Kinetics of Alkane Dehydrogenation on Bulk Zirconia Catalysts

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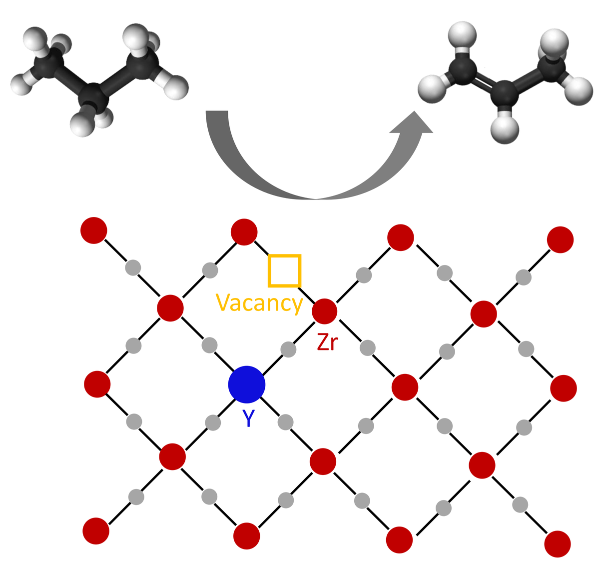
Introduction

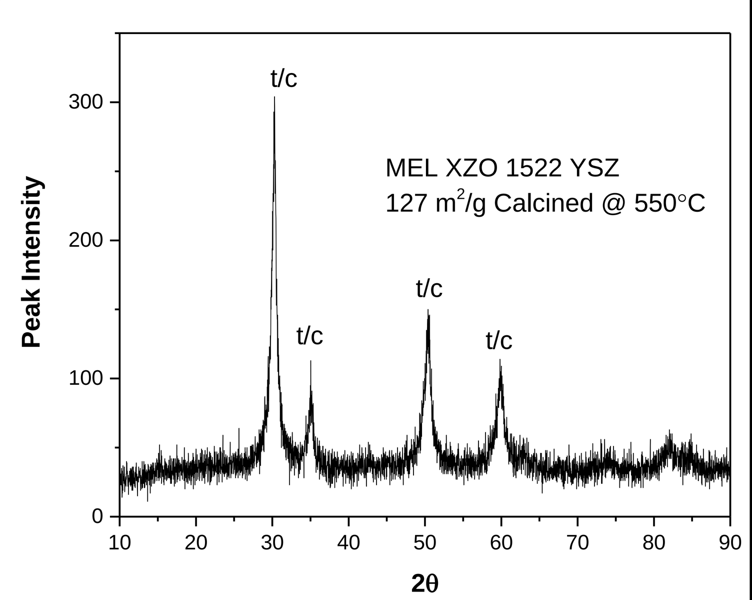


- Propane dehydrogenation accounts for more than 4 million tons of propylene produced annually
- Commercial Pt-based catalysts are too expensive, Cr-based catalysts highly toxic
- Bulk zirconia materials provide an inexpensive, abundant, and highly effective alternative to conventional propane dehydrogenation catalysts

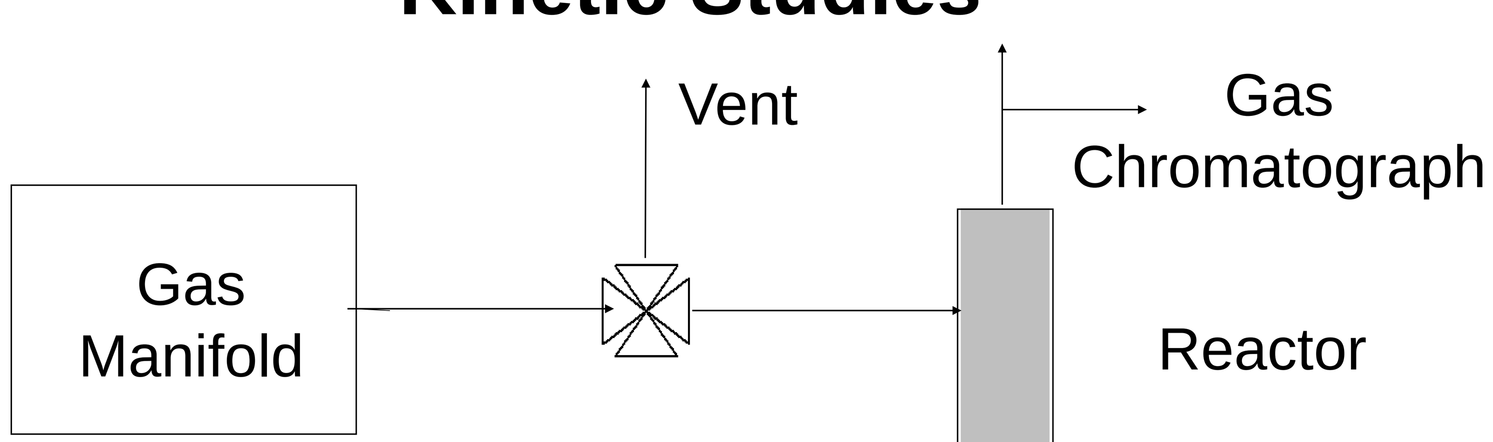
Experimental Methods

Catalyst

- 8.1 wt% Y-doped ZrO_2
- Surface Area: 127 m^2/g
- Tetragonal/cubic structure observed in XRD



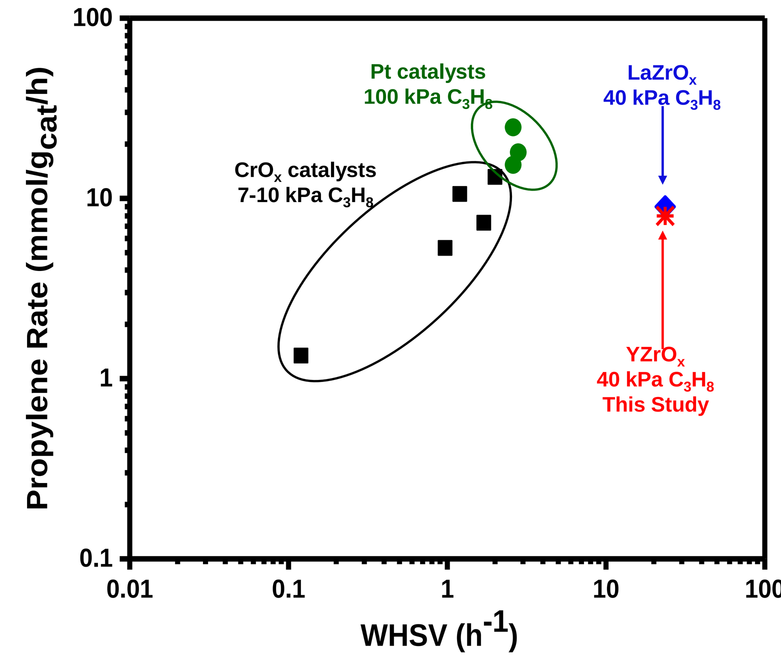
Kinetic Studies



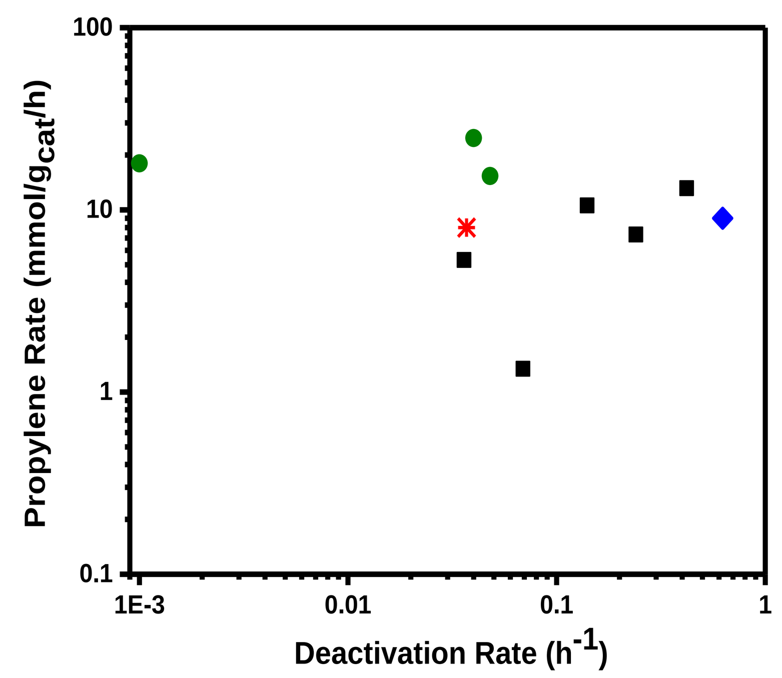
- 550°C reaction temperature
- 0-10 kPa propane pressure
- O_2 pretreatment at 550°C

Results

Catalyst Performance

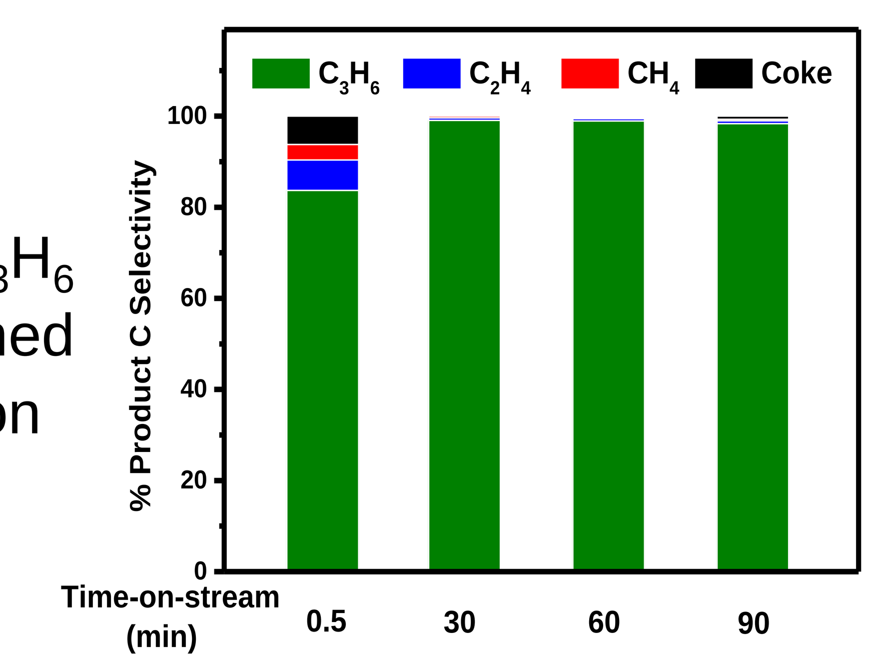


Even at higher space velocities, Zirconia catalysts exhibit rates comparable to commercial catalysts

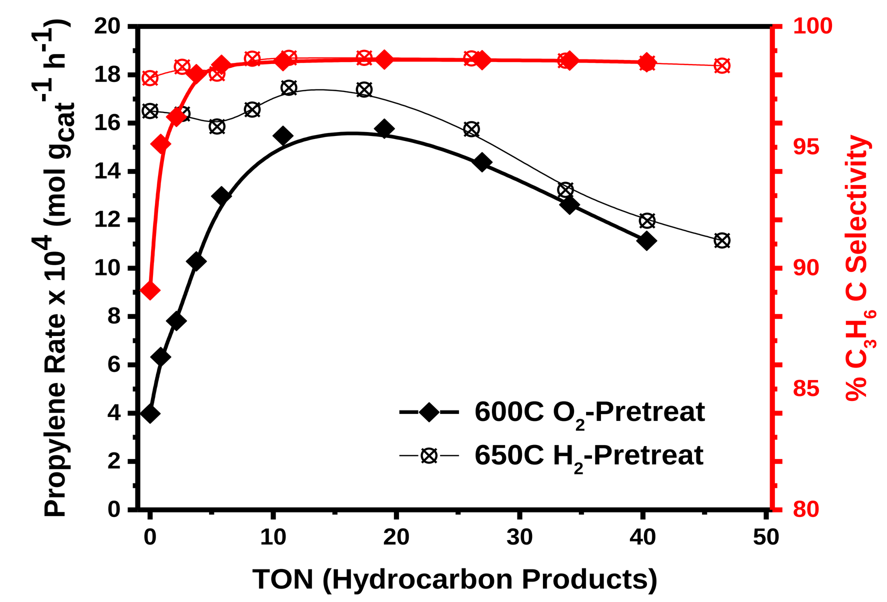


Deactivation rates are lower than most catalysts studied in the literature

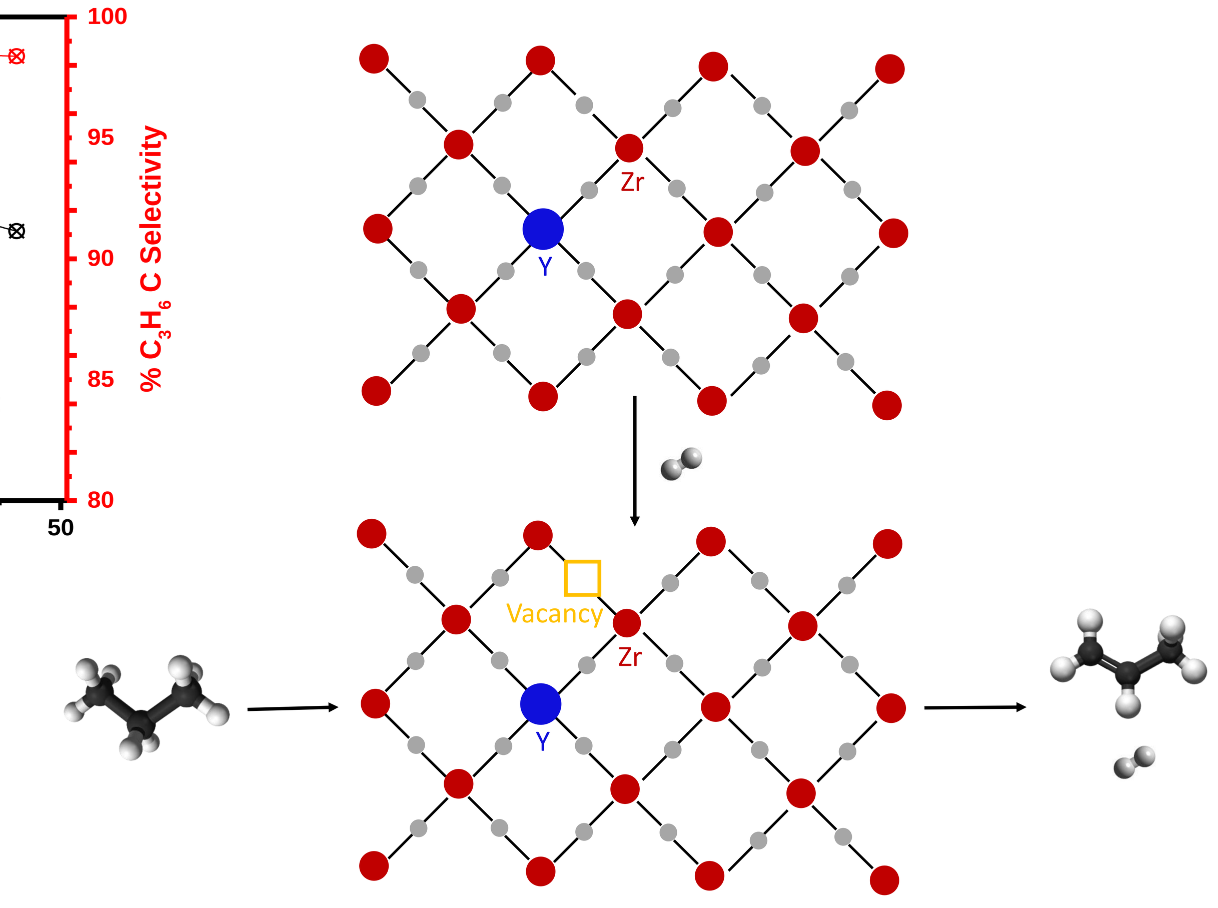
Exceptionally high C_3H_6 selectivity maintained despite the deactivation



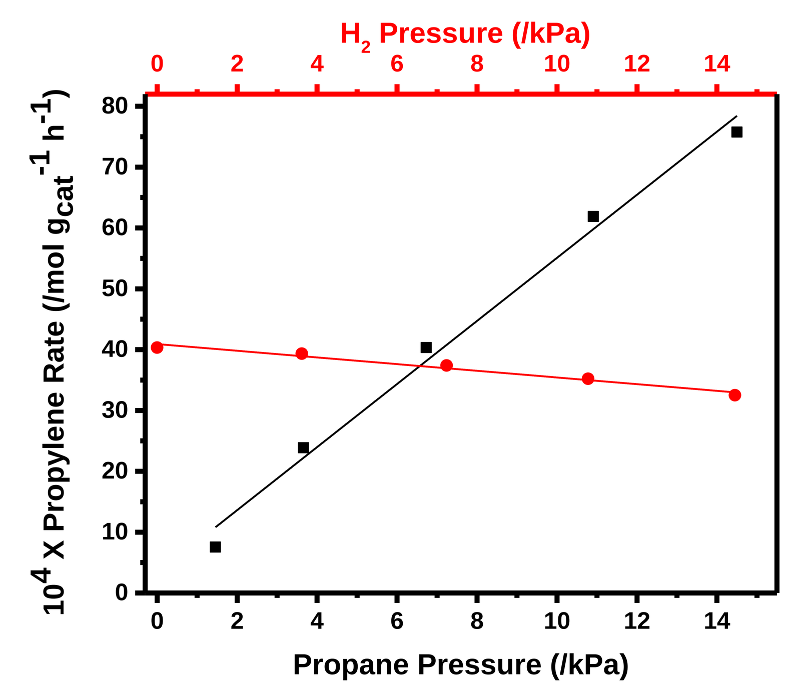
H_2 Pretreatment

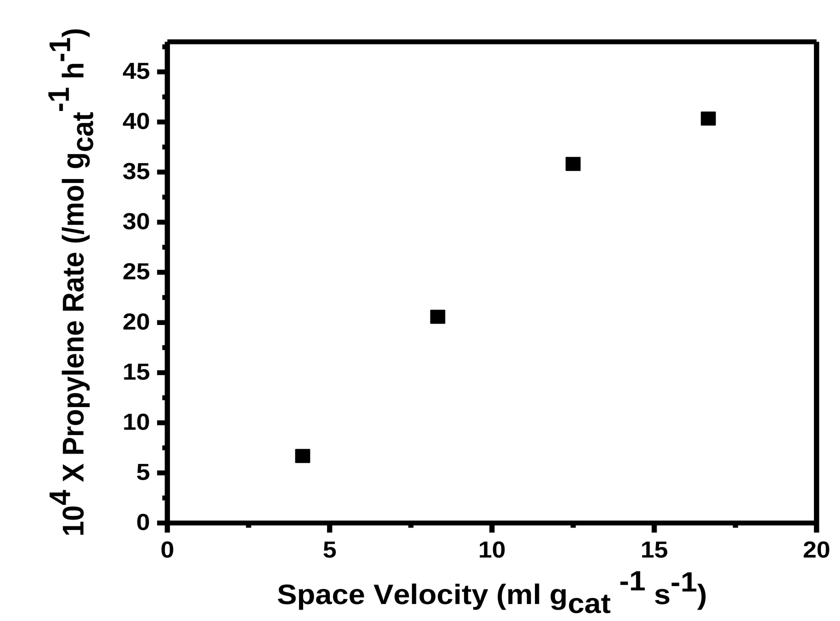


650°C H_2 pretreatment reduces the induction period by creating oxygen vacancies



Reaction Kinetics





- Rates are first order in propane and zero order in H_2
- Reaction orders imply propane coverage controls rates
- Product inhibition likely occurs on the catalyst surface

Conclusions

- Bulk zirconia materials are emerging, promising materials for alkane dehydrogenation applications
- Hydrogen generated during reaction helps generate oxygen vacancies that are responsible for dehydrogenation activity
- High-temperature H_2 pretreatment eliminates induction periods
- Reaction kinetics suggest that under the conditions of interest, propane surface coverage determines propylene synthesis rates

References

[1] Otroshchenko, T., *Angew. Chem*., Int. Ed 10.1002, 15880-15883 (2015)
[2] Caspary, H., *Handbook of Heterogeneous Catalysis*, Vol 7., 3206-3228 (2008)

Acknowledgements

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