_f \) is the liquid surface tension.

Like the gas-liquid interfaces, the position of the interlayer is unknown \textit{a priori}. In order to track its location, three conditions — force balance, no-penetration and velocity continuity — need to be satisfied along the interlayer.
\[ \mathbf{n}_I \cdot \mathbf{T}_i = \mathbf{n}_I \cdot \mathbf{T}_b + \sigma_I \frac{\partial \mathbf{u}_i}{\partial s}, \tag{8.9} \]
\[ \mathbf{n}_I \cdot \mathbf{u}_i = \mathbf{n}_I \cdot \mathbf{u}_b = 0, \tag{8.10} \]
\[ \mathbf{t}_I \cdot \mathbf{u}_i = \mathbf{t}_I \cdot \mathbf{u}_b, \tag{8.11} \]
where \( \mathbf{n}_I \) and \( \mathbf{t}_I \) are the unit normal and tangent vectors to the interlayer. \( \mathbf{u}_i \) and \( \mathbf{T}_i \) are velocity and stress tensor for phase \( i \), and \( \sigma_I \) is the interfacial tension.

The interlayer starts at the separation point attached to the mid lip. We used zero shear rate condition (Nam and Carvalho, 2009b) to find the interlayer separation point:
\[ \frac{\partial \mathbf{u}_I}{\partial s} = 0, \tag{8.12} \]
where $u_t$ is the tangential velocity component and $s_n$ is the arc-length coordinate normal to the wall.

In this study, the deformation and location of the moving substrate are predicted using the equations of cylindrical shells [Flügge 1973].

\[
\begin{align*}
\frac{dT}{d\xi} + \kappa \frac{d}{d\xi} (\kappa D) + P_t &= 0 \\
-\frac{d^2}{d\xi^2} (\kappa D) + \kappa T + P_n &= 0 \quad \text{or} \\
\frac{d^2x}{d\xi^2} + \kappa \frac{dy}{d\xi} &= 0 \\
\frac{d^2y}{d\xi^2} - \kappa \frac{dy}{d\xi} &= 0
\end{align*}
\]

(8.13)

$\xi$ is the coordinate along the web. $T$ and $\kappa$ are the web tension and curvature at each position, and $x$ and $y$ are the Cartesian coordinates of points on the web. The web stiffness $D \equiv E t^3/12(1-\nu^2)$ is a function of the elastic modulus $E$, Poison ratio $\nu$, and thickness of the web $t$. $P_t$ and $P_n$ are the loading forces on the web in the tangential and normal direction. Those forces are responsible for the web deformation and can be obtained from the traction exerted by the fluid neighboring the web:

\[
P_t = t_w \cdot (n_w \cdot T) \quad \text{and} \quad P_n = n_w \cdot (n_w \cdot T),
\]

(8.14)

where $t_w$ and $n_w$ are tangent and normal vectors on the flexible moving web.

The shell equations require two corner conditions for position and curvature, and one condition for tension. In our model, corner conditions for the shell equations are

\[
\begin{align*}
x &= x_d, \quad \kappa = 0 \quad &\text{for downstream web end,} \\
x &= x_u, \quad \kappa = 0, \quad T = T_s \quad &\text{for upstream web end}
\end{align*}
\]

(8.15)

Figure 8.2 shows a graphical representation the boundary conditions (a) and the important parameters (b) of the model, including liquid properties and die lip geometries, and operating parameters. The operating parameters and geometric parameters and their values at the base case analyzed here are summarized in Table 8.1. The dimensionless groups with their base case values and dimensionless parameters used in this model are listed in Table 8.2.
Table 8.1: Operating and geometric parameters for base case die lip configuration.

<table>
<thead>
<tr>
<th>Operating parameters</th>
<th>Unit</th>
<th>Base case</th>
</tr>
</thead>
<tbody>
<tr>
<td>Top-layer film thickness($h_{t,w}$)</td>
<td>µm</td>
<td>30</td>
</tr>
<tr>
<td>Bottom-layer film thickness($h_{b,w}$)</td>
<td>µm</td>
<td>30</td>
</tr>
<tr>
<td>Web speed($U_w$)</td>
<td>m/sec</td>
<td>1</td>
</tr>
<tr>
<td>Web tension ($T$)</td>
<td>lb/in</td>
<td>1</td>
</tr>
<tr>
<td>Ambient pressure ($P_{amb}$)</td>
<td>Pa</td>
<td>0</td>
</tr>
<tr>
<td>Surface tension ($\sigma_f$)</td>
<td>dyne/cm</td>
<td>61</td>
</tr>
<tr>
<td>Interfacial tension ($\sigma_I$)</td>
<td>dyne/cm</td>
<td>0</td>
</tr>
<tr>
<td>Density for both layers ($\rho_t = \rho_b$)</td>
<td>g/cm³</td>
<td>1.2</td>
</tr>
<tr>
<td>Top-layer viscosity ($\mu_t$)</td>
<td>cP</td>
<td>23</td>
</tr>
<tr>
<td>Bottom-layer viscosity ($\mu_b$)</td>
<td>cP</td>
<td>23</td>
</tr>
<tr>
<td>Dynamic contact angle($\theta_d$)</td>
<td>deg.</td>
<td>140</td>
</tr>
<tr>
<td>Static contact angle($\theta_s$)</td>
<td>deg.</td>
<td>60</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Geometric parameters (See Fig. 8.2)</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Downstream lip length($L_d$)</td>
<td>mm</td>
<td>1.1</td>
</tr>
<tr>
<td>Mid lip length($L_m$)</td>
<td>mm</td>
<td>0.5</td>
</tr>
<tr>
<td>Upstream lip length($L_u$)</td>
<td>mm</td>
<td>1.1</td>
</tr>
<tr>
<td>Feed slot heights($H_{d,f} = H_{u,f}$)</td>
<td>µm</td>
<td>100</td>
</tr>
<tr>
<td>Downstream lip radius of curvature ($R_d$)</td>
<td>in</td>
<td>0.250</td>
</tr>
<tr>
<td>Mid lip radius of curvature ($R_m$)</td>
<td>in</td>
<td>0.250</td>
</tr>
<tr>
<td>Upstream lip radius of curvature ($R_u$)</td>
<td>in</td>
<td>0.250</td>
</tr>
<tr>
<td>Downstream die lip departure angle ($\theta_{L,d}$)</td>
<td>deg.</td>
<td>9.97</td>
</tr>
<tr>
<td>Upstream die lip approaching angle ($\theta_{L,u}$)</td>
<td>deg.</td>
<td>16.0</td>
</tr>
<tr>
<td>Downstream web wrap angle ($\theta_{d,w}$)</td>
<td>deg.</td>
<td>9.97</td>
</tr>
<tr>
<td>Upstream web wrap angle ($\theta_{u,w}$)</td>
<td>deg.</td>
<td>15.9</td>
</tr>
</tbody>
</table>
### Table 8.2: Dimensionless numbers and parameters used in two-layer TWOSD computational study.

<table>
<thead>
<tr>
<th>Name</th>
<th>Dimensionless numbers</th>
<th>base case value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Top-layer Reynolds number ( (N_{Re,t}) )</td>
<td>( \frac{\rho_t h_{t,w} U_w}{\mu_t} )</td>
<td>1.565</td>
</tr>
<tr>
<td>Bottom-layer Reynolds number ( (N_{Re,b}) )</td>
<td>( \frac{\rho_b h_{b,w} U_w}{\mu_b} )</td>
<td>1.565</td>
</tr>
<tr>
<td>Modified Reynolds number ( (N_{mRe}) )</td>
<td>( \frac{\rho_b R_d U_w}{\mu_b} )</td>
<td>331.3</td>
</tr>
<tr>
<td>Tension number ( (N_T) )</td>
<td>( \frac{\mu_b U_w}{T} )</td>
<td>( 1.314 \times 10^{-4} )</td>
</tr>
<tr>
<td>Elasticity number ( (N_{ES}) )</td>
<td>( \frac{D}{T R_d^2} )</td>
<td>( 2.682 \times 10^{-12} )</td>
</tr>
<tr>
<td>Capillary number ( (N_{Ca}) )</td>
<td>( \frac{\mu_b U_w}{\sigma_f} )</td>
<td>0.377</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Name</th>
<th>Dimensionless parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Top-layer wet thickness ( (h_{t,w}^* ) )</td>
<td>( \frac{h_{t,w}}{R_d} )</td>
</tr>
<tr>
<td>Bottom-layer wet thickness ( (h_{b,w}^* ) )</td>
<td>( \frac{h_{b,w}}{R_d} )</td>
</tr>
<tr>
<td>Dimensionless mid lip radius ( (R_m^* ) )</td>
<td>( \frac{R_m}{R_d} )</td>
</tr>
<tr>
<td>Dimensionless upstream lip radius ( (R_u^* ) )</td>
<td>( \frac{R_u}{R_d} )</td>
</tr>
<tr>
<td>Dimensionless pressure ( (P^* ) )</td>
<td>( \frac{P}{T/R_d} )</td>
</tr>
</tbody>
</table>
Since there is no fixed representative characteristic length scale for the model, we evaluate Reynolds number based on flow rate — product of wet thickness and web speed — for each layer. However, it is convenient to define another Reynolds number based on the fixed length unit to represent the relative importance of inertia to viscous force. Here, we define modified Reynolds number based on the downstream die lip radius:

\[ N_{Re}^m = \rho b R_d U_w \mu_b. \]  \hspace{1cm} (8.16)

During computational study, we did not vary the density, \( \rho = \rho_t = \rho_b \). Therefore, there are simple relationships between the Reynolds numbers for both layers and the modified Reynolds number:

\[ N_{Re,t} = h_{*t.w}^* N_{Re}^m, \quad N_{Re,b} = h_{*b.w}^* N_{Re}^m. \]  \hspace{1cm} (8.17)

The governing equations and their boundary conditions, Eqs. (8.1) to (8.15), are posed in an unknown flow domain. This type of problem is often called a free boundary problem. Kistler and Scriven (1984) explained the basis of solving viscous free surface problems. The method of coupling viscous free surface flow and flexible moving substrate used here was discussed by in detail Carvalho (2003) and Nam and Carvalho (2009a).

To solve a free boundary problem by means of standard techniques for boundary value problems, the set of differential equations and boundary conditions posed in the unknown physical domain have to be transformed to an equivalent set defined in a known, fixed computational domain. This transformation is made by mapping \( x = x(\xi) \) that connects two domains. The physical domain is parameterized by the position vector \( x = (x, y) \), and the reference domain by \( \xi = (\xi, \eta) \). The mapping used here is the one described by de Santos (1991). He showed that a functional of weighted smoothness can be used successfully to construct the sort of map involved here. The inverse mapping is governed by a system of elliptic differential equations identical to those encountered in the dilute regime of diffusional transport.

\[ \nabla \cdot D_{\xi}(\xi, \eta) \nabla \xi = 0, \quad \nabla \cdot D_{\eta}(\xi, \eta) \nabla \eta = 0. \]  \hspace{1cm} (8.18)

\( D_{\xi} \) and \( D_{\eta} \) are mesh diffusivities which control the steepness of gradients in the node-spacing by adjusting the potentials \( \xi \) and \( \eta \). Curves of constant \( \xi \) and \( \eta \) define the
8.2. ELASTOHYDRODYNAMIC MODEL AND SOLUTION METHOD

Boundaries of elements used to describe the domain. The cross point of these curves sets the position of a node. Boundary conditions are needed to solve the second-order differential equations (8.18). Solid walls and inflow and outflow planes are described by the function that defines their geometry and nodes were distributed along them by a specified hyperbolic tangent type stretching function (Vinokur, 1983a).

Because the model needs to capture the movement of the interlayer separation point, the mid die lip edges cannot be described as a mathematical sharp corner which would lead to artificial pinning of the point. We describe the lip geometry as a combination of arc of circles: each corner has radius of curvature \( R_c = 50 \mu m \) and the mid lip has radius of curvature \( R_m \). These circles are patched together such that the slopes of each circle are continuous across the patched points. In order to have a smooth but maintain high node concentration around the rounded corners, we used the weighting function based on curvature of lip geometry to distribute nodes along the lip (Thompson et al., 1985):

\[
    w(s) = 1 + \alpha |\kappa(s)|, \tag{8.19}
\]

In order to prevent the discontinuity on the weighting function \( w(s) \), which may cause discretization error, we smoothed \( w(s) \) based on the method proposed by Nam and Carvalho (2009b).

The location of gas-liquid interface and the interlayer are implicitly determined by the kinematic condition, Eq. (8.8), and the location of the flexible substrate is implicitly determined by the thin cylindrical shell equations, Eqs. (8.13). The discrete version of the mapping equations is generally referred to as mesh generation equations. Detailed procedure and boundary conditions used for mesh equation are discussed in de Santos (1991).

The physical domain, including the liquid flow and the curved web, was tessellated as shown in Fig. 8.3. The zoomed mesh plot, insert (a), shows the downstream region of the mid lip where the downstream rounded corner of the mid lip, the interlayer and the interlayer separation point are located. See Appendix A for details about mesh constructions for rounded corners in the mid die lip.
Figure 8.3: Tessellation of tensioned-web-over-slot-die coating model. Mesh used to solve the governing equations by G/FEM 927 elements and 17379 unknown coefficients of the finite element basis functions are used to discretize the system. Note that each quadrilateral represents a element that have nine nodes.
8.2.2 Solution of the Navier–Stokes / thin inextensible cylindrical shell system for free surface flow by G/FEM

Galerkin finite element method was used to solve the Navier–Stokes equation (8.1) coupled with the shell equations (8.15) and the mesh generation equations (8.18). Each independent variable, velocity, pressure, nodal position, tension and curvature, was approximated by a linear combination of a finite number of basis functions, which are the unknowns of the discretized problem. For two-dimensional liquid domain, velocity and nodal position were represented in terms of Lagrangian bi-quadratic function \( \phi^i(\xi, \eta) \) and pressure field was represented using piecewise linear basis function \( \psi^k(\xi, \eta) \). This particular choice of basis functions for velocity and pressure made velocity continuity at the interlayer, Eq. (8.11), redundant; and physical pressure jump across the interlayer can be handled naturally without any modification (Nam and Carvalho, 2009b). For the one-dimensional shell elements, velocity and nodal position values are shared with the neighboring liquid element, and tension and curvature are expanded by continuous quadratic functions \( \varphi^j(\eta) \).

The weak forms of Eqs. (8.1) and (8.18) were obtained by multiplying each equation by weighting functions, integrating over the physical domain, and applying the divergence theorem to the appropriate terms. Essential boundary conditions were imposed by replacing the corresponding weighted residual equation with the desired velocity or node specification. Natural boundary condition were applied through the boundary integral that comes from the divergence theorem.

Along the liquid/web boundary, the mesh equations were replaced by the thin cylindrical shell equations (8.13). After applying integral by parts on the second derivative terms, residual equations for the tangential and normal force balance equations can be decomposed into normal and tangential forces from the curved web and loading forces from the liquid. As discussed in Nam and Carvalho (2009a), these two terms can be evaluate at the shell element and the liquid element, separately. Since there is only one equation for the nodal position, third equation of Eqs. (8.13), we impose a hyperbolic stretching function to distribute nodes for the other residual equation for nodal position. The \( x \) and \( y \) momentum weighted residual along the substrate are replaced by the no-slip/no-penetration boundary condition. See Nam and Carvalho (2009a).
8.2. ELASTOHYDRODYNAMIC MODEL AND SOLUTION METHOD

(2009a) for details of coupling between the Navier–Stokes equation and the thin shell equations in Galerkin finite element context.

In sum, the Galerkin finite element method (G/FEM) reduces the Navier–Stokes, mesh generation and thin cylindrical shell equations to a set of nonlinear algebraic equations on the basis function coefficients.

\[ R(z, \lambda) = 0, \]  

where \( z \) is the solution vector, that consists of the finite element coefficients for velocity \( u \), pressure \( P \), position \( x \), tension \( T \) and curvature \( \kappa \), and \( \lambda \) is a vector that contains the \( M \) parameters on which the system depends. The non-linear system of algebraic equations (8.20) is solved iteratively by Newton’s method:

\[ \begin{align*}
J^{(i)}(z^{(i)}, \lambda) \delta z^{(i)} & = -R^{(i)}(z^{(i)}, \lambda), \\
z^{(i+1)} & = z^{(i)} + \delta z^{(i)},
\end{align*} \]  

the indices \( i \) and \( i + 1 \) indicate the current and next Newton’s step. \( J^{(i)} \equiv \partial R^{(i)}/\partial z^{(i)} \) is the Jacobian matrix. The iteration continues until \( \| R^{(i)} \|_2 < 10^{-8} \).

8.2.3 Limit flow states in two-layer tensioned-web-over-slot die coating flow

In this study, we identify five flow features that may lead to coating defects or degraded product qualities — weeping, mid-gap invasion, bead breakup, upstream feed slot vortex and downstream feed slot vortex.

An important parameter to be set is the downstream wrap angle \( \theta_{w,d} \) indicated in Fig. 8.2(b). When \( \theta_{w,d} \) is too large, the downstream static contact line will wet the die shoulder leading to a non-uniform downstream meniscus. The opposite case, too small \( \theta_{w,d} \), will create a diverging channel between the die lip and the moving web near the meniscus that may cause a large adverse pressure gradient. This pressure gradient can cause a ribbing instability that destroys cross-web coating uniformity. Here, we set \( \theta_{w,d} = \theta_{L,d} \) by adjusting the \( x \)-coordinate of the downstream web extreme.
Figure 8.4: Change of flow states as wet thickness changes for base die lip configuration. Tension number is set to $N_T = 3.14 \times 10^{-4}$. $h_{tw}$ and $h_{bw}$ are top-layer and bottom-layer wet thicknesses, respectively.
8.2. ELASTOHYDRODYNAMIC MODEL AND SOLUTION METHOD

point. At a fixed die geometry, wrapping angles and tension number $N_T$, the flow in the coating bead is a function of the flow rate (or wet thickness) of both layers.

Figure 8.4 presents the evolution of the flow states as the top-layer and/or bottom-layer change. The range of desirable wet thickness for both layers, $h_{b,w}$ and $h_{t,w}$, is limited by the different flow states that lead to coating defects. At high bottom layer thickness $h_{b,w}$, the upstream static contact point reaches the end of the upstream corner of the die lip and liquid may drip, losing the pre-metered action of the method, a phenomenon referred as *weeping* (Fig. 8.4(e)). As $h_{b,w}$ decreases, the contact point moves toward the upstream feed slot and a vortex inside the upstream feed slot (Fig. 8.4(c)) will appear when the thickness is below a certain critical value. Below another critical value for $h_{b,w}$, the interlayer separation point can travel from the downstream corner of the mid lip to the mid lip surface, the event called *mid-gap invasion* (Fig. 8.4(a)). Also at low $h_{b,w}$, the dynamic contact point can invades the upstream feed slot leading to cross web non-uniformities and ultimately to *bead breakup* (Fig. 8.4(b)).

The flow in the coating bead is also a function of the top layer flow rate. Above a critical top layer thickness, the interlayer separation point moves along the mid lip, leading also to mid-gap invasion. Depending on the value of the bottom layer thickness, the high top layer thickness can cause weeping of the upstream meniscus before mid-gap invasion occurs. On the other extreme, if the top layer thickness falls below a critical value, a vortex inside the upstream feed slot can be observed.

8.2.4 Construction of operating window and vortex-free operating window

It is important to map not only the parameters at which the upstream contact point reaches both extremes of the upstream die lip and the interlayer separation point arrives at the mid lip surface, but also the conditions at which recirculations appear inside the flow. Vortices can lead to undesired effects on the final coated product, such as cross-web non-uniformities, due to a nodular structure of the vortex, or point defects, due to unwanted polymerization or particle aggregation because of the long
residence time.

The critical parameters at which undesired flow states occur can be found computationally when the model is combined with the mathematical conditions associated with the onset of these flow states. The onset conditions for weeping and bead breakup can be formulated using the upstream meniscus location [Nam and Carvalho (2009a)]:

\[
\begin{align*}
\gamma_{SCP} &= \gamma_{uc,u} & \text{for weeping,} \\
\gamma_{DCP} &= \gamma_{uc,uf} & \text{for bead breakup,}
\end{align*}
\]  

where \( \gamma_{SCP}, \gamma_{DCP}, \gamma_{uc,u} \) and \( \gamma_{uc,uf} \) are the \( y \)-coordinates of the upstream static contact point, dynamic contact point, the upstream corner of upstream die lip and the upstream corner of the upstream feed slot exit, respectively.

Similarly, the onset of mid-gap invasion can be expressed using \( y \)-coordinate of the downstream extreme of the mid lip surface \( y_{dc,m} \):

\[
\gamma_{ISP} = y_{dc,m} \quad \text{for mid-gap invasion,}
\]

where \( \gamma_{ISP} \) is the \( y \)-coordinates of the interlayer separation point.

Both feed slot vortices in the coating flow are detached from the feed slot wall. This type of vortex birth was analyzed in detail by Nam et al. (2009) and they show that the vortex occur when the shear rate and its derivative with respect to the arc length along the wall vanish.

\[
\begin{align*}
\dot{\gamma}_{tn}(s^*_t) &= 0 \\
\frac{\partial \dot{\gamma}_{tn}}{\partial s_t}(s^*_t) &= 0,
\end{align*}
\]

where \( \dot{\gamma}_{tn} \) is shear stress and \( s^*_t \) is the arc-length coordinate along the solid surface at the vortex birth point.

[Nam and Carvalho (2009a)] proposed an efficient way to construct the coating window for the single-layer TWOSD using the direct tracking method proposed by Nam et al. (2009). It is a multiparameter continuation method that can be used to track the limit flow states. Here, we use the same method to construct the vortex-free operating window for the dual slot TWOSD. The crucial point of the method is to solve an augmented system consisting of the original Navier–Stokes system for free surface
flow and tensioned web plus the conditions that defines the flow feature being tracked, Eqs (8.22) – (8.25). The augmented set of non-linear algebraic equations are solve by Newton’s method. The iteration to solve them proceeds until the $L^2$ norm of the augmented residual is less than $10^{-6}$. Note that one of the additional unknowns for this augmented Navier–Stokes system is the critical parameter for the onset of a limit flow feature. Combining the augmented system and a multi-parameter continuation, the critical curve that defines the onset of limit flow states can be constructed. Here, we use a tangent predictor (Seydel 1994) for the continuation.

Figure 8.5 shows an example of an operating window as a function of the dimensionless top-layer wet thickness $h^*_{t,w}$ and bottom-layer wet thickness $h^*_{b,w}$ at a fixed die geometry, liquid properties, web speed and line tension. The path of the continuation procedures used to construct the lines that define the bead breakup, mid-gap invasion, weeping and birth of vortex inside both feed slots are shown in the plot. First, the onset of mid-gap invasion curve (I) was tracked using the direct tracking method explained previously. During the mid-gap invasion tracking, the flow encounters four limit features, bead breakup, both feed slots vortex and weeping. Selecting each limit feature

**Figure 8.5:** Construction of vortex-free operating window for base die lip configuration using direct tracking of flow features.
flow state as an initial condition for each direct tracking of flow features, the critical
curves (II), (III), (IV) and (V) were constructed. The area bounded by (I), (III), (IV)
and (V) — shaded area in the plot — corresponds to the set of parameters at which
the coating flow is feasible (without weeping, mid-gap invasion) and free of vortices.

8.3 Result & Discussion

8.3.1 Minimum thickness in two-layer tensioned-web-over-
slot die coating

The minimum wet thickness that can be coated without defects at a given operating
condition is one of the most important information need in the decision process of
selecting a coating method. Since the dual slot coating method deals with two liquids,
there are two minimum wet thickness to be considered. The bottom-layer minimum
wet thickness is related to the upstream meniscus location — bead breakup. The bead
breakup curve (II) in Fig. 8.5 shows that the bead breakup is virtually independent
to top-layer flow rate: for the set of parameters used, the event occurs at $h^*_{b,w} \sim 6.1 \times 10^{-4}$. However, we could not find the minimum wet thickness for the top-layer
during computation. Figure 8.6 shows the flow states evolution as the top-layer wet
thickness falls. From (i) to (iii), the interlayer moves toward the downstream die
lip and the interlayer separation point climb up along the downstream corner of the
mid lip. When the wet thickness is smaller than the critical top-layer wet thickness
for the downstream feed slot vortex birth, as flow state (ii) in Fig. 8.6 a vortex is
detached from the downstream wall of the feed slot near the upstream corner of the
downstream die lip. The size of separation bubble that contains the vortex grows
as the thickness decreases, as shown by the evolution from flow state (ii) to (iii).
The top layer thickness continuation stopped at flow state (iii) due to the high mesh
distortion.

Up to this point, we could not find any physical event, like the interlayer separation
point invasion into the downstream feed slot, as claimed by Nakama and Chin (1999),
that defines the minimum top-layer wet thickness. In our opinion, this is a limitation
8.3. RESULT & DISCUSSION

**Figure 8.6:** Finding minimum top-layer wet thickness at given bottom-layer wet thickness. $h_{t,w}$ continuation was performed from $h_{t,w}^* = 3.15 \times 10^{-3}$ to $7.38 \times 10^{-5}$ at $h_{b,w}^* = 4.72 \times 10^{-3}$. (i), (ii) and (iii) was computed at $h_{t,w}^* = 3.25 \times 10^{-3}$, $8.35 \times 10^{-4}$ and $7.38 \times 10^{-5}$, respectively.
of the two-dimensional model. The minimum top layer thickness that can be coated may be defined by a three-dimensional event. The 2-D flow may become unstable to cross web perturbation and the separation point location may vary in the cross web direction, leading to non-uniform coating and eventually to discontinuous top layer coating. Another event that may define the minimum top layer wet thickness comes from the large size of the separation bubble, as shown in insert (iii) of Fig. 8.6. The vortex may become nodular along cross web direction and some parts of the separation bubble may touch the interlayer, disrupting the uniform coating.

### 8.3.2 Mid-gap invasion in two-layer tensioned-web-over-slot die coating

As in the fixed-gap dual slot coating, the interlayer separation point does not always dwell on the downstream corner of the mid lip. When the separation point is located on the mid lip, the interlayer and top liquid layer invade the mid-gap region in a phenomenon *mid-gap invasion*. Nam and Carvalho (2009b) showed that mid-gap invasion in fixed-gap slot coating can lead to undesired flow states with micro vortices and periodic oscillation which can cause coating defects. They also showed that a simple criterion can be used to define the operating conditions at which mid-gap invasion occurs: the bottom layer thickness needs to be smaller than $1/3$ of the coating gap. In tensioned-web coating, the coating gap is not an independent process parameter. The web position is determined by the elastohydrodynamic interaction between the coating liquid and the tensioned web. In this case the simple criterion proposed by Nam and Carvalho (2009b) is not valid.

Furthermore, unlike in fixed-gap coating method, we could not found any micro vortices under the mid lip during the computations. Moreover, the mid-gap invasion is a function not only of the bottom layer flow rate but also of the top layer flow rate. Mid-gap invasion flow states can be reached by increasing the top-layer flow rate or decreasing bottom-layer flow rate as shown in Fig. 8.7 (a). Figure 8.7 (b) and (c) represent the location of the interlayer separation with respect to both $h_{t,w}$ and $h_{b,w}$. When the point pass through the patched point between the downstream corner of the mid lip and the mid lip surface, the location of the separation point becomes
8.3. RESULT & DISCUSSION

Figure 8.7: Mid-gap invasion analysis for two-layer TWOSD. plot (a) shows the two different flow rate continuations that go across the critical mid-gap invasion curve for base case die configuration. The location of interlayer separation point was tracked during the continuations. Plot (b) and (c) show the point with respect to $h_{t,w}$ and $h_{b,w}$ continuations, respectively.
very sensitive with respect to both $h_{t,w}$ and $h_{b,w}$. As discussed in Nam and Carvalho (2009b), this high sensitivity may bring coating defects, because small fluctuations that comes from the liquid feeding system will be amplified to a large movement of the separation point that will result nonuniform interlayer configuration.

Figure 8.8 shows the evolution of pressure profile along the coating bead as the top-layer flow rate (a) and the bottom-layer flow rate (b) vary. As the top layer flow rate rises, the pressure near the downstream feed slot remains almost constant, but the gap between the web and the coating die becomes larger. This larger gap leads to smaller pressure near the upstream feed slot, as shown in Fig. 8.8(a), and consequently higher adverse pressure gradient under the mid lip. If the adverse pressure gradient is above a critical value, the separation point is pushed upstream, leading to mid-gap invasion.

As the bottom flow rate falls, the gap near the upstream feed slot becomes smaller and a converging channel under the mid lip is formed. Consequently, the adverse pressure gradient becomes stronger, as shown in Fig. 8.8(b). Again, if the adverse pressure gradient is strong enough, the separation point is pushed upstream, leading to mid-gap invasion.

Since the gap height is determined by the elastohydrodynamic interaction, both the top-layer and bottom-layer flow rates play an important role in changing pressure profile near the mid lip. Hence, the location of interlayer separation point is controlled by both flow rates. This is a major difference between the TWOSD coating and the fixed-gap coating in which only the bottom-layer flow rate plays a key role in the mid-gap invasion.

8.3.3 Effect of operating conditions on vortex-free operating window

As discussed previously, at a fixed die geometry, web tension and velocity and liquid properties, the operability window and vortex-free window of two-layer TWOSD coating can be presented as a function of the top and bottom layer flow rates (or wet thickness).
8.3. RESULT & DISCUSSION

![Graph showing pressure profile changes during different conditions](image)

**Figure 8.8:** Plot (a) and plot (b) show pressure profile changes during $h_{t,w}$ continuation and $h_{b,w}$ continuation, respectively. Solid lines are the pressure profile at the critical mid-gap invasion.
8.3. RESULT & DISCUSSION

The flow in the coating bead and the menisci positions are a function of the balance between initial, pressure forces and normal stress component that comes from the curved substrate under tension. Therefore, as this balance is modified by changes in the operating conditions, the process windows will also change. In this section we discuss how different liquid properties and process conditions affect the coating window, shown in Fig. 8.5.

Effect of viscosity ratio

The coating window was computed at three different viscosity ratio, \( m = \mu_t / \mu_b = 0.5, 1 \) and \( 2 \), to show the effect of this parameter ratio on the operating window of the process (Fig. 8.9). In the results shown, the bottom layer viscosity was kept constant. The results show that the vortex-free operating window shrinks and is shifted down in the plot, i.e. toward thinner top-layer wet thickness, as the top-layer viscosity \( \mu_t \) rises. The insert presented in Fig. 8.9(b) shows that the viscosity of the top layer has little effect on the upstream feed slot vortex birth.

Direct tracking of the different limit flow states, presented in Fig. 8.10, explicitly shows the effect of the viscosity ratio on the critical wet thickness for each coating failure.

Figure 8.10(a) shows how the critical top layer thickness (at a fixed bottom layer thickness) and the critical bottom layer thickness at the onset of mid-gap invasion vary with the viscosity ratio. As discussed before, at a given set of conditions, mid-gap invasion occurs as the top layer thickness rises or the bottom layer thickness falls. At high viscosity ratio, i.e. high top layer viscosity, the pressure near the downstream feed slot rises and so does the adverse pressure gradient along the mid lip consequently, as shown in Fig. 8.11. At fixed flow rates, mid-gap invasion may occur just by raising the viscosity of the top layer. Another way to look is that mid-gap invasion occurs at lower values of the top layer flow rate and higher value of bottom layer flow rate as the viscosity rises.

The effect of the viscosity ratio on the critical conditions at bead breakup is shown in Fig. 8.10(b). The minimum bottom layer thickness necessary to avoid bead breakup
Figure 8.9: Effect of viscosity ratio $m = \mu_t/\mu_b$ on vortex-free operating windows for base die lip configuration. Note that the bead breakup flow state could not be obtained for $m = 0.5$ due to high mesh distortion near the upstream meniscus.
8.3. RESULT & DISCUSSION

Figure 8.10: Effect of viscosity ratio on mid-gap invasion (a), bead breakup (b), downstream feed slot vortex birth (c) and upstream feed slot vortex birth (d). Note that “$h_{b,w}$ continuation” solution branch in (b) is obtained from the arc-length continuation from the bead breakup flow status at $m = 2.78$. 

\[
Wet \text{ thickness } [h^*_t,w (= h_{t,w}/R_d) \text{ or } h^*_b,w (= h_{b,w}/R_d) \times 10^{-3}]
\]

\[
Viscosity \text{ ratio } [m (= \mu_t/\mu_b)]
\]

\[
\text{Increase } h_{t,w} \text{ cause mid-gap invasion}
\]

\[
\text{Decrease } h_{b,w} \text{ cause mid-gap invasion}
\]

\[
\text{Critical mid-gap invasion } h^*_{t,w} \text{ at } h^*_b,w = 3.15 \times 10^{-3}
\]

\[
\text{Before bead breakup}
\]

\[
\text{Bead breakup at } h^*_{t,w} = 6.30 \times 10^{-4}
\]

\[
\text{Stop direct tracking due to high mesh distortion}
\]

\[
\text{After bead breakup}
\]

\[
\text{With downstream feed slot vortex}
\]

\[
\text{Without downstream feed slot vortex}
\]

\[
\text{With upstream feed slot vortex}
\]

\[
\text{Without upstream feed slot vortex}
\]

\[
\text{Upstream feed slot vortex birth at } h^*_{t,w} = 1.57 \times 10^{-3}
\]
Figure 8.11: Change of pressure profile along the moving web near bead breakup condition as viscosity ratio increases for base die lip configuration.
8.3. RESULT & DISCUSSION

rises as the top layer viscosity increase. The reason is that the thickness of the top layer below the downstream die lip at a fixed flow rate becomes larger as that layer becomes more viscous. Consequently, the web is pushed away from the die favoring the invasion of the upstream meniscus.

Figure 8.10(c) shows that at a constant set of flow rates the vortex at the top layer feed slot can be eliminated by raising its viscosity. As the liquid becomes more viscous, the pressure gradient necessary to cause a flow reversal that leads to a recirculation rises. Therefore, the critical thickness at which a vortex in the feed slot is born falls as the liquid becomes more viscous, as shown in Fig. 8.12.

Apparently, the effect of top-layer viscosity on the upstream feed slot vortex is weak, because the bottom-layer viscosity is fixed in the results presented here (Fig. 8.10(d)).

---

**Figure 8.12:** Critical pressure profile at the downstream feed slot vortex birth from the downstream feed slot wall to the downstream die lip surface for base die lip configuration.
8.3. RESULT & DISCUSSION

As in the single-layer TWOSD, the feed slot vortex birth can be delayed by increasing inertia or decreasing viscous force, i.e. increasing bottom-layer Reynolds number $N_{Re,b} = \rho_b h_{b,w} U_w/\mu_b$. Therefore one can expect that the vortex birth may be delayed by decreasing the bottom-layer viscosity, which was not explored in this study.

Effect of web tension

Since the method exploits elastohydrodynamic interaction, the pressure inside the coating bead is proportional to the web tension. At a constant wrapping angle and die geometry, lower web tension corresponds to smaller normal force. Therefore the coating bead become less pressurized at a given flow rate by lowering the web tension. The smaller bead pressure favors towards the invasion of the upstream free surface that leads to bead breakup.

Figure 8.13 shows the effect of web tension on the operating window and on the vortex-free window. As tension number $N_T = \mu_b U_w/T$ increases, bead breakup, mid-gap invasion, and weeping curves shift to thicker bottom-layer wet thickness regime. However, the both vortex birth curves virtually are not affected by the web tension.

The effect of the web tension, represented as the tension number $N_T$, on the critical thickness conditions at mid-gap invasion bead breakup and feed slot vortices is shown in Fig. 8.14.

As discussed before, at a fixed bottom layer thickness $h_{w,b}$, mid-gap invasion occurs as the top layer thickness rises above a critical value. As shown in Fig. 8.14(a), this critical value falls as the web tension falls, i.e. as the tension number $N_T$ rises. In the same way, at a fixed top layer thickness, mid gap invasion occurs as the top layer falls below a critical value. As also shown in Fig. 8.14(a), this critical value rises with tension number. As the web tension falls, the gap between the die and the substrate becomes larger, as shown in Fig. 8.15 favoring mid-gap invasion.

Web tension also affects the bead breakup phenomenon by changing the pressure field near the upstream meniscus. It falls as the web tension decreases. To compensate the lower pressure, a higher flow rate is needed to push the upstream meniscus away
8.3. RESULT & DISCUSSION

Figure 8.13: Effect of tension $T$ on vortex-free operating windows for base die lip configuration.
8.3. RESULT & DISCUSSION

Wet thickness $h^*_{t,w} = h_{t,w}/R_d$ or $h^*_{b,w} = h_{b,w}/R_d \times 10^3$

Tension number $N_T = \mu_b U_w / T \times 10^4$

Increase $h_{t,w}$ causes mid-gap invasion
Decrease $h_{b,w}$ causes mid-gap invasion

Critical mid-gap invasion $h^*_{t,w}$ at $h^*_{b,w} = 3.15 \times 10^{-3}$

Critical mid-gap invasion $h^*_{b,w}$ at $h^*_{t,w} = 3.15 \times 10^{-3}$

Increase $h_{t,w}$ causes bead breakup
Decrease $h_{b,w}$ causes bead breakup

Bead breakup at $h^*_{t,w} = 6.30 \times 10^{-4}$

Without downstream feed slot vortex
With downstream feed slot vortex

Downstream feed slot vortex birth at $h^*_{b,w} = 3.15 \times 10^{-3}$

Without upstream feed slot vortex
With upstream feed slot vortex

Upstream feed slot vortex birth at $h^*_{t,w} = 1.57 \times 10^{-3}$

Figure 8.14: Effect of tension $T$ on mid-gap invasion (a), bead breakup (b), downstream feed slot vortex birth (c) and upstream feed slot vortex birth (d).
8.3. RESULT & DISCUSSION

Figure 8.15: Change of pressure profiles, web curvatures, and web profiles as web tension increase for base die lip configuration. Note that data are obtained from the critical mid-gap invasion solutions specified at Fig. 8.14 (a).
from the feed slot, as shown in Fig. 8.14(b).

Figures 8.14(c) and (d) show that the effect of the web tension on the feed slot vortex birth is insignificant. Note that the modified Reynolds number $N_{Re}^m$ is set to 331.3 for these results. Since the viscous and inertia forces do not change significantly, the vortex birth is virtually the same for different web tension.

**Effect of Web speed**

Tension number can also be adjusted by web speed. However, the web speed also changes the Reynolds number. Figure 8.16 shows the effect of web speed on the operating windows. Similarly to lowering the line tension, raising the web speed pushes the weeping and the bead breakup limits to higher bottom layer thickness, and the mid-gap invasion limit to higher bottom layer thickness and lower top layer thickness. The main difference when compared to the effect of the web tension is that the web speed does change the critical condition for vortex birth inside the upstream and downstream feed slots. As discussed before, as the web speed is raised at constant wet thickness, the flow rate of each layer also needs to be raised. Therefore, the inertial force of the liquid inside the feed slots is stronger and a larger adverse pressure gradient is needed to cause flow reversal and vortex birth. Consequently, vortex birth occurs at lower wet thickness, as shown in Fig. 8.16.

Direct tracking of limit flow states, shown in Fig. 8.17, explicitly shows the effect of the web speed. The critical mid-gap invasion (a) and bead breakup (b) curves are almost identical to Fig. 8.14 (a) and (b). Clearly, these flow limit states are controlled by the tension number. High web speed causes large viscous force and it requires higher curvature of the web to yield larger normal stress resultant for balancing force.

However, the critical wet thicknesses for both vortex births, Figs. 8.17(c) and (d), decrease as the web speed rises. Figure 8.18 (a) and (b) are re-plots of vortex birth direct-tracking results in terms of Reynolds number of each layer and dimensionless wet thickness in unit of feed slot height. The critical upstream feed slot vortex curve (b) is similar to the single-layer TWOSD, as reported by Nam and Carvalho (2009a). As in the single layer case, the critical dimensionless bottom-layer wet thickness is
8.3. RESULT & DISCUSSION

Figure 8.16: Effect of web speed $U_w$ on vortex-free operating windows for base die lip configuration. Note that $U_w$ can adjust tension number $N_T$ and modified Reynolds number $N_{Re}^m$ at the same time.
8.3. RESULT & DISCUSSION

<table>
<thead>
<tr>
<th>Wet thickness [ h^*<em>{t,w} = (h</em>{t,w} / R_d) \times 10^3 ]</th>
<th>Tension number [ N_T = (\mu_b U_w / T) \times 10^4 ]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Increase ( h^*_{t,w} ) cause mid-gap invasion</td>
<td></td>
</tr>
<tr>
<td>Decrease ( h^*_{t,w} ) cause mid-gap invasion</td>
<td></td>
</tr>
<tr>
<td>Critical mid-gap invasion ( h^*_{t,w} = 3.15 \times 10^{-3} )</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Modified Reynolds number [ N_{Re}^{m} = (\rho_b R_d U_w / \mu_b \times 10^{-4}) ]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Without downstream feed slot vortex</td>
</tr>
<tr>
<td>With downstream feed slot vortex</td>
</tr>
<tr>
<td>Without upstream feed slot vortex</td>
</tr>
<tr>
<td>With upstream feed slot vortex</td>
</tr>
</tbody>
</table>

**Figure 8.17:** Effect of web speed \( U_w \) on mid-gap invasion (a), bead breakup (b), downstream feed slot vortex birth (c) and upstream feed slot vortex birth (d). Note that \( U_w \) will change both tension number \( N_T \) and modified Reynolds number \( N_{Re}^{m} \).
8.3. Result & Discussion

Top-layer wet thickness \( h + t, \omega ( = h_{t, \omega} / H_{d, f}) \)

Top-layer Reynolds number \( N_{Re} (= \rho \frac{h_{t, \omega} U_w}{\mu_t}) \)

Without downstream feed slot vortex

With downstream feed slot vortex

Bottom-layer wet thickness \( h + b, \omega ( = h_{b, \omega} / H_{u, f}) \)

Bottom-layer Reynolds number \( N_{Re} (= \rho_b \frac{h_{b, \omega} U_w}{\mu_b}) \)

Without upstream feed slot vortex

With upstream feed slot vortex

Figure 8.18: Re-plot of Fig. 8.17 (c) and (d) using dimensionless wet thickness in unit of feed slot width and Reynolds number.

about \( h_{b, \omega}^{b} = 0.15 \) for the range of Reynolds number explored. The downstream feed slot vortex birth, however, is more sensitive to Reynolds number (Fig. 8.18(a)). The critical ratio between the wet thickness and the feed slot height for the top layer falls from \( h_{t, \omega} / H_{d, f} \approx 0.12 \) to 0.05 as the Reynolds number is raised from \( N_{Re} \approx 0.6 \) to 1.5. In sum, it is possible to coat top layer thinner than bottom layer without invoking feed slot vortex. Moreover, higher production rate, i.e. high web speed, helps in terms of preventing the downstream feed slot vortex.

8.3.4 Effect of die lip geometry on operating window

In a tensioned-web-over-slot die (TWOSD) coating, the pressure is directly related to the web curvature and consequently to the die lip geometry, the geometry may be changed to optimize the process and change the operating window, especially when flow rates are small. There were several attempts to improve die design, but most of them are based on intuition from experiments or simple reasoning ([Chino et al., 1989] [Tomaru, 1995] [Takahashi and Shibata, 1995]).

In this study, the effect of die lip geometry was analyzed by varying four different parameters: the downstream die lip radius of curvature \( R_d \), the mid die lip radius of
8.3. RESULT & DISCUSSION

List of die lip configurations:

- BC
- LRd
- SRd
- LRm
- SRm
- DLu
- MLa

Nam and Carvalho (2009d) proposed an efficient way to construct the coating window for the two-layer TWOSD using the direct tracking method based on Nam et al. (2009c)'s method. It is a multiparameter continuation method that can be used to track particular flow states. Using the tracking method, they created the operating windows in a plane of the top and bottom layer wet thicknesses for different operating conditions, such as viscosity ratio, tension number, and Reynolds number. Here, we use the same method to construct the operating windows for different die design parameters, namely downstream lip radius, mid lip radius, downstream lip offset and mid lip apex point. Unlike operating parameters, the geometry continuation is not trivial to perform. We will describe in detail each geometry continuation details in the following section.

3. Result & Discussion

3.1. Effect of die lip geometry on operating window

In a tensioned-web-over-slot die (TWOSD) coating, the curvature of web is adjusted such that the normal stress resultant from the tensioned web is balanced by the pressure inside the coating bead. The die lip shape is one way to control the curvature of the moving web, especially when flow rates are small. Proper die lip design can enlarge the operability limits of the process.

In this study, the effect of die lip geometry was analyzed by varying four different parameters: the downstream die lip radius of curvature $R_d$, the mid die lip radius of curvature $R_m$, the downstream die lip underbit $H_{d,u}$, and the mid lip apex point location $l_{m,a}$. By adjusting these four parameters, seven different die lip configurations were studied. They are sketched in Fig. 2 and their geometric parameters are listed in Tab. 3.

3.1.1. Effect of downstream die lip radius

As claimed by Chino et al. (1989), improper choice of die lip radii may bring coating defects. Here, we consider three different downstream die lip configurations (BC, LRd, SRd) in order to study the effect of downstream die lip curvature, as sketched in Fig. 3. However, when the downstream web extreme point $x_{d,w}$ is fixed, changes in the lip curvature will bring excessive diverging or converging channel near the die lip exit. This will change pressure profile inside the downstream coating bead and consequently critical operating parameters. In order to decouple this channel shape effect

Figure 8.19: Die lip configurations considered in this study. BC, LRd, SRd, LRm, SRm, DLu and MLa stand for base case lip, large downstream die lip radius, small downstream die lip radius, large mid die lip, small mid die lip, downstream die lip underbite, mid lip with apex point in the middle configurations, respectively. Big dot point on the mid lip stands for the mid lip apex point. $R_d$ and $R_m$ are downstream die lip and mid die lip radii. $H_{d,u}$ and $l_{m,a}$ are downstream underbite depth and mid lip apex point location, respectively. All die lip configuration has the same upstream die lip radius. See Table 8.3 for geometric parameters details.
8.3. RESULT & DISCUSSION

<table>
<thead>
<tr>
<th>Name</th>
<th>$R_d$ (&quot;&quot;ths)</th>
<th>$R_m$ (&quot;&quot;ths)</th>
<th>$H_{d,u}$ (µm)</th>
<th>$l_{m,a}$ (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BC</td>
<td>0.250&quot;</td>
<td>0.250&quot;</td>
<td>0.0 µm</td>
<td>0.0 mm</td>
</tr>
<tr>
<td>LRD</td>
<td>0.375&quot;</td>
<td>0.250&quot;</td>
<td>0.0 µm</td>
<td>0.0 mm</td>
</tr>
<tr>
<td>SRD</td>
<td>0.167&quot;</td>
<td>0.250&quot;</td>
<td>0.0 µm</td>
<td>0.0 mm</td>
</tr>
<tr>
<td>LRM</td>
<td>0.250&quot;</td>
<td>0.375&quot;</td>
<td>0.0 µm</td>
<td>0.0 mm</td>
</tr>
<tr>
<td>SRM</td>
<td>0.250&quot;</td>
<td>0.167&quot;</td>
<td>0.0 µm</td>
<td>0.0 mm</td>
</tr>
<tr>
<td>DLU</td>
<td>0.250&quot;</td>
<td>0.250&quot;</td>
<td>10.0 µm</td>
<td>0.0 mm</td>
</tr>
<tr>
<td>MLA</td>
<td>0.250&quot;</td>
<td>0.250&quot;</td>
<td>0.0 µm</td>
<td>0.5 mm</td>
</tr>
</tbody>
</table>

curvature $R_m$, the downstream die lip offset $H_{d,u}$, and the mid lip apex point location $l_{m,a}$. By adjusting these four parameters, seven different die lip configurations were studied. They are sketched in Fig. 8.19 and their geometric parameters are listed in Tab. 8.3.

Effect of downstream die lip radius

As claimed by Chino et al. (1989), improper choice of die lip radii may bring coating defects. Here, we consider three different downstream die lip configurations (BC, SRD, and LRD) in order to study the effect of downstream die lip curvature, as sketched in Fig. 8.20. However, when the downstream web extreme point $x_{d,w}$ is fixed, changes in the lip curvature will bring excessive diverging or converging channel near the die lip exit. This will change pressure profile inside the downstream coating bead and consequently critical operating parameters. In order to decouple this channel shape effect from the die lip curvature, $x_{d,w}$ is adjusted such that the downstream wrap angle $\theta_{d,w}$ has the same value as downstream die lip departure angle $\theta_{L,d}$, as sketched in Fig. 8.20. The operability and vortex birth limits obtained with each of these three configurations is shown in Fig. 8.21. For comparison purpose, we choose
8.3. RESULT & DISCUSSION

Figure 8.20: Downstream lip radius change scheme. In order to minimize the effect from converging or diverging channel near the downstream die lip and the moving web, the downstream web extreme point \( x_{d,w} \) needs to be adjusted to \( x_{d,w,s} \) or \( x_{d,w,l} \) that depends on the downstream lip radius \( R_d \).
the downstream die lip radius of the BC configuration, $R_{d,\text{base}}$ as the characteristic length unit. As discussed by Nam and Carvalho (2009c), the vortex-free operability limit is bounded by different undesired flow states, such as weeping, that occurs at high top and bottom layer wet thickness, mid-gap invasion, that occurs at high top layer wet thickness, downstream feed slot vortex, that occurs at low top layer thickness, upstream feed slot vortex and bead breakup, that occurs at low bottom-layer thickness.

As the radius of the die lip decreases, the vortex-free window shrinks. The mid-gap invasion and the weeping curves are changed significantly. Like in single-layer case, higher curvature of the web leads to higher pressure in the downstream coating bead. Therefore, even a small flow rate is enough to push the meniscus upstream causing weeping phenomena. However, the lip radius does not change the bead breakup and the downstream feed slot vortex birth curves. Note that the model fails to capture bead breakup phenomenon at the small lip radius due to a high mesh distortion, as shown in Fig. 8.21 (b).

Direct tracking of limit flow states, shown in Fig. 8.22, explicitly represents the effect of the downstream die lip radius on each limiting flow state.

When the lip radius decreases, i.e. the curvature increases, the region near the downstream feed slot becomes more pressurized, as shown in Fig. 8.23. Therefore, for smaller downstream die lip radius $R_d$ and at fixed bottom-layer wet thickness $h_{b,w}$, a small value of the top-layer wet thickness $h_{t,w}$ is enough to create a large adverse pressure gradient near the downstream corner of the mid lip that pushes the interlayer separation point to the mid lip surface.

According to Fig. 8.22 (b), bead breakup is not significantly affected by the downstream die lip radius. As flow rates fall, the shape of the moving web becomes similar to the shape of die lip surfaces. Since, at bead breakup flow state, the upstream meniscus is close to the mid lip, the pressure field is mostly controlled by the mid lip radius, not the downstream lip radius.

Similar to single-layer TWOSD coating flow, the lip radius does not have an impact on the downstream feed slot vortex. However, the bottom-layer wet thickness $h_{b,w}$
Figure 8.21: Effect of downstream lip radius on vortex-free operating windows by comparing three different configurations: BC, LRd and SRd. Note that SRd configuration could not find bead breakup flow state due to high mesh distortion.
8.3. RESULT & DISCUSSION

Figure 8.22: Effect of downstream lip radius on mid-gap invasion (a), bead breakup (b), downstream feed slot vortex birth (c) and upstream feed slot vortex birth (d).
8.3. RESULT & DISCUSSION

for the upstream feed slot vortex is markedly changed when the lip radius is small. Figure 8.24 shows the critical pressure profile at the upstream feed slot vortex birth, along from the upstream feed slot wall and the mid lip surface. The pressure profile for small lip radius reveals that the pressure along the mid lip changes remarkably when $R_d^*$ is small. The change in pressure will enhance the adverse pressure gradient, that accelerates the vortex birth, along the surface of the mid lip near the exit of upstream feed slot.

Effect of mid die lip radius

As the downstream die lip radius, the mid lip radius can also adjust the pressure profile inside the coating bead, especially near the mid lip and upstream lip surface, or more precisely the upstream coating bead. Because the most of the limit flow states is related to the mid lip pressure profile, the proper design of the mid lip geometry is critical. Two variations of the mid lip geometry from the base the base case are

![Diagram](image)

**Figure 8.23:** Change of critical pressure profile along the moving web for the onset of mid-gap invasion condition as the downstream die lip radius $R_d$ changes.
Figure 8.24: Change of critical pressure profile from the upstream feed slow wall to the mid lip surface for the onset of upstream feed slot vortex as the downstream die lip radius $R_d$ changes.
8.3. RESULT & DISCUSSION

**Figure 8.25:** Mid lip radius change scheme. In order to minimize the effect of underbite and overbite of upstream die lip position, the upstream die lip corner of the upstream die lip \( x_{u,d} \) needs to be shifted to \( x_{u,w,s} \) or \( x_{u,w,l} \), and the upstream extreme point web point \( x_{u,w} \) to be shifted to \( x_{u,w,s} \) or \( x_{u,w,l} \), whether mid lip radius \( R_m \) decrease to \( R_{m,s} \) or increase to \( R_{m,l} \).
In order to prevent a misalignment of the upstream and mid die lips as the radius of curvature is changed, the position of the upstream die lip is shifted accordingly, as explained in Fig. 8.25.

As in the previous case, the upstream web extreme point $x_{u,w}$ is also adjusted to eliminate the possibilities of converging and diverging channel near the upstream die lip. Note that in all these configurations, the mid lip apex point is fixed at the downstream corner of the mid lip.

The vortex-free operating window obtained with these three configurations is shown in Fig. 8.26. It expands and shifts toward thicker bottom-layer regime as the mid lip radius rises. The bead breakup, the upstream feed slot vortex birth, and the weeping curves change significantly with the mid lip radius. The mid-gap invasion and the downstream feed slot vortex birth curves are virtually unchanged. As discussed before, the mid lip radius have direct control of the pressure inside the upstream coating bead. Therefore, the limit flow features near the upstream coating bead, like the bead breakup and the upstream feed slot vortex birth, are significantly affected by the mid lip radius.

This argument is confirmed by the direct tracking results, shown in Fig. 8.27 (b) and (d) that present the critical bottom-layer thickness at bead breakup and upstream feed slot vortex birth as a function of mid lip radius. At very small value of mid lip radius, the effect of the lip geometry on the critical thickness at bead breakup is reversed, i.e. smaller lip radius yields larger critical bottom layer wet thickness.

The reason is the inevitable converging channel between the die lips and moving web that is formed when the mid lip radius is very small, as sketched in Fig. 8.25 (a). Figure 8.28 shows the pressure profile along the web at the bead breakup flow states for different $R_m^*$. The pressure peak is remarkably shifted to the middle of the mid lip for the small lip radius, e.g. $R_m^* = 0.51$. This pressure peak shifting explains the channel shape change, leading to large gap height in the upstream section of the mid lip.

The converging channel also affects the upstream feed slot vortex birth. Figure 8.29
8.3. RESULT & DISCUSSION

Figure 8.26: Effect of mid lip radius on vortex-free operating windows by comparing three different configurations: BC, LRm and SRm.
8.3. RESULT & DISCUSSION

![Diagram](image)

**Figure 8.27:** Effect of mid lip radius on mid-gap invasion (a), bead breakup (b), downstream feed slot vortex birth (c) and upstream feed slot vortex birth (d).
Figure 8.28: Change of critical pressure profiles at the bead breakup flow states near the mid lip along the moving web for different mid lip radius $R_m$. Pressure profiles obtained from point (i), (ii), and (iii) in Fig. 8.27(b).
8.3. RESULT & DISCUSSION

Figure 8.29: Change of critical pressure profile from the upstream feed slow wall to the mid lip surface for the onset of upstream feed slot vortex as the downstream die lip radius $R_m$ changes.
8.3. RESULT & DISCUSSION

shows the critical pressure profiles along the feed slot and mid lip surface at the upstream feed slot vortex birth flow conditions. The profile at the small mid lip radius $R_m^* = 0.51$ has a peak in the middle of the mid lip. The adverse pressure gradient that comes after the peak promote a vortex birth near the upstream corner of the mid lip: the vortex can be born at higher bottom-layer flow rate.

Effect of die lip offset (underbite)

In a practical point of view, underbite and overbite die lip configurations are easy to achieve. It can be done by controlling the offset between the die lips, simply pushing or pulling one die lip surface with respect to the others. When the downstream die lip is

![Figure 8.30](image-url)

**Figure 8.30:** Underbite downstream die lip change scheme. In order to prevent diverging channel near downstream coating gap, the downstream web extreme point $x_{d,w}$ shifts to $x_{d,w,u}$, as upstream corner of the downstream die lip $x_{d,u}$ recedes to $x_{d,u,u}$. 

267
8.3. RESULT & DISCUSSION

**Figure 8.31**: Effect of mid lip radius on mid-gap invasion (a), bead breakup (b), downstream feed slot vortex birth (c) and upstream feed slot vortex birth (d).
8.3. RESULT & DISCUSSION

**Figure 8.32:** Effect of underbite downstream die lip on vortex-free operating windows by comparing two different configurations: BC and DLu.
8.3. RESULT & DISCUSSION

![Diagram showing dimensionless pressure and Y coordinate for mid-gap invasion and moving web profile.](image)

**Figure 8.33:** Change of critical pressure profile along the moving web (a) for the onset of mid-gap invasion and the corresponding web profile (b). Pressure and web profiles for (i), (ii) and (iii) are obtained from the critical mid-gap invasion top-layer wet thickness curve of Fig. 8.32 (a).
considered, only a positive offset $H_{d,u}$ (Fig. 8.30) or the underbite configuration helps to widen the operating window. Also most of patents (Tomaru [1995], Takahashi and Shibata [1995]) claimed that properly chosen underbite downstream die lip can prevent coating defects, like streak line(s) or unevenness of the coated layer(s). A negative offset or a overbite configuration will increase the pressure near the downstream feed slot, creating a strong adverse pressure gradient along the mid lip, which will push the interlayer separation point towards the mid lip surface — the onset of mid-gap invasion.

The use of the underbite downstream die lip configuration, however, needs special care. The downstream web extreme point $x_{d,w}$ should be adjusted in order to prevent a diverging channel near the exit of the downstream die lip, as sketched in Fig. 8.30. The diverging channel creates an adverse pressure gradient that may pull the downstream static contact point towards the die lip, which may lead to coating defects. In this study, we shift $x_{d,w}$ in order to match the downstream wrap angle $\theta_{d,w}$ to the downstream die lip departure angle $\theta_{L,d}$.

The positive offset, the underbite configuration, mostly affect the mid-gap invasion and weeping, as shown in Fig. 8.31. The onset of these limiting flow states are delayed to thicker top-layer wet thickness regime. Bead breakup and vortex birth flow conditions are virtually not affected by the underbite configuration.

The direct tracking of limit flow features as a function of the die lip offset, shown in Fig. 8.32 confirms the argument that the offset of the downstream die lip controls primarily the pressure near the exit of the downstream feed slot. Therefore, the effect of offset is mostly related to the mid-gap invasion (a) and slightly related to the downstream feed slot vortex (c). The pressure profiles at the mid-gap invasion conditions, Fig. 8.33 (a), and the web profiles, Fig. 8.33 (b), show why the critical top-layer thickness $h_{t,w}$ rises with the offset value $H_{d,u}$. In order to have the same pressure and web profile near the downstream feed slot exit, a top-layer flow rate is need to fill the larger downstream coating gap.
Effect of mid lip apex point

The apex point of the die lip is another way to control the pressure field inside the coating bead, as discussed in Nam and Carvalho (2009a). In the two-layer case, the mid lip apex point can change the pressure profile along the mid lip, which is critical to mid-gap invasion and weeping. Figure 8.34 shows the mid lip apex point shifting scheme. In order to minimize the effect from the offset of the die lip position for the downstream and upstream lips, both lip apex points are shifted to the mid lip apex point — the three die lips remain on the same circle, dotted lines in Fig. 8.34. Also both web extreme points $x_{d,w}$ and $x_{a,w}$ need to be adjusted to match the both wrap angles, $\theta_{d,w}$ and $\theta_{a,w}$ to the departure and the approach lip angles, $\theta_{L,d}$ and $\theta_{L,u}$, respectively.

By adjusting the mid lip apex point and consequently the pressure profile, the locations of the interlayer separation point and the dynamic contact point are shifted. The critical conditions at mid-gap invasion occur at higher top-layer wet thickness, and the bead breakup and the weeping flow conditions are shifted to thicker bottom-layer wet thickness, as shown in Fig. 8.35. The upstream feed slot vortex birth curve virtually does not change, but the downstream feed slot vortex birth is shifted slightly towards thicker bottom-layer wet thickness regime. Unlike the downstream die lip offset, the effect of apex point grows as the flow rates increases, i.e. thicker top- and bottom-layer wet thickness regime. For lower flow rates, the curvature of the web is virtually the same as the curvature of the die lip, whether the apex point is located at the downstream corner or the upstream corner. Therefore the pressure inside the coating bead should be similar at low flow rates for the different apex point locations.

Figure 8.36 shows the critical conditions at four different limiting flow states as a function of the mid gap invasion. The location of the apex point affect mostly the mid-gap invasion, Fig. 8.36 (a), especially the critical top-layer wet thickness curve, and the bead breakup, Fig. 8.36 (b).

Figure 8.37 (a) shows the pressure profiles at the critical condition of mid-gap invasion at $h_{b,w}^* = 3.15 \times 10^{-3}$. The adverse pressure gradients near the downstream feed
### Table 1: Operating and geometric parameters for base case die lip (BC configuration)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Base case</th>
</tr>
</thead>
<tbody>
<tr>
<td>Upstream lip length (L_u)</td>
<td>mm</td>
<td>16.0</td>
</tr>
<tr>
<td>Downstream lip length (L_d)</td>
<td>mm</td>
<td>15.9</td>
</tr>
<tr>
<td>Upstream web wrap angle (θ_u,w)</td>
<td>deg</td>
<td>9.97</td>
</tr>
<tr>
<td>Downstream web wrap angle (θ_d,w)</td>
<td>deg</td>
<td>16.0</td>
</tr>
<tr>
<td>Upstream lip approaching angle (θ_L,u)</td>
<td>deg</td>
<td>9.97</td>
</tr>
<tr>
<td>Downstream lip departure angle (θ_L,d)</td>
<td>deg</td>
<td>15.9</td>
</tr>
<tr>
<td>Mid lip length (L_m)</td>
<td>mm</td>
<td>10.0</td>
</tr>
<tr>
<td>Downstream lip radius of curvature (R_d,w)</td>
<td>mm</td>
<td>2.5</td>
</tr>
<tr>
<td>Upstream lip radius of curvature (R_u,w)</td>
<td>mm</td>
<td>3.0</td>
</tr>
<tr>
<td>Mid lip radius of curvature (R_m,a)</td>
<td>mm</td>
<td>1.5</td>
</tr>
<tr>
<td>Feed slot heights (H_u,w)</td>
<td>mm</td>
<td>30.0</td>
</tr>
<tr>
<td>Web tension (T_w)</td>
<td>lb/in</td>
<td>100</td>
</tr>
<tr>
<td>Web speed (U_w)</td>
<td>m/sec</td>
<td>250</td>
</tr>
<tr>
<td>Surface tension (σ_b)</td>
<td>dyne/cm</td>
<td>61</td>
</tr>
<tr>
<td>Dynamic contact angle (θ_c,b)</td>
<td>deg</td>
<td>60</td>
</tr>
<tr>
<td>Static contact angle (θ_s,b)</td>
<td>deg</td>
<td>50</td>
</tr>
<tr>
<td>Bottom-layer viscosity (µ_b)</td>
<td>cP</td>
<td>2.3</td>
</tr>
<tr>
<td>Top-layer viscosity (µ_t)</td>
<td>cP</td>
<td>1.5</td>
</tr>
<tr>
<td>Bottom-layer film thickness (h_b)</td>
<td>µm</td>
<td>30</td>
</tr>
<tr>
<td>Top-layer film thickness (h_t)</td>
<td>µm</td>
<td>10</td>
</tr>
<tr>
<td>Density for both layers (ρ)</td>
<td>g/cm³</td>
<td>61</td>
</tr>
<tr>
<td>Ambient pressure (P_amb)</td>
<td>Pa</td>
<td>100</td>
</tr>
</tbody>
</table>

**Figure 8.34:** Mid lip apex point change scheme. Mid lip apex point is presented as \( l_{m,a} \) which is the distance measured from the downstream corner of the mid lip. In order to minimize the effect of underbite and overbite of upstream die lip position, the die lips have the same radius of curvature. Also, both web extreme points, \( x_{d,w} \) and \( x_{u,w} \), shift to \( x_{d,w,a} \) and \( x_{u,w,a} \), correspondingly.
Figure 8.35: Effect of mid lip apex point on vortex-free operating windows by comparing two different configurations: BC and MLa.
8.3. RESULT & DISCUSSION

Figure 8.36: Effect of mid lip apex point on mid-gap invasion (a), bead breakup (b), downstream feed slot vortex birth (c) and upstream feed slot vortex birth (d).
Figure 8.37: Change of critical pressure profile along the moving web (a) for the onset of mid-gap invasion and the corresponding web profile (b). Pressure and web profiles for (i), (ii) and (iii) are obtained from the critical mid-gap invasion top-layer wet thickness curve of Fig. 8.36 (a).
8.4. FINAL REMARKS

slot exit are similar to each other even with different critical $h^*_{r,w}$ for the invasion. However, the pressure gradient near the upstream changes as the apex point changes, as shown in Fig. 8.37 (b). According to the plot, the upstream section of the mid lip becomes more diverging as the apex point shifts towards the upstream corner of the mid lip, $l^*_{m,a} \rightarrow 1$.

The critical feed slot bottom-layer wet thickness $h_{b,w}$ for the bead breakup increases slightly as the apex point shifts to upstream direction. One may expect that the wet thickness decrease as the apex point approaches to the upstream corner of the mid lip, $l^*_{m,a} \rightarrow 1$, because a pressure peak is created due to shifted apex point. However, the wet thickness increases slightly as shown in Fig. 8.36 (b). Figure 8.38 (a) shows the pressure profiles at the bead breakup for three different apex points location. The profiles inside the mid lip are shifted up as $l^*_{m,a} \rightarrow 1$, but the pressure peak does not move significantly. This is due to the wrap angles adjustment scheme, Fig. 8.34, that is intended to prevent excessive diverging and converging channel near the upstream and downstream die lips. Both wrap angles change as the apex point moves, as shown in Fig. 8.38. Since the location of the pressure peak point also depends on how the moving web wraps the die lip surfaces, the wrap angles are also important to control the bead breakup phenomenon, especially the upstream wrap angle $\theta_{u,w}$. However, an excessive wrap angle cause the web to scratch the die lip surface that may lead to damage the lip or tear the web.

8.4 Final remarks

A computational analysis of two-dimensional dual slot two-layer tensioned-web-over-slot die coating flow was carried out by means of Galerkin/finite element method. To obtain an accurate description of the web configuration, thin inextensible shell theory was used. The inter-diffusion zone is approximated as a sharp interface with zero interfacial tension for miscible coating liquids. The accurate interlayer location and the interlayer separation point was predicted by the method proposed by Nam and Carvalho (2009b). The different mechanisms that limit the process were associated to particular flow features. Bead breakup and weeping is directly linked to the position
8.4. FINAL REMARKS

Figure 8.38: Change of critical pressure profile along the moving web for the bead breakup (a) and the wrap angle changes for the apex point continuation (b). Pressure and web profiles for (iv), (v) and (vi) are obtained from the critical bottom-layer wet thickness curve of Fig. 8.36 (b).
of the upstream meniscus. The first occurs when the free surface invades the feed slot and the later when it reaches the end of the upstream die lip. Mid-gap invasion is associated with the position of the separation point. Vortex birth inside the upstream and downstream feed slot is clearly related to a particular flow state.

Deploying direct tracking method, the conditions at which limiting flow states (mid-gap invasion, bead breakup, weeping and vortex birth) occur were computed automatically. In this study, we focused on the effect of operating conditions on the process limits.

The results show the operability and vortex-free windows in the plane of top and bottom layer wet thicknesses at different operating conditions. The pressure gradient in the coating bead controls the different flow states. From a practical stand point, the most important results are that the web tension and web speed change the minimum and maximum flow rates possible, high web speed and low tension increase the minimum possible thickness. Moreover, the vortex inside the downstream feed slot that may lead to coating defects can be avoided by raising the viscosity of the top layer.

The various limiting flow states that may cause coating defects are greatly affected by the pressure profile along the coating. Therefore, the critical conditions that set the operability limits of the process may be shifted to thinner coating by controlling the pressure distribution. Because the pressure is directly related to the web curvature and consequently to the die lip geometry, the later can be optimized. The effect of four different geometric parameters that define the die lips shapes on the critical condition was analyzed here: Downstream lip radius, mid lip radius, downstream lip offset and mid lip apex point.

Each of these geometric features affect the local pressure and consequently the conditions at which the critical flow states occurs. For example, the results show that small downstream lip radius leads to high pressure under the downstream die lip which pushes the separation line towards the mid lip, promoting mid-gap invasion. The results also show that the position of the upstream meniscus, which is directly related to bead breakup, can be shifted upstream by a smaller mid lip radius, leading to thinner coating.
8.4. FINAL REMARKS

In practice, the design of dual slot coating requires special care. For example, too small die lip radius that helps thinner coating requires large wrap angle to prevent ribbing instabilities. In this condition, substantial amount of normal stress resultant from large web curvature may push the web too much against the die surface, which can cause scratching. Also it may cause lateral or cross-web flow from the center to the edge of the die lip due to the extreme pressure between the coating bead and the ambient air. Sometime, the curvature mismatch between die lips cause an inevitable converging or diverging channel between die lips and the moving web, even with proper wrap angle adjustments. Furthermore, when a lip apex point is located at a corner of the feed slots, particles in the coating solution may be trapped near the corner that could create streak lines along the flow direction. For a successful die lip design, these practical limits should also be considered, in addition to the analysis presented in this study.
Chapter 9

Epilogue

The object of this thesis was to analyze tensioned-web-over-slot die (TWOSD) coating flow where the liquid is applied onto the moving substrate under tension. The analysis includes visualization of the coating bead flow using lab-scale coating apparatus and prediction of the flow using the computational model.

One of the important goals in coating flow researches is to find operating window with free of vortex for the given process. Like other slot coating methods, defect-causing flow features for the TWOSD flow are micro recirculations inside the bead flow, bead breakup, weeping, and, for two-layer coating, interlayer separation point movement (mid-gap invasion). In previous works on the coating flows, including the TWOSD coating flow, the window is usually constructed computationally or experimentally by finding undesired flow features inside the coating flow manually. However, it is time-consuming process so that parametric studies on the coating flow were usually limited. Therefore one of the major goals in this study was to propose an efficient method to create such windows automatically.

Since bead breakup and weeping phenomena and their onset conditions are relatively well known for the slot coating flow (Sartor, 1990; Gates, 1999; Musson, 2001) and for the TWOSD slot coating flow (Park, 2008), this thesis focused on micro recirculation inside flow, mid-gap invasion, and interlayer stability. Those flow features were deeply studied computationally and/or experimentally for simpler coating flows.
without elastohydrodynamic interaction. Developed theoretical and computational methods are used for the analysis of the TWOSD coating flow.

The presence of micro recirculations in flows can lead to undesired effects in many industrial processes, including the TWOSD coating. Here, a comprehensive view of vortex birth away from flow boundaries, attached to a liquid/gas interface and attached to a solid wall were presented at Chap. 2. The conditions for vortex birth are expressed in terms of kinematic variables.

In order to compute the critical parameters for the vortex birth efficiently, we developed a computational method to detect or track the birth flow state directly in a generic two-dimensional flow (Chap. 3). In Galerkin finite element context, the vortex birth conditions are expressed as discrete equations. The equations are fully coupled to Navier–Stokes equation to construct augmented Navier–Stokes (NS) system. By solving the augmented system, critical parameter values and the corresponding flow states are obtained simultaneously. The proposed method is extremely efficient compared with post-processing based method that requires checking a large set of solutions manually or semi-automatically. The fixed-gap single-layer slot coating and half-submerged forward roll coating were chosen to show examples of finding vortex-free window for flow process. Note that the proposed method can be combined to other onset of flow features to track critical parameter trajectories in a given parameter space. The extension of the method was shown in Chap. 6 for the single-layer TWOSD coating flow.

Mid-gap invasion is another flow feature that may cause degrading product qualities in two-layer coating. It is related to the location of the interlayer separation point that is the extreme of the interlayer and is attached to the mid die lip surface. When the point moves from the downstream corner of the mid lip to upstream, it is called mid-gap invasion. In Chap. 4, the mid-gap invasion in a fixed-gap two-layer slot coating flow was studied in detail with experiment, computational and theory. The critical operating parameter for onset of the mid-gap invasion turned out to be the bottom-layer flow rate that was identified by visualizing the coating flow. New mesh generation scheme for die lip rounded corners and a zero shear rate for the interlayer separation point boundary condition were proposed to find the accurate the separation
point location. Computational results from two-dimensional Navier–Stokes system combined proposed methods delineate the two different mechanisms that depend on viscosity ratio. Furthermore, a rule of thumb for the onset of the invasion was found by the simplified flow model near the mid die lip: the invasion occurs when the bottom-layer wet thickness is less than $1/3$ of the gap.

Due to the small gap between the die and the high-speed moving web, the interlayer inside the two-layer slot coating suffers enormous shear that may cause unstable interlayer and may create coating defects. In this study, linear stability analysis was done on the approximated rectilinear flow near the downstream die lip to predict the neutral-stability curves that define the region of stable flow as a function of flow rate ratio and viscosity ratio. Exploiting new eigenvalue solving scheme presented in Chap. 3, the stability analysis was done over wide ranges of parameters within reasonable time. The results from the stability analysis were combined with the rule of thumb for the mid-gap invasion to find proper operating conditions.

Based on developed theories and methods mentioned above, both single layer and two layer TWOSD coating flows were analyzed. The computational analysis of two-dimensional TWOSD coating flow was carried out by means of Galerkin finite element method. Free surfaces — both gas/liquid, liquid/liquid and web/liquid interfaces — were tracked by a boundary conforming mesh technique. The technique is based on mapping of unknown flow domain into fixed computational domain and inverse mapping is governed by a system of elliptic differential equations identical to those encountered in the dilute regime of diffusional transport (de Santos 1991). For the accurate description of the web, we used thin inextensible shell theory instead of simple membrane theory (Flügge 1973).

Proposed computational model was validated by flow visualization (Chap. 7). Furthermore, through the visualization, the effect of flow parameters beyond the limits at which the model is valid and to ultimately help the fundamental understanding of the physical mechanism.

Various operating conditions, like web tension, web speed, and viscosity ratio for two layer, affects the possible maximum and minimum wet thickness that are useful guidelines. It turned out that some of die lip design parameters, like radius of curvature and
die lip apex point, has more influence than others in controlling elastohydrodynamic interaction that affect the operating limits.

The analysis done in here assume that the tension is uniform along cross-web direction, i.e. there are no web handling issue. However, it is extremely difficult to achieve in a real industrial process, especially for a thin web. The non-uniformity in the web tension may cause gap variations near the die lip that may spoil cross-web coating uniformity, and sometimes creates dry lanes even in the desirable coating condition suggested by the theoretical model. Also, there are practical limitations on suitable die lip design proposed in this study. For example, too small radius of curvature of die lip requires large wrap angle to prevent ribbing instability at the downstream meniscus. In this condition, substantial amount normal stress resultant from large web curvature may push the web too much against the die surface, with the possibility of scratching. Furthermore, it may cause lateral or cross-web flow from the center to the edge of the die lip due to the extreme pressure difference between the coating bead and the ambient air.

There are several topics can be worth to be investigated further on the TWOSD coating flow, especially for two layer case. For example, the flow feature that leads to the minimum top-layer wet thickness was not identified by two-dimensional steady state model. As mentioned in Chap. 8, the flow feature may be related to three dimensional events and could be signaled by flow instabilities. It may be worthwhile to explore low top-layer flow rate regime in parameter space by flow visualization using lab-scale coater. Also one can perform a two- or three-dimensional linear stability analysis on the two-layer coating flow in order to find a practical limit for the top-layer flow rate or wet thickness with a proper transient model for the thin-inextensible web that extended from shell theory.
Bibliography


BENJAMIN, D. F. 1994 Roll coating flows and multiple roll systems. PhD thesis, University of Minnesota Published by University Microfilms International, Ann Arbor, MI.


BOOTH, G. L. 1970 Coating equipment and processes. Lockwood publishing co., Inc.,


Carvalho, M. S. 1996 Roll coating flows in rigid and deformable gaps. PhD thesis, University of Minnesota. Published by University Microfilms International, Ann Arbor, MI.


Christodoulou, K. N. 1990 *Computational physics of slide coating flow*. PhD thesis, University of Minnesota. Published by University Microfilms International, Ann Arbor, MI.


BIBLIOGRAPHY

HANUMANTHU, R. 1996 Patterned roll coating. PhD thesis, University of Minnesota Published by University Microfilms International, Ann Arbor, MI.


KISTLER, S. F. 1983 The fluid mechanics of curtain coating and related viscous free surface flow with contact line. PhD thesis, University of Minnesota Published by University Microfilms International, Ann Arbor, MI.


LIGHTHILL, M. J. 1964 *Laminar boundary layer (ed. L. Rosenhead)*. Clarendon, chapter Attachment and separation in three-dimensional flow.


MUSSON, L. C. 2001 Two-layer slot coating. PhD thesis, University of Minnesota Published by University Microfilms International, Ann Arbor, MI.


BIBLIOGRAPHY


PARK, E. 2008 Physics of coating tensioned-web over slot die. PhD thesis, University of Minnesota Published by University Microfilms International, Ann Arbor, MI.


PRAUNDL, L. AND TIETJEN, O. G. 1934 Fundamentals of hydro- and aeromechanics. United engineering trustees,
BIBLIOGRAPHY


SCRIVEN, L. E. 2005b Fine-Structured Materials by Continuous Coating and Drying or Curing of Liquid Precursors. In Chemical Engineering Trends and Developments John Wiley & Son,.


SEYDEL, R. 1994 Practical Bifurcation and Stability Analysis. Springer-Verlag,.

SHEPHERD, F. 1995 Modern coating technology systems – for paper, film and foil. Emap maclaren,.


WEISS, H. L. 1977 Coating & laminating machines. Converting tech company.,


Appendix A

A mesh generation method for surface having rounded corner with small radius of curvature

A.1 Smooth node distribution along rounded corner of die lip

A.1.1 Patching straight line and arc of circle

For simplicity, coating die edges can be approximated as mathematically sharp corner. In this case, contact line, end of free surface, is pinned: in other words, it cannot move along die surface. In order to overcome this unrealistic limitation, Gibbs condition (Gibbs, 1961) are used. See Kistler (1983); Gates (1999) for details. Such inequality condition determine whether contact line is pinned or move along die surface.

In reality, coating die edges are not mathematical corners, they are rounded. The contact line slides along the die surface with maintaining static contact angle. For describing rounded corner in the coating die, one possible candidate is a geometry described by connecting straight lines and arc of circles in series, and we called it a “patch geometry”. Die shoulder and die lip are described by straight line, and die edge, a region between shoulder and lip, are described by arc of circle. Typically, radius of curvature of die edge is $25\,\mu m \sim 50\,\mu m$, which is usually limited by machining
A.1. SMOOTH NODE DISTRIBUTION ALONG ROUNDED CORNER OF DIE LIP

Figure A.1: Example of geometry with connecting straight lines and arc of circle. $\mathbf{x}_s$ and $\mathbf{x}_e$ are starting point and ending point of straight line, $\mathbf{x}_c$ and $R_c$ are the center and the radius of arc of circle, and $\mathbf{x}_p$ is patching point between straight line and arc of circle. Patching points are chosen to satisfy continuity of slope between straight line and arc of circle.

Example of patched geometry is shown in Figure A.1. The geometry can be expressed in terms of mathematical functions. Straight line is described as,

$$f_l(x, y) = -(x - x_s)(y - y_s) + (x_e - x_s)(y - y_s) = 0,$$

(A.1)

where $\mathbf{x}_s = ix_s + jy_s$ and $\mathbf{x}_e = ix_e + jy_e$ are the starting point and the ending point of straight line, respectively. Note that arc length increases from $\mathbf{x}_s$ to $\mathbf{x}_e$. Arc of circle is described as,

$$f_r(x, y) = (x - x_c)^2 + (y - y_c)^2 - R_c^2 = 0,$$

(A.2)

where $\mathbf{x}_c = ix_c + jy_c$ is the center of circle. Note that arc length increases in counterclock direction.

The sign of each term in Eqs. (A.1) and (A.2), and the direction of increasing arc length should be kept consistent over the whole geometry. Because the method proposed in here is based on arc length of the geometry, direction of increasing arc length is important.
A.1. SMOOTH NODE DISTRIBUTION ALONG ROUNDED CORNER OF DIE LIP

A.1.2 Node distribution

In Galerkin finite element method, the solutions, i.e. field variables, are approximated over the discretized domain — grid, mesh or nodal points. The system domain boundary, especially for boundary conforming mesh, should be discretized properly in order to reflect effects from the complex geometry. Good choice of node distribution is essential for the proper discretization. For example, for the coating flow near the die lip with rounded corner, a number of nodal points along arc of circle should be sufficient enough to deliver effect of curvature to the flow system.

In order to yield best node distribution with a given number of nodal points, node concentrations near the region where shape varies rapidly need to be high enough to catch those changes. Typically, one-dimensional hyperbolic tangent stretching functions based on truncation error bounds (Vinokur, 1983a) are used to concentrate nodes near rapid geometry variation.

The stretching function method is designed to adjust “strength” of node concentration to control the distribution. However, the method use the function based on computational coordinate or reference coordinate not physical coordinate. In order to concentrate node based on physical coordinate, one may keep track of a relationship between computational domain coordinate and physical domain coordinate. Furthermore, when boundary has more than two regions requiring high node concentrations, the formulation of stretching function becomes extremely complex.

Another method, proposed in this study, is to control node spacing based on curvature of the shape of geometry. In order to achieve it, a function for node distribution need to be constructed based on physical domain coordinate. The main concept is close to adaptive grid generation that the nodal points move to concentrate in regions of large variations in the solution as they emerge (Thompson et al., 1985). The grid generation devised to minimize solution errors that come from an “unsuitable” node distribution.

In one-dimensional adaptive grid generation, the solution error can be reduced by distributing the nodal points. Some positive weight function $w(s)$, which is related to the solution error, is equally distributed over the field, i.e.,

\[ \int_{s_i}^{s_{i+1}} w(s)\, ds = \text{constant}, \quad (A.3) \]

where $s$ is arc length along the curve of the system, $s_i$ and $s_{i+1}$ are the $i^{th}$ and $i+1^{th}$ nodal points along the curve. In proposed mesh generation scheme, the value of
weighting function $w(s)$ is based on curvature of geometry: the higher curvature, the higher weighting function. Therefore, a high curvature region requires small node spacing so as to keep the integral of Eq. (A.3) constant.

The main advantage of this method over the conventional hyperbolic tangent stretching function is flexibility. For example, the method can handle multiple high node concentration regions without modification.

**Equidistribution**

Equation (A.3) can be expressed in discrete form,

$$\Delta s_i w_i = \text{constant}, \quad (A.4)$$

where $\Delta s_i$ is the node interval, i.e., $\Delta s_i = s_{i+1} - s_i$, and $w_i$ is weighting function for $i^{th}$ node.

As mentioned in Thompson et al. (1985), nonuniform node distribution can be considered to be a transformation, $s(\zeta)$, from uniformly spaced mesh in $\zeta$ space, with the coordinate $\zeta$ serving to identify the nodal points. Nodal points are conveniently defined by successive integer values $\zeta$ from 1 to total number of nodal points $N$. In this reference domain, $\Delta \zeta$ is set to be one and $\Delta s = (ds/d\zeta)\Delta \zeta = (\partial s/\partial \zeta) = s_\zeta$. Here, $s_\zeta$ represents the variation in $s$ between nodal points. Then Eq. (A.4) becomes,

$$s_\zeta w = \text{constant}. \quad (A.5)$$

This is called the equidistribution statement (Thompson et al., 1985). According to Eq. (A.5), high weighting function yields small spacing $s_\zeta$.

The constant value in Eq. (A.5) can be evaluated the method described below:

$$s_\zeta w(s) = \frac{ds}{d\zeta} w(s) = \text{constant} = C$$

$$w(s)ds = C\ d\zeta$$

$$\int_{s_s}^{s_e} w(s)ds = \int_1^N C\ d\zeta$$

$$C = \frac{1}{N-1} \int_{s_s}^{s_e} w(s)ds, \quad (A.6)$$
A.1. SMOOTH NODE DISTRIBUTION ALONG ROUNDED CORNER OF DIE LIP

where $s_i$ and $s_e$ are the starting and the ending arc lengths of a given curve.

The node distribution along the geometry of system is to find $i^{th}$ node arc length $s_i$ using the equidistribution statement. Satisfying this statement can be rewritten as follow. From either Eq. (A.3) or Eq. (A.5), one can obtain a relationship for $s_i$,

$$\int_{s_i}^{s_e} w(s) \, ds = \frac{i - 1}{N - 1} \int_{s_i}^{s_e} w(s) \, ds. \tag{A.7}$$

Therefore $s_i$ can be found by solving Eq. (A.7).

In elliptic mesh generation system, the node distribution along the geometry of system is imposed as a boundary condition of the mesh system. However, the boundary condition does not necessarily require arc length value of the nodal point. As will shown in Sec. A.1.3, node spacing information — the derivative of arc length with respect to computational domain coordinate $s_\zeta$ — can be used instead. $s_\zeta$ can be evaluated using Eq. (A.5),

$$s_\zeta = \frac{ds}{d\zeta} = \frac{1}{w(s)(N - 1)} \int_{s_i}^{s_e} w(s) \, ds. \tag{A.8}$$

If one use dimensionless arc length $s^* = s/(s_e - s_i)$ and computational domain $\xi = \zeta/(N - 1)$, Eq. (A.8) becomes,

$$\frac{ds^*}{d\xi} = \frac{1}{w(s^*)} \int_{0}^{1} w(s^*) \, ds^*. \tag{A.9}$$

Weighting function construction

In this study, the weighting function is related to curvature of the geometry. The simplest choice is

$$w(s) = 1 + \alpha |\kappa(s)|, \tag{A.10}$$

where $\kappa(s)$ is the curvature for a given geometry at arc length $s$, and $\alpha$ is user-defined coefficient. $\alpha$ can adjust the effect of curvature on weighting function.

However, two difficulties arose when we chose Eq. (A.10) for the patch geometry:
A.1. SMOOTH NODE DISTRIBUTION ALONG ROUNDED CORNER OF DIE LIP

- Points where weighting functions are evaluated are not known \textit{a priori}.
- Curvature is not continuous across from straight line to arc or circle and \textit{vice versa}.

In simple patch geometry, one may find analytic formula for Eq. (A.10). But, in general, it is difficult to obtain analytic one. To handle this difficulties, we use \textit{dual grid method}: use a “predetermined” grid for weighting function to generate a Galerkin finite element method (GFEM) grid.

A patch geometry usually requires continuous in slope not curvature. However, discontinuity in curvature can destroy “smoothness” of mesh. In this study, we applied artificial smoothing process to weighting function for ensuring smooth changes in node spacing. Therefore, in the proposed method, node spacings and distributions can be controlled by two factors: coefficient $\alpha$ and a number of smoothing process step (it will be discussed later).

\textbf{Dual grid strategy}

The dual grid method is based on maintaining two grids for a given geometry of the system: “predetermined” grid that contains weighting function values and GFEM grid that needs to be computed. Typically, “predetermined” grid has coarser than GFEM one. When weighting function value at a certain arc length is required for either Eq. (A.7) or Eq. (A.8), the value is interpolated using “predetermined” grid. In this study, we use linear interpolation between adjacent two points. A example of dual grid is shown at Fig. A.2.

For slot coating system, radius of curvature is about $25 \mu \text{m} \sim 50 \mu \text{m}$ and die lip length is about $1000 \mu \text{m}$. For die lip or die shoulder which is straight line, small number of node is enough to describe the geometry. For die edge which is arc of circle, large number of node required to capture the curvature. In this study, to minimize storage for “predetermined” grid, straight line and arc of circle sections use 5 nodal points and 30 or less points, respectively.

\textbf{Smoothing process for weighting function}

An artificial smoothing process is applied to weighting function on the “predetermined” grid to get rid of abrupt change in the function. In this study, two types of numerical smoothing process are proposed: averaging and artificial diffusion methods. In general, numerical smoothing process is applied as an iterative manner. Successive
steps of the process smoothed out rapid changes in weighting function. Here, \( w^{(i)}(s^p_k) \) stands for \( i^{th} \) smoothing step weighting function at \( k^{th} \) “predetermined” grid point with arc length \( s^p_k \).

The averaging method can be expressed as:

\[
   w^{(i+1)}(s^p_k) = \frac{w^{(i)}(s^p_{k-1}) + w^{(i)}(s^p_{k+1})}{2}.
\]  

(A.11)

It will create a “ramp” when initial weighting function profile has a step like Fig. A.3. The method yields smoothly change weighting function value but it is combination of straight lines and their slope changes abruptly. Because arc length for GFEM grid is based on integral of weighting function as in Eq. (A.7) or Eq. (A.8), the resulting arc length profile is piecewise quadratic functions. In this study, 50 steps are used for smoothing weighting function.

Another method is to obtain smoothed weighting function profile by solving the time evolution of a diffusion-like processing using \( w(s) \) as the initial condition. It treats weighting function as a concentration and control degree of smoothness by adjusting

"Predetermined" grid for weighting function

Figure A.2: Dual grid strategy for evaluating weighting function. Black circles are node points for “predetermined” grid, and white circle are node points for GFEM grid. Note that values of weighting function to get GFEM grid points are interpolated using “predetermined” grid.
A.1. SMOOTH NODE DISTRIBUTION ALONG ROUNDED CORNER OF DIE LIP

“artificial” time $\hat{t}$:

$$\frac{\partial w}{\partial \hat{t}} = \frac{\partial^2 w}{\partial s^2}. \quad (A.12)$$

Note diffusion coefficient is chosen to be one in this study.

One of the simplest ways to perform the smoothing process uses explicit Euler time integration method with finite difference formula. In uniform spacing “predetermined” grid case, the equation becomes,

$$\frac{w^{(i+1)}(s^p_k) - w^{(i)}(s^p_k)}{\Delta \hat{t}} = \frac{w^{(i)}(s^p_{k-1}) - 2w^{(i)}(s^p_k) + w^{(i)}(s^p_{k+1})}{(\Delta s^p)^2}$$

$$w^{(i+1)}(s^p_k) = w^{(i)}(s^p_k) + \left( \frac{\Delta \hat{t}}{(\Delta s^p)^2} \right) \left( w^{(i)}(s^p_{k-1}) - 2w^{(i)}(s^p_k) + w^{(i)}(s^p_{k+1}) \right), \quad (A.13)$$

where $\Delta \hat{t}$ is arbitrary time step. In non-uniform spacing case, the equation becomes,

$$w^{(i+1)}(s^p_k) = w^{(i)}(s^p_k) + \frac{\Delta \hat{t}}{(\Delta s^p_k + \Delta s^p_{k+1})/2 \left( \frac{w^{(i)}(s^p_{k+1}) - w^{(i)}(s^p_k)}{\Delta s^p_k} + \frac{w^{(i)}(s^p_k) - w^{(i)}(s^p_{k-1})}{\Delta s^p_{k+1}} \right)}, (A.14)$$

![Figure A.3: Numerical smoothing algorithm for weighting function: averaging and "artificial" diffusion system.](image)
where \( \Delta s^p_k = s^p_k - s^p_{k-1} \).

In order to prevent numerical instability in explicit Euler step, time step \( \Delta \tilde{t} \) is chosen as \( 0.5(\Delta s^p_{\text{min}})^2 \), where \( \Delta s^p_{\text{min}} \) is the minimum node spacing in "predetermined" grid. The number of step used in this study is 200.

Diffusion-like system yields error function type weighting function. Computed arc length follows integral of exponential function which is close to hyperbolic tangent function. Hence the resulting node distribution is close to the conventional stretching function. Examples of smoothing process are shown in Fig. A.3.

Calculating arc length based on weighting function

Arc length at \( i^{th} \) node \( s_i \) is computed by solving Eq. (A.7). The computation requires evaluation of integral of weighting function. However, the exact value of integral is not

\[
\int_{s_k}^{s_{i+1}} w(s) \, ds,
\]

where \( s_k \) and \( s_{i+1} \) are the starting point, the ending point, and \( i^{th} \) nodal point on GFEM grid respectively. \( s^p_k \) is the closest "predetermined" grid point to GFEM grid point \( s_i \), but the value has lower than \( s_i \). The value of weighting function at \( s_i \) is evaluated using linear interpolation of "predetermined" grid points. Trapezoid rule is used to evaluate integral.

**Figure A.4:** Scheme for evaluating integral of weighting function from the starting point to the \( i^{th} \) nodal point on GFEM grid. \( s_s, s_e, \) and \( s_i \) are the starting point, the ending point, and \( i^{th} \) nodal point on GFEM grid respectively. \( s^p_k \) is the closest "predetermined" grid point to GFEM grid point \( s_i \), but the value has lower than \( s_i \). The value of weighting function at \( s_i \) is evaluated using linear interpolation of "predetermined" grid points. Trapezoid rule is used to evaluate integral.
A.1. SMOOTH NODE DISTRIBUTION ALONG ROUNDED CORNER OF DIE LIP

demanding, because overall node distribution is important, not precise node locations. Here, we use trapezoid rule to evaluate integral. Integral of right hand side (RHS) in Eq. (A.7) is straight forward to evaluate using “predetermined” grid.

Left hand side (LHS) of Eq. (A.7) is difficult to evaluate because the upper limit of integral $s_i$ is unknown for the equation. We tried two ways to compute $s_i$ from Eq. (A.7). First method is approximate $w(s_i)$ as $w(s_{i-1})$ where $s_{i-1}$ is the arclength of the closest “predetermined” grid to G/FEM grid corresponding to $s_i$. Then Eq. (A.7) become explicit in terms of $s_i$. This is simple to evaluate $s_i$, but it requires large nodal points for “predetermined” grid in order to improve accuracy of the integral.

Second method is to find $s_i$ by solving equation using Newton’s method. Residual $R_d$ is

$$R_d = \int_{s_{i-1}}^{s_i} w(s) \, ds - \int_{s_i}^{s_e} w(s) \, ds, \quad (A.15)$$

Scheme of evaluating integral in first term of RHS in Equation (A.15) is shown in Figure A.4. Jacobian $J_d$ can be approximated by finite difference quotient, $(R_d(s_i + \epsilon) - R_d(s_i))/\epsilon$, where $\epsilon = (s_e - s_{i-1})/(N - 1) \times 10^{-6}$ is used in this study. The initial value for finding $s_i$ is $s_{i-1} + 0.1(s_e - s_{i-1})/(N - 1)$, and typically less than 10 iteration is required to get $s_i$ with $\|R_d\|_2 < 10^{-12}$.

Geometry tracking

Sometime, arc length of G/FEM grid point need to be converted to coordinate values. For example, initial guess mesh for the elliptic mesh generation system requires coordinate values of nodes on boundaries. Here, we propose the methods of converting arc length to coordinate values when the functional form of a boundary is given as $f(x, y) = 0$. The method is similar to pseudo-arc-length continuation (Bolstad and Keller, 1986).

In this study, a boundary can be described by combination of straight line regions and arc of circle regions (patch geometry) that can be expressed in combination of functions as discussed in Sec. A.1.1. However, each region of the boundary should have the same direction of increasing arc length. For example, one can choose counterclockwise direction for increasing arc length.

As in pseudo-arc-length continuation, predictor-corrector step are applied in geometry
A.1. SMOOTH NODE DISTRIBUTION ALONG ROUNDED CORNER OF DIE LIP

tracking. Predictor step used in this study is tangent predictor step.

\[ \mathbf{x}_0(s_{i+1}) = \mathbf{x}(s_i) + \mathbf{t}_f(s_i) \Delta s_{i+1} \]  
(A.16)

where \( \Delta s_{i+1} = s_{i+1} - s_i \), \( \mathbf{t}_f(s_i) \) is the tangent vector of the geometry, and \( \mathbf{x}_0(s_{i+1}) \) is the predicted \( i+1 \)th coordinate which is initial guess for corrector step. Tangent of curve at \( \mathbf{x}(s_i) \) is

\[
\mathbf{t}_f(s_i) = \left. \frac{d\mathbf{x}}{ds} \right|_{s_i} \mathbf{i} + \left. \frac{d\mathbf{y}}{ds} \right|_{s_i} \mathbf{j} \\
= \left( \frac{\text{sign}_x |df/dy|}{\sqrt{(df/dx)^2 + (df/dy)^2}} \right) \left. \mathbf{i} + \left( \frac{\text{sign}_y |df/dx|}{\sqrt{(df/dx)^2 + (df/dy)^2}} \right) \right|_{s_i} \mathbf{j} \quad \text{(A.17)}
\]

where \( \text{sign}_x \) and \( \text{sign}_y \) are chosen such that \( \mathbf{t}_f \) point toward increasing arc length direction. In this study, \( \text{sign}_x \) is the sign of \((df/dy)\), and \( \text{sign}_y \) is the sign of \((-df/dx)\) to follow counter-clock direction. Note that \( \mathbf{i}(df/dy) + \mathbf{j}(-df/dx) \) is a vector tangent to a trajectory of the function \( f(x,y) = 0 \).

For two dimensional case, two residuals are required. The \( j \)th corrector step’s residual is:

\[
R_x = f(x^{(j)}(s_j),y) \quad \text{(A.18)}
\]

\[
R_y = \left. \frac{dx}{ds} \right|_{x(s_i)} (x^{(j)}(s_{i+1}) - x(s_i)) + \left. \frac{dy}{ds} \right|_{x(s_i)} (y^{(j)}(s_{i+1}) - y(s_i)) - \Delta s_{i+1} \quad \text{(A.19)}
\]

where \( x^{(j)}(s_{i+1}) = i x^{(j)}(s_{i+1}) + j y^{(j)}(s_{i+1}) \) which is a position vector for \( i+1 \)th node at \( j \)th step. Equation (A.18) restricts nodal point to a given geometry and Eq. (A.19) is linearized version of the pseudo-arc-length condition that keeps the distance between \( s_i \) and \( s_{i+1} \) to specified \( \Delta s_{i+1} \). The \((i+1)\)th nodal position \( \mathbf{x}(s_{i+1}) \) can be obtained by solving the following system at each iteration:

\[
\begin{bmatrix}
\frac{df}{dx} \\
\frac{df}{ds}
\end{bmatrix}
\begin{bmatrix}
x^{(j)}(s_{i+1}) \\
x^{(j)}(s_i)
\end{bmatrix}
\begin{bmatrix}
\Delta x^{(j)} \\
\Delta y^{(j)}
\end{bmatrix}
= -
\begin{bmatrix}
R_x \\
R_y
\end{bmatrix}
\]

\[
x^{(j+1)}(s_{i+1}) = x^{(j)}(s_{i+1}) + \Delta x^{(j)}. \quad \text{(A.20)}
\]

The iteration continues until \( L_2 \) norm of residual is less than \( 10^{-12} \). Typically, less than 10 iterations are required to get \( i+1 \)th node coordinate. A schematic drawing of the geometry tracking procedure is shown in Fig. A.5.
A.1.3 Coupling with elliptic mesh generation system

Residual for diffusional transport elliptic mesh generation system

Constructed arc length distribution, based on either Eq. (A.7) or (A.8), are coupled to elliptic mesh generation system as boundary conditions. In two dimension, when nodes on a boundary are allowed to shift their positions along a specified geometry, first mesh boundary condition describes the geometry and second one distributes node:

\[
R_1 = f(x, y) 
\]

\[
R_2 = \int_{\partial \Omega_0} \phi^j \left( \frac{d}{d\xi} \left( \frac{1}{D_{ij}} \frac{ds}{d\xi} \right) \right) d\xi 
\]

where \(D_{ij}\) is mesh diffusivity for mesh coordinate potential \(\xi\) in diffusional transport elliptic mesh generation system. See de Santos (1991); Benjamin (1994) for details. \(\partial \Omega_0\) is the computational domain boundary for a specified geometry.

Equation (A.21) uses the functional form of the geometry, Eq (A.1) or (A.2) in the patch geometry. Whereas Eq. (A.22) comes from the equidistribution statement:

\[
\omega \frac{ds}{d\xi} = C = \text{constant} \\
\frac{d}{d\xi} \left( \omega \frac{ds}{d\xi} \right) = 0 
\]

Figure A.5: Scheme for converting arc length of curve to position — geometry tracking. \(x(s_i)\) is the nodal position vector on curve, \(x_0(s_{i+1})\) is the position vector by tangent predictor step, and \(t_f\) is tangent vector to geometry curve at \(x(s_i)\).
A.1. SMOOTH NODE DISTRIBUTION ALONG ROUNDED CORNER OF DIE LIP

where $\omega$ is the weighting function. Note that it is different from $w$ for the patched geometry. In diffusional transport elliptic mesh system, $\omega = (\partial F/\partial \xi)^{-1} = 1/D_\xi$, where $F$ is a stretching function, or dimensionless arc length $s^*$, for boundary along $\xi$ (de Santos, 1991). Therefore the node distribution residual, Eq. (A.22), is a weighted residual of the equidistribution statement, Eq. (A.23). Using integral by part, the residual become,

$$R_2 = \int_{\partial \Omega_0} \phi^j \frac{d}{d\xi} \left( \frac{1}{D_\xi} \frac{ds}{d\xi} \right) d\xi$$

Note that the end-point term in Eq. (A.24) is not evaluated. The extreme points of the boundary, which corresponding to $s_f$ and $s_e$, are specified either by description of geometry or by angle between geometry and subdomain boundary of mesh.

However, there are several difficulties, especially evaluating residual Eq. (A.21), during solving the mesh generation system using Newton’s method. In each step or iteration of the Newton’s method, boundary nodal positions may not be necessarily on the boundary a described by a function until it satisfies Eq. (A.21). When the patch geometry are used, two types of difficulties arise in evaluating mesh residuals:

- It is not straightforward to determine which region of the geometry use what kind of functions, either Eq. (A.1) or (A.2), to compute Eq. (A.21).
- It is ambiguous to evaluate the derivative of stretching function for Eq. (A.22) in a given position.

In order to overcome these difficulties, we evaluated the residual equations based on projections of nodal points onto the geometry described by the functions during each step of Newton’s method.

**Evaluating arc length with given position**

In this study, the projection onto a geometry stands for finding the point on the geometry that ensure the *shortest* distance from the given point, i.e. normal projection point. For find the shortest distance for the patch geometry, distances from a
A.1. SMOOTH NODE DISTRIBUTION ALONG ROUNDED CORNER OF DIE LIP

given point to each region of the geometry are computed and compared. Computed
distances are accepted only when a normal projection point are on the boundary of
the geometry.

In Fig. A.6, for example, $x_1$ and $x_2$ are located outside the geometry. $x_1^p$ and $x_2^p$ are the
shortest normal projection points on the curve. The “pseudo” arc length along the
curve are evaluated based on those points, $s_1$ and $s_2$. The residual equation (A.21), is
evaluated based on the projected points. For $x_1$, the function for describing “Straight
line 1” and $x_1^p$ are used to evaluate the residual equation. The residual equation for
distributing nodes, Eq. (A.22), uses arc length $s_1$ to evaluate $D_ξ$.

Overall scheme

The method proposed here is one way to provide boundary conditions for the diffu-
sional transport elliptic mesh generation system. The main additional feature is to

---

**Figure A.6:** Example of estimating arc length when point under “surveillance”
are off from the geometry curve. $x_1$ and $x_2$ are GFEM mesh points inside Newton
loop. They can be off from the geometry curve before converging to solution
of the mesh system. $s_1$ and $s_2$ are “pseudo” arc length on curve for $x_1$ and $x_2$
which are measured from the starting point of the curve to projected points on
the curve, $x_1^p$ and $x_2^p$. 
A.1. SMOOTH NODE DISTRIBUTION ALONG ROUNDED CORNER OF DIE LIP

1.3 Coupling with elliptic mesh generation system inside Newton loop

Basic Newton scheme is described in Figure 7. However, when geometry continuation is applied to the flow/mesh system, especially, pseudo-arc-length continuation; geometry parameters, for example, patching point, length of straight, etc., are also the part of unknown in the system. When this is the case, the geometry initialization procedure should be executed inside Newton loop of corrector step, to make sure changes in the geometry are properly reflected.

Figure A.7: Scheme for coupling patched geometry and G/FEM system inside Newton step.
A.2  Example: downstream slot coating — Low flow limit

In order to test new mesh scheme for patch geometry, we choose downstream slot coating flow as depicted in Fig. A.8. Edge in coating dies have a specified radius of curvature. We choose 25\(\mu\)m as a base case in this study that is extremely sharp convex corner. de Almeida (1995); Musson (2001) claimed that elliptic mesh generation system cannot concentrate nodes near convex sharp corner. Romero et al. (2006) solved the same coating flow but the die lip has large radius of curvature 100\(\mu\)m that is relatively easy to handle by the mesh generation system.

Figure A.8 shows the downstream slot die coating flow. In relatively high flow rate, the downstream meniscus wet the die shoulder, i.e. the static contact line located at the die shoulder as shown in Fig. A.8. The contact line moves toward die lip, as the flow rate through inflow boundary decrease. If it decrease further, the contact line invade into the die lip pass through the rounded corner.

The coating flow can be described by the two dimensional, steady-state Navier-Stokes Equation with Newtonian liquid. In this study, we ignore the effect of gravity. Because the size of coating bead is small, solution is aqueous, and web speed is fast; gravitational force is minuscule compared with viscous force and capillary force. The base case operating parameters and dimensionless numbers are summarized in Table A.1. The total domain length from synthetic inlet boundary to outlet boundary is set to 16\(H_0\).

Boundary conditions used in the systems are:

- Inflow velocity profile: At inflow boundary, flow assumed to be rectilinear,
Table A.1: Operating parameter and dimensionless number downstream slot coating flow

<table>
<thead>
<tr>
<th>Operating parameters</th>
<th>Unit</th>
<th>Base case</th>
</tr>
</thead>
<tbody>
<tr>
<td>Film thickness ($h_\infty$)</td>
<td>mm</td>
<td>0.125</td>
</tr>
<tr>
<td>Web speed ($V_w$)</td>
<td>m/sec</td>
<td>1</td>
</tr>
<tr>
<td>Ambient pressure ($P_{amb}$)</td>
<td>Pa</td>
<td>0</td>
</tr>
<tr>
<td>Surface tension ($\sigma$)</td>
<td>dyne/cm</td>
<td>61</td>
</tr>
<tr>
<td>Viscosity ($\mu$)</td>
<td>mPa·s</td>
<td>2.3</td>
</tr>
<tr>
<td>Density ($\rho$)</td>
<td>g/cm³</td>
<td>1.2</td>
</tr>
<tr>
<td>Static contact angle ($\theta_s$)</td>
<td>deg.</td>
<td>60</td>
</tr>
<tr>
<td>Gap height ($H_0$)</td>
<td>$\mu$m</td>
<td>250</td>
</tr>
<tr>
<td>Radius of curvature in die edge ($R_e$)</td>
<td>$\mu$m</td>
<td>25</td>
</tr>
<tr>
<td>Die lip length ($L_d$)</td>
<td>$\mu$m</td>
<td>1250</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Dimensionless variables used in the system</th>
<th>Definition</th>
<th>Base value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dimensionless flow rate ($q^*$)</td>
<td>$\frac{h_\infty}{H_0}$</td>
<td>0.5</td>
</tr>
<tr>
<td>Reynolds number ($Re$)</td>
<td>$\frac{\rho V_w H_0}{\mu}$</td>
<td>13.0</td>
</tr>
<tr>
<td>Capillary number ($Ca$)</td>
<td>$\frac{\mu V_w}{\sigma}$</td>
<td>0.377</td>
</tr>
</tbody>
</table>
A.2. EXAMPLE: DOWNSTREAM SLOT COATING — LOW FLOW LIMIT

Couette-Poisuille velocity profile,

\[
    u(y) = \frac{6V_w h_\infty}{H_0} \left[ \left( -1 + \frac{1}{2} \frac{H_0}{h_\infty} \right) \left( \frac{y}{H_0} \right)^2 + \left( -1 + \frac{2}{3} \frac{H_0}{h_\infty} \right) \left( \frac{y}{H_0} \right) + \frac{1}{6} \frac{H_0}{h_\infty} \right],
\]

\[
v(y) = 0,
\]

(A.25)

where \( u \) and \( v \) are velocities in \( x \) and \( y \) direction, respectively. \( V_w \) is the web speed, and \( h_\infty \) is the wet coating thickness. See Fig. A.8.

- Moving web: Along moving web no-slip / no-penetration condition are imposed:

\[
u = V_w, \quad v = 0.
\]

(A.26)

- Outflow: At synthetic outflow boundary, flow assumed to be fully developed in flow direction:

\[
n \cdot \nabla u = 0,
\]

(A.27)

where \( n \) is the normal vector to the outflow boundary.

- Die surface: Along the die surface, no-slip / no-penetration condition are applied.

\[
u = 0.
\]

(A.28)

Figure A.8: Schematic representation of downstream of the slot coating flow. \( H_0 \) is the gap height, \( h_\infty \) is the final wet film thickness, and \( L_d \) is the die lip length. I, II, and III are the subdomain for structured mesh. \( \theta_A \) and \( \theta_B \) are the angle between artificial subdomain boundary and die lip or free surface. In this study, we choose 110° and 120°, respectively. For fully developed outflow, \( \theta_\infty \) is chosen as 90° for mesh corner boundary condition.
A.2. EXAMPLE: DOWNSTREAM SLOT COATING — LOW FLOW LIMIT

- Free surface: Along the downstream meniscus, force balance and no-penetration condition are imposed.

\[ n_f \cdot T = \sigma \frac{dt_f}{ds} - n_f P_{amb}, \quad (A.29) \]

\[ n_w \cdot u = 0, \quad (A.30) \]

where \( t_f \) and \( n_f \) are the local unit tangent and unit normal to the free surface, \( s \) is the arc length coordinate along the free surface, \( T \) is the stress tensor, \( \sigma \) is the liquid surface tension, and \( P_{amb} \) is the ambient pressure.

- Static contact line: The contact line is free to move with the contact angle \( \theta_s \) between the die surface and the free surface,

\[ n_{ds} \cdot n_f = \cos \theta_s, \quad (A.31) \]

where \( n_{ds} \) is the unit normal to die surface, and \( n_f \) is unit normal to the free surface.

Here, diffusional transport elliptic mesh generation system is deployed to discretize the flow domain. In sum, the mass/momentum system and the mesh system are fully coupled to track the position of the interface. See Kistler and Scriven (1984) for details about treating free surface problem in coating flows. The coupled systems use mapping the physical domain \( \Omega \) to the computational domain \( \Omega_0 \) by means of a coordinate transformation \( \mathbf{x} = \mathbf{x}(\xi) \), where \( \mathbf{x} \) and \( \xi \) denote position coordinate in physical and computational domain, respectively. Inverse transformation, \( \xi = \xi(\mathbf{x}) \) is governed by diffusional transport elliptic mesh generation system with variable mesh diffusivity \( D_\xi \) and \( D_\eta \):

\[ \nabla \cdot (D_\xi \nabla \xi) = 0, \quad \nabla \cdot (D_\eta \nabla \eta) = 0. \quad (A.32) \]

Boundary conditions for mesh system follows geometry description, Eq. (A.21), and node distribution, Eq. (A.22). In geometry description, inflow, outflow and moving web are described by straight line, Eq. (A.1). Die surface is described by the patched geometry: straight lines, Eq. (A.1), are for die lip and die shoulder; and arc of circle, Eq. (A.2), is for die corner. Along the free surface, no penetration condition, Eq. (A.30), is used instead.

Node distributions along inflow, outflow and moving web are chosen to be uniform spacing, i.e. \( D_\xi \) or \( D_\eta \) is chosen to be one. Along free surface, node are concentrated toward the static contact point using hyperbolic stretching function (Vinokur, 1983a). Node distribution along die surface is computed using the method proposed in this study based on the curvature of the die surface. See Sec. A.1.2 for details.
A.2. EXAMPLE: DOWNSTREAM SLOT COATING — LOW FLOW LIMIT

The set of differential equations for mass/momentum and mesh system are solved on computational domain by means of Galerkin/Finite Element Method (G/FEM). The position and velocity fields are spanned by bi-quadratic basis function, and the pressure pressure field is approximated by linear discontinuous basis function. The set of nonlinear algebraic equations from the method of weighted residuals is solved by Newton’s method. The trajectory in solution space, for example tracking the changes in solution state by changing flow rate, are tracked by pseudo-arc-length continuation (Bolstad and Keller [1986]).

A.2.1 Results for downstream slot coating with rounded corner

The effect of the static contact angle is shown in Figure A.9 when dimensionless flow rate $q^*$ is the same as 0.5. Low contact angle (Fig. A.9(b)) wet die shoulder more than high contact angle (Fig. A.9(a)). Both figures show high node concentration near the rounded die corner even with small radius of curvature of the corner, 50 $\mu$m.

Figures A.10 and A.11 shows mesh and streamline plot snap shots of the downstream slot coating flow during pseudo-arc-length continuation, which are used to find low flow limit in slot coating in Figure A.13 The results is based on static contact angle 60°. When the flow rate or wet thickness is large, liquid wets die shoulder and the separation bubble that contains vortex is observed. This type of corner vortex between solid surface and free surface, was studied in details by Moffatt [1963]. Near

![Figure A.9: Downstream single-layer slot coating mesh with different static contact angles.](image)

(a) 100° static contact angle with $h_\infty = 1/2H_g$.

(b) 60° static contact angle with $h_\infty = 1/2H_g$. 

Figure A.9: Downstream single-layer slot coating mesh with different static contact angles.
$H_0/h_\infty = 2.922$, the static contact line approaches the rounded corner (Figs. A.10(b) and A.11(b)) and, at the same time, the corner vortex shrink as well. The flow recirculation near the die lip appear near $H_0/h_\infty = 3$ and grow its size as the wet thickness $h_\infty$ decreases. The static contact line is located at the middle of the die lip corner at $H_0/h_\infty = 3.683$ (Figs. A.10(c) and A.11(c)). The contact line hit the die lip surface at $H_0/h_\infty = 4.796$ (Figs. A.10(d) and A.11(d)) and the flow state is very close to the low flow limit: minimum film thickness that can be achieved by the coating flow. After this flow state, the contact line invade into the die lip (Figs. A.10(e) and A.11(e)).

Figure A.12 depicts how to measure the arc length of the static contact line and arc length range of rounded corner of the die edge. Using the arc length, the location of static contact line can be tracked with respect to wet thickness using pseudo-arc-length continuation (Fig. A.13). The contact line stays within the rounded corner from $H_0/h_\infty = 3.1$ to 4.8. At low flow limit, the gap over film thickness has the maximum value, $H_0/h_\infty = 4.8$. After the maximum value, $H_0/h_\infty$ decreases as the arc length of the static contact line $s_{sc}$ decreases. However, the sensitivity of the arc length with respect to the flow rate or wet thickness is extremely large. Therefore the meniscus will invade into the coating bead, as shown in the progression from Fig. A.11(d) to A.11(e), that may lead to break up of the bead from downstream meniscus to create dry lanes.
2.1 Results for downstream slot coating with rounded corner

A.2. EXAMPLE: DOWNSTREAM SLOT COATING — LOW FLOW LIMIT

Figure A.10: Zoomed mesh plots as static contact line moves with static contact angle 60°. Note that Figure A.10(e) is the snapshot which already passed the low flow limit in Figure A.13.
Figure A.11: Zoomed Streamline plots as static contact line moves with static contact angle 60°. Note that Figure A.11(e) is the snap shot which already passed the low flow limit in Figure A.13.
Figure A.12: Schematic representation of how to measure arc length of static contact line. \( x_s \) is the starting point of measuring arc length along die surface. In Cartesian coordinate, it is (0.3,0). \( s_{s,R} \) and \( s_{e,R} \) are the starting and ending arc length of the rounded corner. \( s_{sc} \) is the arc length of the static contact line.
Figure A.13: Static contact line arclength position $s_{sc}$ at different flow rates of the radius of curvature ($R = 25\mu m$) of the die edge in parallel die lip configuration with $H_0 = 250\mu m$. Turning point, which indicate low flow limit, occurs at the downstream end of rounded corner, and gap to film thickness ratio $H_0/h_\infty$ is about 4.79. The streamline contour plot at the turning point is Figure A.11(d) and mesh plot is Figure A.10(d).
Appendix B

Inter-regional mesh boundary condition for structured mesh

B.1 Mesh boundary conditions for region interfaces

In order to tessellate a complicate physical domain with quadrilateral subdomains with structured mesh, the physical domain may need to be break into smaller subdomains or regions like Fig. B.1. Here, we confined our focus to an elliptic mesh generation system where nodal points were determined by cross points between two coordinate potentials. See Christodoulou (1990); de Santos (1991); Benjamin (1994) for details. In this multi-region meshes, a region boundary is either a physical boundary, when it corresponds to part of the boundary of the physical domain, or an inter-regional boundary when it is internal or borders with another region (Benjamin, 1994). Mesh generation equations in each region separately and coupling the regions through inter-regional mesh boundary conditions. This artificial boundary can be fixed in shape or the distribution of coordinate potential along the boundary can be controlled by imposing spacing information with an essential boundary condition, and the boundary may be move freely inside the physical domain with a Neumann-type boundary condition.

One major source of errors in approximate solution of GFEM system comes from abrupt changes of element size inside the domain or losing “smoothness” of mesh. When a diffusional transport type elliptic mesh generation scheme is considered (de San-
B.1. MESH BOUNDARY CONDITIONS FOR REGION INTERFACES

The size of the elements depends only on the flux of “ether” or “aether”\(^1\), which is the product of gradient of coordinate potential, \(\xi\) and \(\eta\) in two dimensional, and coordinate diffusivity, \(D_{\xi}\) and \(D_{\eta}\) in two dimensional. Therefore the flux matching across inter-regional boundary can be used to get smooth element size change and boundary shape.

A simple geometry flow system is used to test the validity of inter-regional boundary conditions proposed here. The system requires a rather complicated subdomain division and it is called a pentagon lid-driven cavity.

B.1.1 Example of pentagon lid-driven cavities

Pentagon lid-driven cavity flow system has a pentagon-shaped cavity with the moving lid on a top. Basically, this system is the similar to the “conventional” lid-driven cavity flow system except shape of the cavity. Because of the shape, which has five sides, a single subdomain cannot describe the whole physical domain without having large mesh deformations. One possible multi-region mesh is shown in Fig. B.1: five quadrilateral subdomains are used to fit the pentagon-shaped cavity.

The subdomain division shown in Fig. B.1, however, yields the special subdomain corner: an internal five region-confluence. Thompson et al. (1985) show the treatments of multiple region vertices in finite difference and finite volume method. de Santos (1991); Benjamin (1994) considered an internal three-region confluence, an external three-region confluence on a physical boundary, and an internal four-region confluence in a finite element context.

Likewise in other multi-region confluence points, this special corner point also requires two conditions to determine coordinate values for the point in two-dimensional case. When one tried to fix the special corner in physical domain, it is straight forward: just specifying \(x\) and \(y\) in essential way. When the point is located on interlayer or free surface, one condition is based on physics for confining the point on the physical boundary and the other condition is artificial one, for example, specifying angle between two adjacent boundaries at the five region confluence point. In the pentagon lid-driven cavity flow case, however, the point can be chosen arbitrary. Here, we did not fix the point. Instead, we tried to minimize the mesh distortion by letting the mesh generation system locate the point naturally.

\(^1\)Prof. L. E. “Skip” Scriven use this word, because it is not physical quantity. It is an imaginary or artificial material used for determining mesh nodal points.
### B.1. Mesh Boundary Conditions for Region Interfaces

**1.1 Example of Pentagon Lid-Driven Cavities**

*Figure B.1:* Pentagon lid-driven cavity example. One possible subdomain division of physical and Computational domain are shown above. Roman numerals, numbers inside circle and alphabet inside square stand for subdomain numbers, subdomain boundary numbers and subdomain connection type, respectively. “Regular” subdomain connections occur when types of coordinate potentials are the same across subdomain boundaries. In this example, it only occurs when the difference between adjacent subdomain boundary numbers are two under the numbering scheme shown in here. For example, connection B is “regular” type: the connection between side 4 and side 2. However, connection A is “irregular” type connection. Across the inter-regional boundary from I to III, $\xi$ becomes $-\eta$, and $\eta$ becomes $\xi$. See text for details.
B.1. MESH BOUNDARY CONDITIONS FOR REGION INTERFACES

Other challenging aspect of the system comes from connection between subdomains. In general, two types of subdomain connection: “regular” and “irregular” subdomain connection. “Regular” subdomain connections occur when types of coordinate potentials are the same across subdomain boundaries. In Fig. [B.1] all subdomain connections except connection A is “regular” type, when one use the counter clockwise subdomain boundary numbering scheme, like in Fig. [B.1] “regular” connections occurs when the difference between adjacent subdomain boundary numbers are two. However, connection A is “irregular” type connection. Across the inter-regional boundary from I to III, \( \xi \) becomes \(-\eta\), and \( \eta \) becomes \( \xi \). This special connection requires special care on boundary conditions.

B.1.2 Boundary condition for inter-regional boundary

Inter-regional boundary is a fictitious boundary that has no physical law to specify locations. Possibly, the best way to determine its location is minimizing errors of the hosted solution. As mentioned before, significant errors are comes from an abrupt changes in element size. In two-dimensional elliptic mesh generation system, boundaries of the element are defined by contours of two coordinate potentials, \( \xi \) and \( \eta \). Therefore the spacing between each coordinate potential govern element size. In diffusional elliptic mesh generation system, proposed by de Santos (1991); Benjamin (1994), the spacing between coordinate potential are controlled by flux of “ether”:

\[
q_\xi = D_\xi \nabla \xi, \quad q_\eta = D_\eta \nabla \eta.
\]  

(B.1)

At the same ether flux, the smaller diffusivity yields the larger spacing between coordinate potentials.

In principle the diffusional elliptic mesh generation system minimizing the ether flux difference by imposing,

\[
\nabla \cdot (D_\xi \nabla \xi) = 0, \quad \nabla \cdot (D_\eta \nabla \eta) = 0.
\]  

(B.2)

Therefore, when diffusional coefficient is remained constant across a subdomain boundary, the spacing of element across the boundary solely depend on the gradient of coordinate potential, \( \nabla \xi \) or \( \nabla \eta \), depending on the direction of the boundary.
B.1. MESH BOUNDARY CONDITIONS FOR REGION INTERFACES

The mesh equation residual for \( k \)\(^{th} \) nodes are

\[
R_x^k = -\int_\Omega D_\xi \nabla \zeta \cdot \nabla \phi_k \, d\Omega + \int_\Gamma D_\xi (n \cdot \nabla \zeta) \phi_k \, d\Gamma \\
= -\int_\Omega D_\xi \left( \frac{\partial y}{\partial \eta} \frac{\partial \phi_k}{\partial x} - \frac{\partial x}{\partial \eta} \frac{\partial \phi_k}{\partial y} \right) \, d\Omega_0 \\
+ \int_\Gamma_0 D_\xi \frac{1}{|J|} \left( \frac{\partial y}{\partial \eta} n_x - \frac{\partial x}{\partial \eta} n_y \right) \phi_k \left( \frac{d\Gamma}{d\Gamma_0} \right) \, d\Gamma_0,
\]

\[
R_y^k = -\int_\Omega D_\eta \nabla \eta \cdot \nabla \phi_k \, d\Omega + \int_\Gamma D_\eta (n \cdot \nabla \eta) \phi_k \, d\Gamma \\
= -\int_\Omega D_\eta \left( \frac{\partial y}{\partial \xi} \frac{\partial \phi_k}{\partial x} - \frac{\partial x}{\partial \xi} \frac{\partial \phi_k}{\partial y} \right) \, d\Omega_0 \\
+ \int_\Gamma_0 D_\eta \frac{1}{|J|} \left( -\frac{\partial y}{\partial \xi} n_x + \frac{\partial x}{\partial \xi} n_y \right) \phi_k \left( \frac{d\Gamma}{d\Gamma_0} \right) \, d\Gamma_0.
\]  

(B.3)

where \( \Omega \) is physical domain, \( \Omega_0 \) is computational domain, \( \Omega \) is physical domain boundary, \( \Gamma_0 \) is computational subdomain boundary, \( \phi_k \) is the basis function associated to \( k \)\(^{th} \) node, \( n = i n_x + j n_y \) is the outward normal vector with respect to subdomain, and \( |J| \) is the determinant of Jacobian of transformation between physical domain and computational domain,

\[
|J| \equiv \frac{\partial x}{\partial \xi} \frac{\partial y}{\partial \eta} - \frac{\partial y}{\partial \xi} \frac{\partial x}{\partial \eta}.
\]  

(B.4)

Note that \( d\Omega_0 \) is \( d\xi \, d\eta \) and \( d\Gamma_0 \) is \( d\xi \) or \( d\eta \) which depends on alignment of subdomain boundary. In boundary integral term on Eq. (B.3), basis function \( \phi_k \) are belong to subdomain boundary node. For more detail, see Carvalho (1996).

“Regular” subdomain connection, like the connection between subdomain I and V of pentagon lid-driven cavity (Fig. B.2(a)), is considered, the mesh equation residuals of \( i \)\(^{th} \) node on inter-regional boundary are

\[
R_i^x = -\int_{\Omega_A} D_{\xi_A}^i \nabla \xi_A \cdot \nabla \phi_i \, d\Omega_A - \int_{\Omega_B} D_{\xi_B}^i \nabla \xi_B \cdot \nabla \phi_i \, d\Omega_B \\
+ \int_\Gamma \left[ D_{\xi_A}^i (n_A \cdot \nabla \xi_A) + D_{\xi_B}^i (n_B \cdot \nabla \xi_B) \right] \phi_i \, d\Gamma,
\]

where

\[
\Gamma = \Gamma_0 + \Gamma_A + \Gamma_B.
\]
B.1. MESH BOUNDARY CONDITIONS FOR REGION INTERFACES

1.2 Boundary condition for inter-regional boundary

\[ \eta_A \rightarrow \eta_B, \quad \xi_A \rightarrow \xi_B, \]

Figure 2: Example of “regular” connection between subdomains

Figure B.2: Two-types of subdomain connections in the pentagon lid-driven cavity flow (Fig. B.1). Note that, in (a), coordinate potential are changed across inter-regional boundary: \( \xi_A \) becomes \( \xi_B \), and \( \eta_A \) becomes \( \eta_B \), and, in (b), coordinate potentials are changed across inter-regional boundary: \( \xi_A \) becomes \( -\eta_B \), and \( \eta_A \) becomes \( \xi_B \).
B.1. MESH BOUNDARY CONDITIONS FOR REGION INTERFACES

\[ R_{i}^{y} = - \int_{\Omega_{A}} D_{\xi_{A}} \nabla \xi_{A} \cdot \nabla \phi_{i} \, d\Omega_{A} - \int_{\Omega_{B}} D_{\xi_{B}} \nabla \xi_{B} \cdot \nabla \phi_{i} \, d\Omega_{B} \]

\[ + \int_{\Gamma} \left[ D_{\xi_{A}}^{A} (n_{A} \cdot \nabla \xi_{A}) + D_{\xi_{B}}^{B} (n_{B} \cdot \nabla \xi_{B}) \right] \phi_{i} \, d\Gamma, \tag{B.5} \]

where subscript \( A \) and \( B \) mean variables are associated with element \( A \) and \( B \), and superscript of coordinate potential diffusivity stands for the element where the diffusivity was evaluated. Note that \( \xi_{A} \) and \( \eta_{A} \) are the same as \( \xi_{B} \) and \( \eta_{B} \).

When continuity of “ether” flux across the boundary is considered, the boundary integral terms in Eq. (B.5) vanishes, because

\[ D_{\xi_{A}}^{A} (n_{A} \cdot \nabla \xi_{A}) + D_{\xi_{B}}^{B} (n_{B} \cdot \nabla \xi_{B}) = 0, \tag{B.6} \]

\[ D_{\xi_{A}}^{A} (n_{A} \cdot \nabla \eta_{A}) + D_{\eta_{B}}^{B} (n_{B} \cdot \nabla \eta_{B}) = 0. \tag{B.7} \]

Note that the outward normal vectors, \( n_{A} \) and \( n_{B} \), take care of the direction of flux.

In Fig. B.2(a) case, inter-regional boundary is aligned to \( \eta \) direction. Therefore “ether” flux continuity on \( \xi \) direction, Eq. (B.6), helps to change element size across the boundary smoothly (continuity of element size), and “ether” flux continuity on \( \eta \) direction, Eq. (B.7), helps to align coordinate potential across the boundary (continuity of inclination of coordinate potential).

“Irregular” subdomain connection, like subdomains between subdomain I and III of the pentagon lid-driven cavity as in Fig. B.2(b) are considered, Eq. (B.5) cannot be used. Because \( \xi_{A} \) changes to \( \eta_{A} \), and \(-\eta_{B}\) changes to \( \xi_{B} \) across subdomain boundary, mesh residual equations is not straightforward to obtain. When only \( \xi_{A} \) and \( \eta_{A} \) are chosen to describe “ether” flux for both elements \( A \) and \( B \), one can describe residual equations without confusion:

\[ R_{i}^{y} = - \int_{\Omega_{A}} D_{\eta_{A}}^{A} \nabla \eta_{A} \cdot \nabla \phi_{i} \, d\Omega_{A} - \int_{\Omega_{B}} D_{\eta_{B}}^{B} \nabla \eta_{B} \cdot \nabla \phi_{i} \, d\Omega_{B} \]

\[ + \int_{\Gamma} \left[ D_{\eta_{A}}^{A} (n_{A} \cdot \nabla \eta_{A}) + D_{\eta_{B}}^{B} (n_{B} \cdot \nabla \eta_{B}) \right] \phi_{i} \, d\Gamma, \tag{B.8} \]

\[ R_{i}^{y} = - \int_{\Omega_{A}} D_{\eta_{A}}^{A} \nabla \eta_{A} \cdot \nabla \phi_{i} \, d\Omega_{A} - \int_{\Omega_{B}} D_{\eta_{B}}^{B} \nabla \eta_{B} \cdot \nabla \phi_{i} \, d\Omega_{B} \]

\[ + \int_{\Gamma} \left[ D_{\eta_{A}}^{A} (n_{A} \cdot \nabla \eta_{A}) + D_{\eta_{B}}^{B} (n_{B} \cdot \nabla \eta_{B}) \right] \phi_{i} \, d\Gamma. \]
The “ether” flux continuity conditions changes to

\[ D^A_{\xi_A} (\mathbf{n}_A \cdot \nabla \xi_A) + D^B_{\xi_B} (\mathbf{n}_B \cdot \nabla \xi_A) = 0 \]  
(B.9)

\[ D^A_{\eta_A} (\mathbf{n}_A \cdot \nabla \eta_A) + D^B_{\xi_B} (\mathbf{n}_B \cdot \nabla \eta_A) = 0. \]  
(B.10)

Again, boundary integral term in Eq. (B.8) vanishes as well by Eqs. (B.9) and (B.10).

Because, \( \xi_B \) and \( \eta_B \) are the same as \( \eta_A \) and \( -\xi_A \) respectively, Eq. (B.8) can be represented in terms of coordinate potential for each element that happened to be coincide with computational domain coordinate in the elliptic mesh generation system for each element:

\[
R_x^i = -\int_{\Omega_{0,A}} D^A_{\xi_A} \left( \frac{\partial y}{\partial \eta_A} \frac{\partial \phi_i}{\partial x} - \frac{\partial x}{\partial \eta_A} \frac{\partial \phi_i}{\partial y} \right) d\xi_A d\eta_A - \int_{\Omega_{0,B}} D^B_{\xi_B} \left( \frac{\partial y}{\partial \eta_B} \frac{\partial \phi_i}{\partial x} - \frac{\partial x}{\partial \eta_B} \frac{\partial \phi_i}{\partial y} \right) d\xi_B d\eta_B
\]

\[
R_y^i = -\int_{\Omega_{0,A}} D^A_{\eta_A} \left( \frac{\partial y}{\partial \xi_A} \frac{\partial \phi_i}{\partial x} - \frac{\partial x}{\partial \xi_A} \frac{\partial \phi_i}{\partial y} \right) d\xi_A d\eta_A - \int_{\Omega_{0,B}} D^B_{\eta_B} \left( \frac{\partial y}{\partial \xi_B} \frac{\partial \phi_i}{\partial x} - \frac{\partial x}{\partial \xi_B} \frac{\partial \phi_i}{\partial y} \right) d\xi_B d\eta_B
\]

Note that the sign of internal contribution of \( \Omega_B \) in \( R^x_i \) changes due to the fact that increasing \( \eta_B \) direction is the same as decreasing \( \xi_A \) direction, and the physical coordinate derivative terms are not affected by change in choice of computational domain coordinate.

In typical finite element programming, residual equation are evaluated in element-wise and are summed up at last. The mesh residual contributions from element \( A \) are evaluated using \( \xi_A \) and \( \eta_A \), and the other contributions from element \( B \) are evaluated.
B.1. MESH BOUNDARY CONDITIONS FOR REGION INTERFACES

using $\xi_B$ and $\eta_B$. For simplicity, each elemental contribution can be represented as,

$$R_{i}^{x,A} = - \iint_{\Omega_{A}} D_{\xi_A}^{A} \left( \frac{\partial y}{\partial \eta_A} \frac{\partial \phi_i}{\partial x} - \frac{\partial x}{\partial \eta_A} \frac{\partial \phi_i}{\partial y} \right) d\xi_A d\eta_A$$

$$R_{i}^{y,A} = - \iint_{\Omega_{A}} D_{\eta_A}^{A} \left( \frac{\partial y}{\partial \xi_A} \frac{\partial \phi_i}{\partial x} - \frac{\partial x}{\partial \xi_A} \frac{\partial \phi_i}{\partial y} \right) d\xi_A d\eta_A$$

$$R_{i}^{x,B} = - \iint_{\Omega_{B}} D_{\xi_B}^{B} \left( \frac{\partial y}{\partial \eta_B} \frac{\partial \phi_i}{\partial x} - \frac{\partial x}{\partial \eta_B} \frac{\partial \phi_i}{\partial y} \right) d\xi_B d\eta_B$$

$$R_{i}^{y,B} = - \iint_{\Omega_{B}} D_{\eta_B}^{B} \left( \frac{\partial y}{\partial \xi_B} \frac{\partial \phi_i}{\partial x} - \frac{\partial x}{\partial \xi_B} \frac{\partial \phi_i}{\partial y} \right) d\xi_B d\eta_B. \quad (B.12)$$

Note that $\xi$ and $\eta$ mesh equations are assigned to $x$ and $y$ residual.

Using Eq. (B.12), Eq. (B.11) can be written as

$$R_{i}^{x} = R_{i}^{x,A} - R_{i}^{x,B}$$

$$R_{i}^{y} = R_{i}^{y,A} + R_{i}^{x,B}. \quad (B.13)$$

In sum, elemental residual equations can be evaluated at each element like regular finite element programming. But when they are summed up at a “irregular” boundary, special care is required: the residual equations may need to be swapped or, sometimes, change the sign of them. Figure B.2(b) case requires both swapping equations and changing sign.

Corresponding Jacobian entries are

$$\frac{\partial R_{i}^{x}}{\partial x_j} = \frac{\partial R_{i}^{x,A}}{\partial x_j} - \frac{\partial R_{i}^{x,B}}{\partial x_j}$$

$$\frac{\partial R_{i}^{y}}{\partial y_j} = \frac{\partial R_{i}^{y,A}}{\partial y_j} - \frac{\partial R_{i}^{y,B}}{\partial y_j}$$

$$\frac{\partial R_{i}^{x}}{\partial x_j} = \frac{\partial R_{i}^{x,A}}{\partial x_j} + \frac{\partial R_{i}^{x,B}}{\partial x_j}$$

$$\frac{\partial R_{i}^{y}}{\partial y_j} = \frac{\partial R_{i}^{y,A}}{\partial y_j} + \frac{\partial R_{i}^{y,B}}{\partial y_j}. \quad (B.14)$$

The mesh generation result for the pentagon cavity flow with “ether” flux continuity boundary conditions are shown at Fig. B.3.
Figure B.3: Mesh of pentagon lid-driven cavity generated by diffusional elliptic mesh generation system using the ether flux continuity boundary condition.
B.1. MESH BOUNDARY CONDITIONS FOR REGION INTERFACES

1.3 Automatic elemental residual construction scheme for inter-regional boundary condition

When one uses a subdomain boundary numbering, or naming, scheme and computational coordinate direction as shown in Fig. B.4, mesh residual equations can be constructed following Table B.1 which is based on the difference of boundary numbers between two adjacent elements.

1.4 Corner condition at internal five region-confluence

Figure B.5 shows the effect of two different corner conditions on the center point location. According to the figure, neglecting contribution from the subdomain III (see Fig. B.1) results in better mesh than including all contributions.

**Figure B.4:** Subdomain boundary numbering scheme and computational coordinate direction used in GFEM program. Note that boundary number, or side number, increases with counter-clockwise direction and computational coordinate increase along left to right and top to bottom.
Table B.1: Residual equations for inter-regional node \( i \). Elements \( A \) and \( B \) are adjacent elements and share inter-regional boundary between them. \( N^b_A \) and \( N^b_B \) are boundary number for inter-regional boundary of element \( A \) and \( B \). See Fig. B.4. \( R^{x,i}_i, R^{y,i}_i, R^{x,i}_i \) and \( R^{x,i}_i \) are from Eq. (B.12).

<table>
<thead>
<tr>
<th>( \Delta N^b = N^b_A - N^b_B )</th>
<th>x residual equation</th>
<th>y residual equation</th>
</tr>
</thead>
<tbody>
<tr>
<td>2 or -2</td>
<td>( R^{x,i}_i = R^{x,i}_i + R^{x,i}_i )</td>
<td>( R^{y,i}_i = R^{y,i}_i + R^{y,i}_i )</td>
</tr>
<tr>
<td>0</td>
<td>( R^{x,i}_i = R^{x,i}_i - R^{x,i}_i )</td>
<td>( R^{y,i}_i = R^{y,i}_i - R^{y,i}_i )</td>
</tr>
<tr>
<td>1 or -3</td>
<td>( R^{x,i}_i = R^{x,i}_i - R^{x,i}_i )</td>
<td>( R^{y,i}_i = R^{y,i}_i + R^{y,i}_i )</td>
</tr>
<tr>
<td>-1 or 3</td>
<td>( R^{x,i}_i = R^{x,i}_i + R^{x,i}_i )</td>
<td>( R^{y,i}_i = R^{y,i}_i - R^{y,i}_i )</td>
</tr>
</tbody>
</table>

**Figure B.5:** Comparison between two corner conditions. (a) and (b) are zoomed mesh near the center vertex point of pentagon lid-driven cavity. In (a) case, elemental residual from all subdomain are summed up and contribute to the center vertex point, but in (b) case, elemental residual from subdomain III are excluded. Note that the position of the points are different in both case. the point in (a) is off to left, but the point in (b) locat at the center.
Appendix C

Filtering eigenvalues at infinity for linear stability analysis of two-layer plane incompressible flow

C.1 Introduction

Stability of parallel flow of parallel two-layer fluid is focused in many engineering technology fields. From macroscopic oil transportation (Hu and Joseph, 1989) to microscopically coating process (Severtson and Aidun, 1996), the nature of instability can be effectively studied by considering two-layer parallel Couette flow, Poiseuille flow, or combination of both. The linear stability of a given two-layer base flow is determined by tracking an infinitesimal disturbance of the flow and the interlayer that is placed between other fluid layers. Unlike a single fluid case, the flow system is susceptible to various instabilities. For example, not only viscosity difference between both layers but also density difference may bring the growing disturbance in the interlayer, so called the interfacial mode found by Yih (1967).

In many case, stability of the flow system consists of two inter-connected flow domains is described by Orr-Sommerfeld equation with streamfunction formulation. When the system is not belong to short-wavelength limit or long-wavelength limit, an analytical solution using a conventional perturbation scheme is difficult to get and the equation needs to be treated numerically. However, the equation is well-known for suffering parasitic growth problem or stiff eigenvalue problem, and this is a driving force for development of various numerical methods (Drzazin and Reid, 2004).
element method (Li and Kot, 1981; Yiantsios and Higgins, 1986), compound matrix method (Yiantsios and Higgins, 1988b), and spectral Chebyshev tau method (Su and Khomami, 1992; Severtson and Aidun, 1996) are usually used to overcome this problem. While the compound matrix method can only track a single eigenmode with a proper initial guess, the finite element method and the spectral method can be obtained the whole eigenspectrum at once without the initial guess.

The discretization of the system of linear differential equations that describe the infinitesimally small amplitude of the perturbation leads to a non-Hermitian, generalized eigenvalue problem (GEVP), \( Jc = \omega M c \), where \( J \), \( M \), \( \omega \) and \( c \) are Jacobian, mass matrix, (generalized) eigenvalue and corresponding eigenvector, respectively. However, finding eigensolutions of a GEVP is challenging task. In general, accurate eigenvalues requires high degree of discretization that leads to large matrices. The size of the full eigenproblem rules out any algorithm that has to compute the whole eigenspectrum; rather, only the portion of the spectrum — typically, eigenvalues with larger real part or leading modes — is calculated. As Saad (1989) pointed out, various numerical techniques that are based on iterative methods, for example, subspace iteration method and projection method, for approximating the original eigenproblem to a small-sized problem. Typically, these methods are coupled with preconditioning, for example, exponential preconditioner (Christodoulou and Scriven, 1988) or Chebyshev acceleration technique (Saad, 1984) for amplifying leading modes.

Moreover, the eigenvalue computation of GEVP is susceptible to additional, but obviously non-physical, eigenvalues to the eigenspectrum, or called eigenvalues at infinity, because their moduli are unrealistically large and they come from inevitable perturbations of original GEVP problem during numerical computation. The simple ways to overcome this difficulty are mapping eigenvalues at infinity to specified values, shift-and-invert or “mapping” techniques (Goussis and Pearlstein, 1989), or evade direct computations of them, QZ algorithm (Golub and Loan, 1996) or LZ algorithm (Kaufman, 1975). But the dimension of the matrix problem does not changed by these methods, thus one has to combine these with the subspace or projection method to handle a large-scale problem.

In order to get full eigenspectrum efficiently, one has to decrease the dimension of matrices. For a single layer case, Orszag (1971) use Chebyshev spectral method on the stability of a plane Poiseuille flow, and solve the system by QR algorithm (Golub and Loan, 1996) with “reducing” method (Gary and Helgason, 1970) that reduce the size of the original matrix problem produced by finite differencing scheme. Recently, Valerio et al. (2007) showed that the GEVP resulting from the linear stability analysis of viscous flows in Galerkin finite element framework can be reduced to a smaller non-singular eigenvalue problem. By exploiting the structure of the mass and Jacobian
C.2. FILTERING EIGENVALUES AT INFINITY

matrices, eigenvalues at infinity are filtered by an algebraic procedure.

Here, we revisit finite element method again for the linear stability analysis of a two-layer parallel flow, but with a different approach: the flow system for the stability analysis is formulated in terms of infinitesimal disturbances of primitive variables, i.e. velocity and pressure. Unlike fourth-order Orr-Sommerfeld equation that needs Hermite cubic basis function, primitive variables can be expanded using quadratic basis function in our formulation. Expanding Valerio et al. (2007)’s theory, we propose an efficient way to filtering eigenvalues at infinity and decrease the size of the matrix problem for the two-layer flow case, simultaneously. We also check the validity of the results from our method by comparing with the results from other numerical methods.

We consider a two-layer parallel channel flow as shown in Fig. 5.2 for the linear stability analysis. Detail formulations and discretization by Galerkin’s method and finite element basis functions are discussed in Secs. 5.2.1 and 5.2.2. In sum, the set of differential equations leads to the generalized eigenvalue problem (GEVP)

\[ \mathbf{J}_c = \omega \mathbf{M}_c. \]  

(C.1)

The Jacobian and mass matrix entries are summarized in Tables 5.2 and 5.3.

The choice of dimensionless parameters and variables depends on the type of flow — either (nearly) Couette flow or (nearly) Poiseuille flow — and they are summarized in Table C.1.

C.2 Filtering eigenvalues at infinity with matrix transformation

The mass matrix \( \mathbf{M} \) is singular, because the continuity equation for incompressible fluid, no-slip boundary conditions and interfacial conditions, except kinematic condition, have no time derivative. Thus, the number of finite eigenvalues of generalized eigenvalue problem Eq. (C.1) is smaller than the dimension of the problem \( 6N + 5 \). The missing eigenvalues are commonly referred to as \textit{eigenvalues at infinity}, because if the mass matrix is slightly perturbed to remove the singularity, e.g. \( \mathbf{M}' = \mathbf{M} + \epsilon \mathbf{I} \), large eigenvalues appear in the spectrum, and they grow unbounded as \( \epsilon \to 0 \) (Valerio et al. 2007). During the numerical computation of the eigenspectrum, truncation errors and round-off errors may cause perturbations of the mass matrix and lead to the eigenvalues at infinity — unrealistically large eigenvalues — of the GEVP.
C.2. FILTERING EIGENVALUES AT INFINITY

### Table C.1: Dimensionless variables used in stability analysis

<table>
<thead>
<tr>
<th>Name</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reynolds number</td>
<td>( N_{Re,C} = \frac{\rho_2 U_w H_2}{\mu_2} ) \quad ( N_{Re,P} = \frac{\rho_2 U_2^P(z=0) H_2}{\mu_2} )</td>
</tr>
<tr>
<td>Viscosity ratio</td>
<td>( m = \frac{\mu_1}{\mu_2} )</td>
</tr>
<tr>
<td>Density ratio</td>
<td>( r = \frac{\rho_1}{\rho_2} )</td>
</tr>
<tr>
<td>Thickness ratio</td>
<td>( n = \frac{H_1}{H_2} )</td>
</tr>
<tr>
<td>Dimensionless pressure gradient</td>
<td>( N_{G,C} = \frac{(dP/dx) H_2^2}{\mu_2 U_w} ) \quad ( N_{G,P} = \frac{(dP/dx) H_2}{\rho_2 U_2^P(z=0)^2} )</td>
</tr>
<tr>
<td>Interfacial tension number</td>
<td>( N_{T,C} = \frac{\sigma}{\mu_2 U_w} ) \quad ( N_{T,P} = \frac{\sigma}{\rho_2 H_2 U_2^P(z=0)^2} )</td>
</tr>
<tr>
<td>Froude number</td>
<td>( N_{F,C} = \frac{\rho_2 (d-1) g H_2^2}{\mu_2 U_w} ) \quad ( N_{F,P} = \frac{\rho_2 (d-1) g H_2^2}{U_2^P(z=0)^2} )</td>
</tr>
</tbody>
</table>

The eigenvalues \( \omega \) of Eq. (C.1) are the roots of the characteristic polynomial \( p(\omega) = \det(J - \omega M) \). In other words, it is the same as to find the value of \( \omega \) such that the homogeneous system \((J - \omega M)c = 0\) has non-trivial solution \(c\). One may put another matrix pair \(\tilde{J}\) and \(\tilde{M}\) in place of the matrix pair \(J\) and \(M\) for the new GEVP \((\tilde{J} - \omega \tilde{M})\tilde{c} = 0\) with new non-trivial solution \(\tilde{c}\) but with the same value of \(\omega\). \(\tilde{J}\) and \(\tilde{M}\) can be constructed by multiplying \(\omega\)-independent, full-ranked matrices \(X\) and \(Y\) to left and right side of both \(J\) and \(M\), like \(\tilde{J} = XJY\) and \(\tilde{M} = XMY\). The matrices \(X\) and \(Y\) arises while solving \(Jc = \omega Mc\) with a two-sided Gaussian elimination, in the sense that row and column operations are allowed. The non-trivial solutions of the original system \((J - \omega M)c = 0\) and the transformed system \((\tilde{J} - \omega \tilde{M})\tilde{c} = 0\) are related by \(Y\tilde{c} = c\). Therefore one can recover the solution of the original system only by simple matrix multiplications to the solution of the transformed system.

#### C.2.1 The algorithm

The matrix transformation to filter eigenvalue at infinity are summarized as below:

1. Order Jacobian and mass matrix entries and partition the matrices,
C.2. FILTERING EIGENVALUES AT INFINITY

2. Eliminate no-slip boundary conditions,

3. Eliminate parts of interfacial conditions (shear stress continuity, velocity continuity),

4. Transform matrices to “condense” eigenvalue informations to small block matrix.

After ordering entries of the matrix system and using the numbering scheme as in Fig. 5.3, both the mass and Jacobian matrices can be partitioned in $5 \times 5$ block structure with square blocks along the diagonal. The configuration of the matrices is shown below.

\[
M = \begin{pmatrix}
M_{11} & M_{12} & 0 & M_{14} & M_{15} \\
M_{22} & M_{22} & 0 & M_{24} & M_{25} \\
0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 \\
\end{pmatrix}
\]

\[
J = \begin{pmatrix}
J_{11} & J_{12} & J_{13} & J_{14} & J_{15} \\
J_{21} & J_{22} & J_{23} & J_{24} & J_{25} \\
J_{31} & J_{32} & 0 & J_{34} & J_{35} \\
J_{41} & J_{42} & 0 & J_{44} & J_{45} \\
0 & 0 & 0 & 0 & I_{[4]} \\
\end{pmatrix}
\]

Note that most of the subblocks are zero, especially in the mass matrix, and the last block of Jacobian $I_{[4]}$ is identity matrix of dimension four.

Algebraic equations associated with Dirichlet boundary conditions — no-slip conditions — do not have a time derivative, and the perturbed velocity field vanishes at these boundaries. In the matrix configuration of Eq. (C.2), they are located at the last blocks of $J$ and $M$, and they are identity matrix and zero matrix, respectively. Therefore they can be eliminated from the system without affecting the eigenspectrum.

After elimination of the rows and columns related to no-slip boundary conditions, upper $4 \times 4$ subblocks of the mass matrix $M^b$ and the Jacobian matrix $J^b$ remains. It is convenient to redefine the block structure of $B = J^b - \omega M^b$ as below:

\[
B = \begin{pmatrix}
B_{11}(\omega) & B_{12}(\omega) & B_{13} & B_{14}(\omega) \\
B_{21}(\omega) & B_{22}(\omega) & B_{23} & B_{24}(\omega) \\
B_{31} & B_{32} & 0 & B_{34} \\
B_{41} & B_{42} & 0 & B_{44} \\
\end{pmatrix}
\]

Note that $B_{44}$ is always invertible when $J^b$ is not singular. Because $4 \times 6N + 1$ size sub block $[J_{41}, J_{42}, 0, J_{44}]$ should have rank 3, but $J_{41}$ and $J_{42}$ are always rank deficient matrix. Exploiting this fact, one can construct transformation matrix $T_r^{(1)}$ to eliminate
block matrices $B_{14}$ and $B_{41}$.

$$\mathbf{\tilde{B}} = \begin{pmatrix}
\tilde{B}_{11}(\omega) & \tilde{B}_{12}(\omega) & \tilde{B}_{13} & B_{14}(\omega) \\
\tilde{B}_{21}(\omega) & \tilde{B}_{22}(\omega) & B_{23} & B_{24}(\omega) \\
\tilde{B}_{31} & \tilde{B}_{32} & 0 & B_{34} \\
0 & 0 & 0 & B_{44}
\end{pmatrix} = \mathbf{B} \mathbf{T}_{r}^{(1)} \quad \text{(C.4)}$$

where

$$\mathbf{T}_{r}^{(1)} = \begin{pmatrix}
I_{[2N]} & 0 & 0 & 0 \\
0 & I_{[2N-2]} & 0 & 0 \\
0 & 0 & I_{[2N]} & 0 \\
-B_{44} & -B_{41} & -B_{44} & B_{42} & 0 & I_{[3]}
\end{pmatrix} \quad \text{(C.5)}$$

When we take of first $3 \times 3$ blocks of $\mathbf{\tilde{B}}$, then

$$\mathbf{A} = \begin{pmatrix}
\tilde{B}_{11}(\omega) & \tilde{A}_{12}(\omega) & \tilde{B}_{13} \\
\tilde{B}_{21}(\omega) & \tilde{B}_{22}(\omega) & B_{23} \\
\tilde{B}_{31} & \tilde{B}_{32} & 0
\end{pmatrix}.$$

\text{(C.6)}$$

Since the matrix $\mathbf{T}_{r}^{(1)}$ is a lower triangular matrix with diagonal entries equals to one, its determinant is equal to one. The characteristic polynomial of $\mathbf{\tilde{B}}$ is equal to the original polynomial of $\mathbf{B}$,

$$p_{\mathbf{\tilde{B}}}(\omega) = \det \mathbf{\tilde{B}} = \det \mathbf{B} \det \mathbf{T}_{r}^{(1)} = \det \mathbf{B} = p_{\mathbf{B}}(\omega). \quad \text{(C.7)}$$

Furthermore, the characteristic polynomial of $\mathbf{\tilde{B}}$ is related to that of $\mathbf{A}$,

$$p_{\mathbf{\tilde{B}}}(\omega) = \det \mathbf{\tilde{B}} = \det \mathbf{B}_{44} \det \mathbf{A} = \det \mathbf{B}_{44} p_{\mathbf{A}}(\omega). \quad \text{(C.8)}$$

Because $\mathbf{B}_{44}$ does not have $\omega$, i.e. its determinant is just a number, roots of characteristic polynomial of $\mathbf{\tilde{B}}$ are the same as roots of $\det \mathbf{A}$. It does not affect the eigenspectrum of $\mathbf{A}$. Note that the rank of $\mathbf{\tilde{B}}, \mathbf{B}$, and $\mathbf{J}^{\theta}$ are the same, because $\mathbf{B}_{44}$ is invertible.

Now, the matrix structure become similar to a plane single-layered flow with Newtonian fluid [Valerio et al., 2007]. As mentioned in their work, $\tilde{B}_{31}$ and $\tilde{B}_{13}$ are invertible, when matrix $\mathbf{\tilde{B}}$ is nonsingular. Therefore, one can construct matrices $\mathbf{T}_{r}^{(2)}$ and $\mathbf{T}_{l}^{(2)}$ such that eliminate $\tilde{B}_{32}$ and $\tilde{B}_{23}$ from $\mathbf{A}$, when Jacobian is not singular, there exist matrices:

$$\tilde{\mathbf{A}} = \begin{pmatrix}
\tilde{B}_{11}(\omega) & \tilde{A}_{12}(\omega) & \tilde{B}_{13} \\
\tilde{A}_{21}(\omega) & \tilde{A}_{22}(\omega) & 0 \\
\tilde{B}_{31} & 0 & 0
\end{pmatrix} = \mathbf{T}_{l}^{(2)} \mathbf{A} \mathbf{T}_{r}^{(2)} \quad \text{(C.9)}$$
C.2. FILTERING EIGENVALUES AT INFINITY

where

\[
T_l^{(2)} = \begin{pmatrix}
I_{[2N]} & 0 & 0 \\
-B_{23} & I_{[2N-2]} & 0 \\
0 & 0 & I_{[2N]}
\end{pmatrix} \quad T_r^{(2)} = \begin{pmatrix}
I_{[2N]} & -\tilde{B}_{31}^{-1} \tilde{B}_{32} & 0 \\
0 & I_{[2N-2]} & 0 \\
0 & 0 & I_{[2N]}
\end{pmatrix}
\]  \hspace{1cm} (C.10)

Likewise in Eq. (C.7), the multiplication of \(T_l^{(2)}\) and \(T_r^{(2)}\) does not change the spectrum of \(A\):

\[
p_\tilde{A}(\omega) = \det \tilde{A} = \det T_l^{(2)} \det A \det T_r^{(2)} = \det A = \pm p_A(\omega).
\]  \hspace{1cm} (C.11)

Furthermore, one can rewrite the characteristic polynomial of \(\tilde{A}\) in terms of block matrices:

\[
p_\tilde{A}(\omega) = \det \tilde{A} = \det \tilde{B}_{31} \det \tilde{\tilde{A}}_{22}(\omega) \det B_{13} = \det \tilde{B}_{31} \det B_{13} p_{A_{22}}(\omega).
\]  \hspace{1cm} (C.12)

Again, \(\tilde{B}_{31}\) and \(B_{13}\) do not have \(\omega\), so the eigenspectrum of \(\tilde{A}\) are the same as that of \(\tilde{A}_{22}\).

From the relationship between the characteristic polynomials of transformed matrix, Eq. (C.7) and (C.11), the final relationship between original matrix and final transformed matrix becomes

\[
p_{A_{22}}(\omega) = \pm \frac{p_\tilde{A}(\omega)}{\det \tilde{B}_{31} \det B_{13}} = \pm \frac{p_B(\omega)}{\det B_{44} \det \tilde{B}_{31} \det B_{13}}.
\]  \hspace{1cm} (C.13)

Therefore the eigenspectrum related to \(\tilde{A}_{22} = \tilde{J}_{22} - \sigma \tilde{M}_{22}\) is the same as the original generalize eigenvalue problem Eq. (C.1) when the original Jacobian matrix is nonsingular.

The smaller GEVP which contain the finite portion of the spectrum of the original can be written as

\[
\tilde{J}_{22} c_2 = \omega \tilde{M}_{22} c_2
\]  \hspace{1cm} (C.14)

where \((2N-2) \times (2N-2)\) matrices \(\tilde{M}_{22}\) and \(\tilde{J}_{22}\) are given by

\[
\tilde{J}_{22} = \left( -J_{23} J_{13}^{-1} \tilde{J}_{22} \right) \left( -J_{31}^{-1} \tilde{J}_{32} \right) + \left( -J_{23} J_{13}^{-1} \tilde{J}_{22} \right) J_{12} + \tilde{J}_{22},
\]  \hspace{1cm} (C.15)

\[
\tilde{M}_{22} = \left( -J_{23} J_{13}^{-1} \tilde{M}_{22} \right) \left( -J_{31}^{-1} \tilde{J}_{32} \right) + \left( -J_{23} J_{13}^{-1} \tilde{M}_{22} \right) M_{12} + \tilde{M}_{22},
\]
and matrices with bar are computed by

\[
\begin{align*}
\bar{J}_{11} &= -J_{14} \bar{J}_{44}^{-1} J_{41} + J_{11}, \\
\bar{J}_{12} &= -J_{14} \bar{J}_{44}^{-1} J_{42} + J_{12}, \\
\bar{J}_{21} &= -J_{14} \bar{J}_{44}^{-1} J_{41} + J_{21}, \\
\bar{J}_{22} &= -J_{14} \bar{J}_{44}^{-1} J_{42} + J_{22}, \\
\bar{J}_{31} &= -J_{34} \bar{J}_{44}^{-1} J_{41} + J_{31}, \\
\bar{J}_{32} &= -J_{34} \bar{J}_{44}^{-1} J_{42} + J_{32}, \\
\bar{M}_{11} &= -J_{14} \bar{J}_{44}^{-1} M_{41} + M_{11}, \\
\bar{M}_{12} &= -J_{14} \bar{J}_{44}^{-1} M_{42} + M_{12}, \\
\bar{M}_{21} &= -J_{14} \bar{J}_{44}^{-1} M_{41} + M_{21}, \\
\bar{M}_{22} &= -J_{14} \bar{J}_{44}^{-1} M_{42} + M_{22}.
\end{align*}
\]

As in discussed by Valerio et al. (2007), both \(\bar{J}_{22}\) and \(\bar{M}_{22}\) are invertible, when the original Jacobian \(J\) is non-singular. Therefore the eigenvalue \(\omega\) can be obtained by either

\[
\bar{M}_{22}^{-1} \bar{J}_{22} c_2 = \omega c_2, \quad (C.17)
\]

or

\[
\bar{J}_{22}^{-1} \bar{M}_{22} c_2 = \frac{1}{\omega} c_2. \quad (C.18)
\]

In most of our computations, \(\bar{M}_{22}\) is usually well-conditioned, i.e. small condition number, comparing with \(\bar{J}_{22}\). One may attempt to use Eq. (C.17) instead of Eq. (C.18), because the error from the inversion of \(\bar{M}_{22}\) may be less than that from the inversion of \(\bar{J}_{22}\). However, the accuracy of eigenvalue depends not only on the condition of matrix but also on the conditioning of eigenvalue.

Let the original matrix \(A\) is perturbed proportional to the matrix \(E\) with an arbitrary parameter \(t\): \(A(t) = A + tE\) — then the eigenvalue problem becomes \(A(t) c(t) = \lambda(t) c(t)\), where \(\lambda(t)\) is a eigenvalue perturbed from the original eigenvalue \(\lambda\). When \(t\) is small enough, the perturbed eigenvalue can be approximated as \(\lambda(t) = \lambda + (d\lambda/dt) t\). According to the eigenvalue perturbation theory (Golub and Loan 1996), the sensitivity of eigenvalue for simple eigenvalue \(|d\lambda/dt|\) can be bounded as

\[
\left| \frac{d\lambda}{dt} \right| \leq \frac{\|E\|_2 \|c_f\|_2 \|c_r\|_2}{\|c_f \cdot c_r\|_{\kappa(\lambda)}}.
\]

(C.19)
where $\mathbf{c}_l$ and $\mathbf{c}_r$ are the left and right eigenvector of $\mathbf{A}$, and $\kappa(\lambda)$ is the condition number for eigenvalue $\lambda$. It is reasonable to approximate that the $L_2$ norm of the perturbation $\|\mathbf{E}\|_2$ for the matrix depends on the $L_2$ norm of the original matrix $\|\mathbf{A}\|_2$: $\|\mathbf{E}\|_2 \approx \epsilon_m \|\mathbf{A}\|_2$, where $\epsilon_m$ is a machine epsilon (Anderson et al., 1999).

Using these relationships, one can rewrite estimated error bound of the computed eigenvalue as below. When Eq. (C.17) is considered, the error bound become

$$|\omega(t) - \omega| \leq \epsilon_m \|\tilde{\mathbf{M}}_{22}^{-1} \tilde{\mathbf{J}}_{22}\|_2 \kappa(\omega). \quad (C.20)$$

For Eq. (C.18), the error bound is roughly approximated as

$$\left|\frac{1}{\omega(t)} - \frac{1}{\omega}\right| \leq \epsilon_m \|\tilde{\mathbf{J}}_{22}^{-1} \tilde{\mathbf{M}}_{22}\|_2 \kappa\left(\frac{1}{\omega}\right) \implies |\omega(t) - \omega| \leq \epsilon_m \frac{\|\tilde{\mathbf{J}}_{22}^{-1} \tilde{\mathbf{M}}_{22}\|_2}{|\omega(t)|^2} \kappa(\omega), \quad (C.21)$$

when $\omega(t)$ is close to $\omega$. Note that Eq. (C.17) and (C.18) share the same left and right eigenvectors, i.e. their condition numbers are the same. Hence, there are possibility that Eq. (C.18) can give better results than Eq. (C.17), especially when the matrix norm is small. During the computation, we sometimes, found spurious wiggles of the most dangerous growth rate versus wavenumber plot from Eq. (C.17) in small wavenumber regime. However, in most case, the resulting values from both equations do not show significant differences.

Most numerical methods for computing eigenvalue are based on an iterative scheme, like QR iteration based on Schur decomposition. The convergence property and accuracy of computed eigenvalues depend largely on the separation between eigenvalues (Golub and Loan, 1996). In other words, an eigenvalue $\omega_i$ having a large modulus ratio to the closet one $\omega_{i+1}$, a large value of $|\omega_i|/|\omega_{i+1}|$, will shows a better accuracy and convergence rate. The separation between eigenvalues can be adjusted easily by the shift-and-invert method:

\[
\left(\tilde{\mathbf{J}}_{22} - \delta \tilde{\mathbf{M}}_{22}\right)^{-1} \tilde{\mathbf{M}}_{22} \mathbf{c}_2 = \frac{1}{\omega - \delta} \mathbf{c}_2, \quad (C.22)
\]

where $\delta$ is shift factor. This enable to get a large separation of eigenvalues near $\delta$. Because the size of the reduced eigenvalue problem is smaller than the original one, the shift-and-invert method applied on the reduced problem can save significant amount of computational time. Note that Eq. (C.18) is a special case of Eq. (C.22).

In this study, the original and reduced GEVP, Eq. (C.1) and Eq. (C.14), are solved by the LAPACK subroutine ZGGEV. For solving reduced eigenvalue problem, Eq. (C.17), (C.18), or (C.22), eigenvalues are computed by subroutine ZGEEV.
C.2. Recover original generalized eigenvector

The generalized eigenvectors \( \mathbf{c} \) of the original GEVP, Eq. (C.1) may be recovered from the eigenvector \( \mathbf{c}_2 \). The procedure of proposed method is the reverse process of the transformation. The overall scheme can be re-written as

\[
\hat{A}\hat{c} = \begin{pmatrix}
\hat{A} & T^{(2)}_{l}\hat{B} \\
0 & B_{44}
\end{pmatrix}
\begin{pmatrix}
\hat{c}_1 \\
\mathbf{c}_4
\end{pmatrix}
= \begin{pmatrix}
T^{(2)}_l & 0 \\
0 & I
\end{pmatrix}
\begin{pmatrix}
A & \hat{B} \\
0 & B_{44}
\end{pmatrix}
\begin{pmatrix}
T^{(2)}_r & 0 \\
0 & I
\end{pmatrix}
\begin{pmatrix}
\hat{c}_1 \\
\mathbf{c}_4
\end{pmatrix}
= \tilde{T}^{(2)}_l B T^{(1)}_r \tilde{T}^{(2)}_r \hat{c} = 0,
\] 

(C.23)

where \( \hat{c}_1 = [c_1, c_2, c_3]^T \) and \( \hat{B} = [B_{14}(\omega), B_{24}(\omega), B_{34}]^T \). Since the original GEVP without boundary conditions is \( B\mathbf{c} = 0 \), the relationship between the original generalized eigenvector is related to the reduced one: \( \mathbf{c}^b = T^{(1)}_r \tilde{T}^{(2)}_r \hat{c} \). Note that \( \tilde{T}^{(2)}_l \) is invertible.

The eigenvector of \( \hat{A}\hat{c} = 0 \) may be written as

\[
\begin{cases}
\tilde{B}_{11}(\omega)c_1 + \tilde{A}_{12}(\omega)c_2 + B_{13}c_3 + B_{14}(\omega)c_4 = 0, \\
\tilde{A}_{21}(\omega)c_1 + \tilde{A}_{22}(\omega)c_2 + \tilde{A}_{24}(\omega)c_4 = 0, \\
\tilde{B}_{31}c_1 + \tilde{B}_{34}c_4 = 0, \\
\tilde{B}_{44}c_4 = 0.
\end{cases}
\] 

(C.24)

where \( \tilde{A}_{12}(\omega) = -(\tilde{J}_{11} - \omega \tilde{M}_{11})\tilde{J}^{-1}_{31}\tilde{J}_{32} + (\tilde{J}_{12} - \omega \tilde{M}_{12}) \). As we discussed before \( B_{14} \) and \( \tilde{B}_{31} \) is non-singular when Jacobian is invertible. Hence, \( B_{14}c_4 = 0 \) has a trivial solution \( c_4 = 0 \). For the same reason, \( c_1 = 0 \). This leads the second equation to the reduced GEVP, Eq. (C.14), \( \tilde{A}_{22}(\omega)c_2 = 0 \). \( c_3 \) can be obtained from the first equation: \( c_3 = -B_{13}^{-1}\tilde{A}_{12}(\omega)c_2 \). Therefore, the eigenvector of \( B\mathbf{c} = 0 \) can be written in terms of the eigenvector of the reduced eigenproblem \( \mathbf{c}_2 \) as

\[
\mathbf{c}^b = T^{(1)}_r \tilde{T}^{(2)}_r \begin{pmatrix}
0 \\
\mathbf{c}_2 \\
-B_{13}^{-1}\tilde{A}_{12}(\omega)c_2 \\
0
\end{pmatrix}
= \begin{pmatrix}
-\tilde{B}_{31}^{-1}\tilde{B}_{32}c_2 \\
\mathbf{c}_2 \\
-\tilde{B}_{13}^{-1}\tilde{A}_{12}(\omega)c_2 \\
B_{44}^{-1}B_{41}\tilde{B}_{31}\tilde{B}_{32}c_2 - B_{44}^{-1}B_{42}c_2
\end{pmatrix}.
\] 

(C.25)

From this equation, the original generalized eigenvector can be directly recovered from the reduced eigenvector.
C.2. FILTERING EIGENVALUES AT INFINITY

C.2.3 An example: the two-layer flow system with four elements

As an illustration, we consider first only four elements $N = 4$, two for the first layer and two for the second layer. The number of unknown coefficients for velocity, pressure, and interfacial height are $2(2N+2) = 20$, $2N = 8$, and 1, respectively. After elimination of no-slip conditions, the matrix structure becomes Fig. C.2(a) with the labeling of entries follows Fig. C.1. The next step is to eliminate the blocks $B_{41}$ and $B_{42}$ using the transformation using the matrix defined in Eq. (C.5). In practice, the inverse operation does not occur explicitly for evaluating $B_{41}^{-1}B_{41}$ and $B_{41}^{-1}B_{42}$. Instead, Gauss elimination was performed on the rectangular matrices $[B_{41},B_{44}]^T$ and $[B_{42},B_{44}]$ in order to decrease number of operations for minimizing round-off error.

![Figure C.1](image-url): Numbering scheme for 4 elements, 10 nodes and 29 degrees of freedom: two elements for both layers, 10 for the $x$-velocity $U$, 10 for the $z$-velocity $W$, 6 for the pressure $P$, and 1 for the interfacial height $h$. The 29 related coefficients $C_1, \ldots, C_{29}$ are inserted in the vector following the order described in Fig. 5.3.
C.2. FILTERING EIGENVALUES AT INFINITY

After the first transformation, the second transformation performed on the upper 3 × 3 block A as in Fig. C.3(a). Similarly, row and column permutation-based Gauss elimination is used to compute \( \mathbf{B}_{22} \mathbf{B}_{13}^{-1} \) and \( \tilde{\mathbf{B}}_{32}^{-1} \tilde{\mathbf{B}}_{32} \) efficiently. The structure of the resulting transformed matrix \( \tilde{\mathbf{A}} \) is shown in Fig. C.3(b). Note that all the information on the eigenspectrum boiled down to the (2\( N - 2 \)) × (2\( N - 2 \)) central block in the transformed matrix \( \tilde{\mathbf{A}}_{22}(\omega) \). In this example, the dimension of the central block matrix is 6. See Fig. C.3(b).

C.2.4 Comparison between matrix transformation and full QZ method

Here, we compare the results from the reduced eigenvalue problem and the original generalized eigenvalue problem. For the original problem, QZ method was chosen to solve Eq. (C.1). For now, FullQZ stands for this solution method. In matrix transformation method, three different methods are used and compared: QZ method to Eq. (C.14) (MTwQZ), QR method to Eq. (C.18) (MTwQR(J)) and QR method to Eq. (C.17) (MTwQR(M)).

Total number of element used in this test is 200, each layer has 100 elements, and

![Figure C.2: Non-zero entries after the eliminating no-slip conditions (a) and after the first matrix transformation Eq. (C.5) (b).](image-url)

343
corresponding degree of freedom is 1207. Flow is majorly driven by the moving substrate and distorted by relatively small pressure gradient: Reynolds number $N_{Re,C}$ viscosity ratio $m$, density ratio $d$ and thickness ratio $n$ are 2.61, 0.5, 1.0, and 1.5, respectively. Dimensionless pressure gradient $N_{G,P}$ is 0.196. There is no gravity in this system, i.e. Froude number $N_{F,C}$ is zero. All dimensionless numbers are based on Table C.1

Table C.2 shows the comparison between each method. Except MTwQZ, MTwQR and FullQZ have similar results in the most dangerous growth rate, in either high and low wavenumber. For example in $a = 0.1$ case, MTwQZ method seems to fail to predict an accurate growth rate as indicated by estimated error bound. Note that the condition number $\kappa(\omega_{MD})$ for MTwQZ is better than the original one, but it does not guarantee small error bound $\epsilon_h(\omega_{MD})$. Even though the condition number is similar to others, the estimated error bound $\epsilon_h(\omega_{MD})$ for MTwQR(J) is extremely small. Therefore one may conclude that MTwQR(J) is the best way to compute the reduced eigenvalue problem. In terms of computational time, MTwQZ and MTwQR are about 10 times faster than FullQZ. MTwQR(J) is the fastest of all.

The most dangerous growth rate versus wavenumber plot are shown in Fig. C.4. The results show that matrix transformation method with QZ iteration fails to get the most dangerous eigenvalue at small wavenumber regime. At high wavenumber, three

![Figure C.3](image_url)

**Figure C.3:** Non-zero entries for before (a) and after (b) the second matrix transformation, Eq. (C.10). Note that the mid block become full matrix after finishing matrix transformation.
### C.2. FILTERING EIGENVALUES AT INFINITY

**Table C.2:** Comparison between full QZ method and matrix transformation method. $\omega_{MD,R}$ means the most dangerous growth rate or real part of the eigenvalue, $\kappa(\omega_{MD})$ stands for condition number of the eigenvalue, and $\epsilon_b(\omega_{MD})$ is estimated error bound from Eq. (C.19) with approximating $\|E\|_2 \approx \epsilon_m \|A\|_2$. Total number of element is 200. Note that condition number for eigenvalue are computed from LAPACK expert driver subroutine (Anderson et al., 1999): `ZGEEVX` for QR iteration and `ZGGEVX` for QZ iteration.

<table>
<thead>
<tr>
<th></th>
<th>MTwQZ</th>
<th>MTwQR(J)</th>
<th>MTwQR(M)</th>
<th>Full QZ</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>CPU time (sec)</strong></td>
<td>10.8</td>
<td>7.2</td>
<td>7.3</td>
<td>80.2</td>
</tr>
<tr>
<td><strong>$\alpha = 0.1$ case</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\omega_{MD,R}$ (sec$^{-1}$)</td>
<td>$-1.0661 \times 10^{-1}$</td>
<td>$6.0527 \times 10^{-3}$</td>
<td>$6.0551 \times 10^{-3}$</td>
<td>$6.0544 \times 10^{-3}$</td>
</tr>
<tr>
<td>$\kappa(\omega_{MD})$</td>
<td>$1.759 \times 10^1$</td>
<td>$8.475 \times 10^1$</td>
<td>$8.478 \times 10^1$</td>
<td>$8.467 \times 10^4$</td>
</tr>
<tr>
<td>$\epsilon_b(\omega_{MD})$</td>
<td>$1.6719 \times 10^1$</td>
<td>$2.5129 \times 10^{13}$</td>
<td>$6.5698 \times 10^{-3}$</td>
<td>$3.207 \times 10^{-8}$</td>
</tr>
<tr>
<td><strong>$\alpha = 10$ case</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\omega_{MD,R}$ (sec$^{-1}$)</td>
<td>$4.0195 \times 10^1$</td>
<td>$4.0196 \times 10^1$</td>
<td>$4.0191 \times 10^1$</td>
<td>$4.0193 \times 10^1$</td>
</tr>
<tr>
<td>$\kappa(\omega_{MD})$</td>
<td>$1.862 \times 10^1$</td>
<td>$4.163 \times 10^3$</td>
<td>$4.161 \times 10^3$</td>
<td>$1.629 \times 10^2$</td>
</tr>
<tr>
<td>$\epsilon_b(\omega_{MD})$</td>
<td>$1.770 \times 10^{-3}$</td>
<td>$5.4627 \times 10^{-12}$</td>
<td>$2.892 \times 10^{-2}$</td>
<td>$1.632 \times 10^{-10}$</td>
</tr>
</tbody>
</table>
methods show similar results. Ten most dangerous eigenvalues in the eigenspectrum (Fig. C.5) also reveal similar trends: predictions from MTwQZ are different from the others. The trends clearly support that solving Eq. (C.17) or (C.18) is better than solving Eq. (C.16), as we discussed before. Therefore one can conclude that either MTwQR(J) or MTwQR(M) saves computational times and compute an accurate value at the same time.

The most dangerous growth rate computed in Fig. C.4 has disturbances mainly on the interlayer, so called the interfacial mode. One way to visualize this disturbance is to plot each perturbed field in eigenvector separately. Figure C.6 shows modulus of entries in the eigenvector at the most dangerous eigenvalue $\omega_{MD} = 6.0527 \times 10^{-1} - i 4.8055 \times 10^2$ with the same operating condition used in Table C.2. Modulus of entries in the eigenvector stands for the magnitude of disturbances. The fact that all peaks of disturbances are located at the interlayer $z = 0$ supports that the most dangerous disturbance is interfacial mode. The velocity component $u$ and pressure $p$ clearly show the jump across the interlayer, as expected in the velocity continuity and normal stress balance condition at the interlayer. Note that the recovered eigenvectors from the method discussed in Sec C.2.2 have virtually the same moduli as the original eigenvectors.

![Figure C.4: Comparison between three eigenvalue solving methods: MTwQZ, MTwQR, and FullQZ. Here MTwQR stands for solving Eq. (C.18): Because MTwQR(J) and MTwQR(M) show virtually the same results, the plot shows only one of them. Note that MTwQZ shows strange patterns at low wavenumber.](image)
Table C.3: Comparison of most dangerous growth rate with different number of elements in matrix transformation method at wavenumber: $1.0 \times 10^{-3} \text{mm}^{-1}$, $10 \text{mm}^{-1}$ and $1.0 \times 10^3 \text{mm}^{-1}$.

<table>
<thead>
<tr>
<th>Number of elements</th>
<th>most dangerous growth rate ($\text{sec}^{-1}$)</th>
<th>$\alpha = 1.0 \times 10^{-3} \text{mm}^{-1}$</th>
<th>$\alpha = 10 \text{mm}^{-1}$</th>
<th>$\alpha = 1000 \text{mm}^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>$6.05569 \times 10^{-7}$</td>
<td>$40.1932$</td>
<td>$9.76113 \times 10^{-2}$</td>
<td></td>
</tr>
<tr>
<td>100</td>
<td>$6.05397 \times 10^{-7}$</td>
<td>$40.1938$</td>
<td>$9.70265 \times 10^{-2}$</td>
<td></td>
</tr>
<tr>
<td>200</td>
<td>$6.03895 \times 10^{-7}$</td>
<td>$40.1664$</td>
<td>$9.69402 \times 10^{-2}$</td>
<td></td>
</tr>
<tr>
<td>300</td>
<td>$6.12567 \times 10^{-7}$</td>
<td>$40.1702$</td>
<td>$9.68871 \times 10^{-2}$</td>
<td></td>
</tr>
</tbody>
</table>

C.2.5 Effect of elements on eigenvalue

In order to examine the effect of elements on eigenvalue, three different wave numbers, $1.0 \times 10^{-3} \text{mm}^{-1}$, $10 \text{mm}^{-1}$ and $1.0 \times 10^3 \text{mm}^{-1}$, are chosen for comparison between different number of elements. Table C.3 shows the most dangerous growth rate for each number of elements, and Fig. C.7 represents ten most dangerous modes in the eigen-spectrum at wavenumber $\alpha = 10 \text{mm}^{-1}$. Surprisingly, even small number of elements gives reasonable growth rates compared with finer mesh flow systems.

Figure C.5: Ten most dangerous leading eigenvalues for $\alpha = 0.1$ computed from different methods: MTwQZ, MTwQR(J), MTwQR(M), and FullQZ. Except the results from MTwQZ, rest of them close to the original generalized eigenvalue problem.
C.2. FILTERING EIGENVALUES AT INFINITY

![Graphs of u, v, and p](image)

(a) Parallel velocity component $u$.

(b) Transversal velocity component $v$.

(c) Pressure $p$.

Figure C.6: Modulus of the velocity component and pressure across the flow direction $z$ related to the most dangerous growth rate $\omega_{MD} = 6.0527 \times 10^{-1} - i4.8055 \times 10^{2} \text{ sec}^{-1}$. “Original” and “Reduced” stand for the eigenvector from the original generalized eigenproblem and the reduced eigenproblem. Especially, the reduced eigenvector computed by solving Eq. (C.18). Number of element is 100, and wavenumber is $1 \text{ mm}^{-1}$. Interlayer is located at $z = 0$. The jump of velocity component and pressure across the interlayer are shown in (a) and (d).
C.2.6 Compare with numerical results

The compound matrix method was formulated by Ng and Reid (1979) and Yiantsios and Higgins (1988b) expand the method to two-layer parallel flow case. In general, the method is superior to the conventional shooting method (Ng and Reid, 1979). Like the shooting method, it can track only single eigenmode and requires a “good” initial guess. As suggested by Yiantsios and Higgins (1988a), it is advantageous to use finite element method or finite difference method to get the whole eigenspectrum for the problem and then the calculation of a desired mode are refined using the compound matrix method. However, finite element formulation for Yiantsios and Higgins (1986) is not the same as our formulation: their equations are based on streamfunction formulation with two Orr-Sommerfeld equations. Here, we compare dimensionless wavespeed $c^* = \omega^*/-i \alpha^*$ between results from our method and the compound matrix method.

At the same time, we also examined the effect of node distribution on the interfacial mode. Fixed-width mesh used the same spacing between each node, and graded mesh

![Figure C.7: Ten most dangerous leading eigenvalues computed from different number of elements: 50, 100, 200, and 300. Eigenvalues are computed by solving Eq. (C.18), and wavenumber is $\alpha = 10\text{mm}^{-1}$.](image-url)
C.2. FILTERING EIGENVALUES AT INFINITY

Table C.4: Comparison between theoretical results and numerical model with different mesh scheme for dimensionless complex wave speed \( c^* \left( N_{Re,P} = 1, \ m = 5, \ n = 1, \ r = 1 \right) \). CM method means compound matrix method, and fixed-width mesh and graded mesh stand for fixed nodal spacing mesh and variable nodal spacing mesh, respectively. Number of element used in fixed-width mesh is 180 and the spacing between nodes are uniform. For graded mesh, 100 elements are used but nodes are concentrated near the interlayer.

<table>
<thead>
<tr>
<th>( \alpha^*N_{T,P} )</th>
<th>( \alpha^* )</th>
<th>( c^*(\text{CM method}) )</th>
<th>( c^*(\text{Fixed-width mesh}) )</th>
<th>( c^*(\text{Graded mesh}) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>10</td>
<td>0.99998 (-i0.008199)</td>
<td>1.000077 (-i0.008191)</td>
<td>0.999968 (-i0.008201)</td>
</tr>
<tr>
<td>1</td>
<td>20</td>
<td>0.99956 (-i0.041184)</td>
<td>1.000171 (-i0.004130)</td>
<td>0.99983 (-i0.004146)</td>
</tr>
<tr>
<td>2</td>
<td>10</td>
<td>0.99996 (-i0.016537)</td>
<td>1.000071 (-i0.016519)</td>
<td>0.999950 (-i0.016440)</td>
</tr>
<tr>
<td>2</td>
<td>20</td>
<td>0.99907 (-i0.008349)</td>
<td>1.000189 (-i0.008278)</td>
<td>0.999980 (-i0.008314)</td>
</tr>
</tbody>
</table>

used variable spacing using stretching functions (Vinokur, 1983a):

\[
\begin{align*}
  s \left( \frac{i}{N_l} \right) &= \frac{\tanh \left( \phi \left( \frac{i}{N_l} \right) \right)}{\tanh \phi} \quad \text{for layer 1,} \\
  s \left( \frac{i}{N_l} \right) &= 1 + \frac{\tanh \left( \phi \left( \frac{i}{N_l} - 1 \right) \right)}{\tanh \phi} \quad \text{for layer 2,}
\end{align*}
\]

where \( i \) stands for the nodal position number counted from bottom to top of a given layer, \( N_l \) is the number of nodal points inside the layer, and \( \phi \) is the strength of the node concentration: a higher value yields more mesh concentration near a specified point. Typically, we choose \( \phi \) as 2 for 100 elements case. Number of mesh points used in the fixed-width mesh and the graded mesh are 180 and 100, respectively.

Table C.4 shows the comparison. Even though the results from the graded mesh has less number of elements than the fixed-width mesh, it predicts wavespeed very close to the compound matrix method. Wavespeed for the graded mesh matched up to about 4 digit in real part and about 3 digit in complex part compared with the compound matrix method. Therefore we can conclude that properly chosen node distribution can help the accuracy of the eigenvalue of the given flow system. Furthermore, accuracy of the finite element method using primitive variable formulation is comparable to the compound matrix method.

We also compare the our results with results from spectral Chebyshev tau method. The spectral Chebyshev tau method was developed by Orszag (1971) to evaluate the linear stability for a plane Poiseuille flow, and Su and Khomami (1992) expand
### Table C.5: Comparison between numerical results by spectral tau method and numerical model for dimensionless complex wave speed $c^*$. Numerical results by spectral tau method were from Su and Khomami (1992).

<table>
<thead>
<tr>
<th>Viscosity ratio $m$</th>
<th>$c$ (Spectral method)</th>
<th>$c$ (G/FEM method)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>$2.71932 + i2.05300 \times 10^{-5}$</td>
<td>$2.71942 + i2.05346 \times 10^{-5}$</td>
</tr>
<tr>
<td>60</td>
<td>$2.56766 + i8.26909 \times 10^{-6}$</td>
<td>$2.56727 + i8.27006 \times 10^{-6}$</td>
</tr>
<tr>
<td>20</td>
<td>$2.06021 + i1.58952 \times 10^{-6}$</td>
<td>$2.06033 + i1.59030 \times 10^{-6}$</td>
</tr>
<tr>
<td>10</td>
<td>$1.67220 + i1.24810 \times 10^{-6}$</td>
<td>$1.67232 + i1.24896 \times 10^{-6}$</td>
</tr>
<tr>
<td>5</td>
<td>$1.33333 + i7.52439 \times 10^{-7}$</td>
<td>$1.33333 + i7.53490 \times 10^{-7}$</td>
</tr>
</tbody>
</table>

$\alpha = 1.0 \times 10^{-5}$, $N_{Re,C} = 10.0$, $r = 1$, $n = 1$, $N_{E,C} = 0$, $N_{T,C} = 0$

The technique to handle the stability for a two-layer plane flow. Table C.5 shows the results from both methods for a plane Couette two-layer flow. The analysis was performed at $N_{Re} = 10.0$, $r = 1$, $n = 1$, $N_{E,C} = 0$ and $N_{T,C} = 0$ with different $m$ and $\alpha$, and the growth rate are converted to the dimensionless wavespeed for comparison purpose. Note that the dimensionless number is based on Table C.1 and number of elements used in G/FEM mesh is 100 with concentrate node near the interlayer (graded mesh configuration). Similar to the previous comparison, wavespeed for the G/FEM matched up to about 4 digit in real part and about 3 digit in complex part compared with the spectral method.

### C.3 Final remarks

The method proposed in here is to formulate the linear stability analysis on a two-layer parallel flow in finite element framework and filter eigenvalues at infinity with matrix transformation. The algorithm, discussed in Sec. C.2, transforms the original generalized eigenvalue problem (GEVP) constructed by GFEM into a small-sized eigenvalue problem (EVP). The size of the transformed EVP is less than 1/3 of the
size of the original GEVP, but it contains all the finite eigenvalues of the original eigenspectrum. Also we propose the way to recover the original eigenvector from the transformed one in Sec. [C.2.2]

The main advantages of the new method are:

- Unlike computationally expensive preconditioning and mapping technique, eigenvalues at infinity are eliminated without changing the value of other finite eigenvalues inside the original eigenspectrum.

- No assumption is made during the transformation process, but the dimension of the transformed eigenvalue problem is less than one-third of that of the original.

- The reduced GEVP is non-singular and, consequently, the original GEVP can be rewritten as an EVP.

- The original invariant subspaces, or eigenspaces, can be easily recovered from simple matrix-vector multiplications.

The method proposed in this study decreases the computation cost of finding the whole eigenspectrum of an two-layer parallel incompressible flow significantly. In the particular example we solved, the method was about $10$ times faster than the conventional eigenvalue solving method of the original GEVP.