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FLUIDS OF DIFFERENTIAL TYPE: CRITICAL REVIEW AND THERMODYNAMIC ANALYSIS.

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ABSTRACT

Thermodynamics, in the form of a dissipation inequality and the commonly accepted idea that the stored energy should have an extremum in equilibrium, is used to find restrictions on the response functions for the stress and the stored energy in incompressible fluids of differential type. For those special fluids of differential type known as *grade n fluids* these thermodynamic restrictions have been a source of some controversy and much confusion notwithstanding the fact that they are in complete harmony with results achieved by either linear or nonlinear stability analysis. In order to clarify the issues that seem to underlie this controversy, we provide an extended analysis of the genesis and development of fluids of differential type. As part of our analysis, we will see that certain ideas of flow retardation and model approximation have been consistently misinterpreted. Additionally, we establish several new results concerning the thermodynamics of these materials. A special application of our results reveals that work of Joseph (1981, 1990) and Renardy (1983) on the instability of the rest state for certain, very special grade n fluids is in fact inapplicable to all those grade n fluids that are consistent with thermodynamics.

0 PROLOGUE

Geological materials, liquid foams, polymeric fluids, slurries, and food products are among the many substances which are capable of flowing but which exhibit flow characteristics that cannot be adequately described by the classical linearly viscous fluid model. In order to describe some of the departures from Newtonian behavior evinced by such materials, many idealized material models have been suggested. One of the earliest class of such material models is what is now commonly referred to as *fluids of differential type** or, informally, as *Rivlin-Ericksen fluids*. In such materials, only a very short (in fact, infinitesimal) part of the history of the deformation gradient has an influence on the stress. More specifically, in an incompressible fluid of differential type, apart from a constitutively indeterminate pressure, the stress is just a function of the velocity gradient and some number of its higher time derivatives. These materials thus lack a gradually “fading memory”; instead, their “memory” fades precipitously: with the instantaneous cessation of all local motion, the stress becomes a pure pressure. One consequence of this is that, while they can experience the phenomenon of *creep*, these materials do not exhibit the phenomenon of *stress relaxation*. Nevertheless, due to their mathematical simplicity, a great deal of interest has been evinced in recent years in the thermomechanics and stability of such fluids. Unfortunately, the results of some of these studies have sometimes been interpreted in ways that suggest logical paradoxes, physical impossibilities, or conflicts between “reality” and theory. A closer scrutiny reveals that all

*For a precise definition, see our (2.1) or Rivlin & Ericksen (1955) or Truesdell & Noll (1965).

such “problems” with fluids of differential type are an illusion and stem from imprecise and inaccurate interpretations of both theoretical and experimental results.

In this paper, in addition to establishing some new theorems about the thermomechanics of fluids of differential type, we will attempt to clarify and dispel several of the confusions which frequently seem to impair understanding of the thermodynamics and stability of these materials.

Among fluids of differential type, one special subclass has received an especially large (perhaps even an inordinately large) amount of attention. This is the incompressible homogeneous fluid of grade 2, and much of the confusion surrounding general fluids of differential type arose from and is rooted in the misunderstood consequences of the thermodynamics and stability of this model. It is natural therefore to begin our discussion by reviewing the history of this rather special material. As we shall see, the false trails taken with it have been retraveled many times.

In Section 1 we present a critical historical survey of those issues that we feel bear most directly on the origins and the consequences of the confusion which surrounds grade 2 fluids. We think particularly remarkable the extent to which the theoretical implications of an experimental fact (*viz.* positive first normal stress data) have been consistently misconstrued. As we discuss at length in Section 1, instead of its sign, it is the *rate of growth* of the first normal stress difference with the shear rate that is the key issue for many fluids of laboratory interest. While occasionally experimenters have remarked that this growth rate is nonstandard for certain liquids, the point does not seem to have attracted the attention from theorists which we think it deserves. Although the matter is not studied in any detail here, we think that this nonstandard growth rate will prove to be an especially fertile area for future investigation.

In Sections 2 and 3 we carry out a thermodynamic analysis of the general fluid of differential type. We obtain constitutive restrictions on this material based on the hypotheses that a familiar dissipation principle holds and that the material possesses a stored energy function that is bounded either below or above. A noteworthy aspect of our analysis is that the restrictions we establish turn out to hold true even if we limit ourselves to the material’s behavior on just steady motions. As a consequence, our results not only complement those based on stability ideas, they also address questions of material response fundamentally inaccessible from a stability point of view.

Joseph (1981, 1990) and Renardy (1983) studied certain, highly special fluids of differential type and showed that for all of them the rest state was unstable. In Section 3 we show that any fluid of differential type which is thermodynamically consistent is necessarily forbidden from belonging to either the class studied by Joseph or the (somewhat larger) class studied by Renardy, *i.e.*, thermodynamically consistent fluids of differential type are to be found only among those materials *not* studied by Joseph and Renardy. From the vantage point of either stability theory or thermodynamics, the particular model materials examined by Joseph and Renardy are thus seen to be physically unnatural. Notwithstanding this, their results, like earlier, similar results of other authors, are sometimes interpreted in a way that incorrectly and unjustly impugns a far larger (sometimes the entire) class of fluids of differential type. One goal of our analysis and review in Section 1 is to expose the logical lacunae inherent in such interpretations. We close in Section 4 with some final, more general comments on the roots and consequences of the misunderstandings that have grown up around fluids of differential type. We will argue that there has been a consistent and long-standing pattern of misinterpretation of the retardation theorem of Coleman & Noll (1960) and that this has led to the adoption of certain flawed notions of model approximation.

We emphasize here, at the outset, that it is not a concern of this paper to argue for or against any particular model of non-Newtonian behavior. Far less is it our concern to argue for or against a particular model’s ability to capture “all of the essential physics” of this or that actual material (say, for example, a polymeric fluid). The manifest difficulty of such a program has been long recognized in the rheological community.[†] Nevertheless, given the misunderstandings that have grown up around them, a remark or two about the applicability of differential fluid models might not be out of place.

As we have already remarked, fluids of differential type cannot experience stress relaxation. One therefore would hardly expect them to be useful in modelling those aspects, if any, of a particular real material’s response that critically hinge on the material’s ability to stress relax. To again pick a concrete example (and to again give them the emphasis they have been given over the last 35 years), if all polymeric fluids are such that the capacity for stress relaxation is an ever present, though oftentimes latent, aspect of their response, then

[†]Thus, for example, Pipkin & Tanner (1972) remark that “In order to understand the physical applicability of the mathematical models that are used to describe viscoelastic fluids, it is absolutely essential to be aware that none of these models are applicable to all states of motion of any particular fluid. Many different forms of constitutive equations can be valid descriptions of the behavior of one fluid, depending on the circumstances, and one form of constitutive equation may describe the behavior of many different fluids in one particular kind of motion.”

differential type fluids would certainly seem to be inappropriate models for them. Rather than being used to model such “strongly viscoelastic” materials (if indeed that is what polymeric fluids are), fluids of differential type might well be better employed to model the usual materials involved in, say, slurry flows and food rheology, where relaxation effects frequently seem to be rather insignificant. Work by Man and his co-workers (1985, 1987, 1992) even suggests that polycrystalline ice might also be so modeled. Issues such as these are, of course, among the most difficult and most fundamental confronting us when it comes time to actually *apply* a given model. Here, however, our concerns are different and, in fact, preparatory to such questions of application.

1 HISTORICAL REVIEW AND COMMENTARY

The Cauchy stress $\mathbf{T}$ in an incompressible homogeneous fluid of grade 2 is related to the fluid motion by the constitutive equation

$$\mathbf{T} = -p\mathbf{1} + \mu\mathbf{A}_1 + \alpha_1\mathbf{A}_2 + \alpha_2\mathbf{A}_1^2, \quad (1.1)$$

where $-p\mathbf{1}$ is the spherical part of the stress due to the constraint of incompressibility and where μ , α_1 , and α_2 are constant material moduli, μ being the viscosity. The Rivlin-Ericksen kinematical tensors $\mathbf{A}_1, \mathbf{A}_2, \dots, \mathbf{A}_n, \dots$, are defined recursively through

$$\mathbf{A}_1 = \mathbf{L} + \mathbf{L}^T, \quad (1.2)$$

and

$$\mathbf{A}_{n+1} = \overset{\Delta}{\mathbf{A}}_n \equiv \dot{\mathbf{A}}_n + \mathbf{A}_n\mathbf{L} + \mathbf{L}^T\mathbf{A}_n, \quad (1.3)$$

where $\mathbf{L} = \text{grad } \mathbf{v}$ denotes the spatial gradient of the velocity field, where a superposed dot denotes the material time derivative, and where a superposed triangle denotes the *convected* time rate. It can be shown that the $\mathbf{A}_i$ are objective or frame indifferent measures of the higher rates of material straining.*

The stress form (1.1), with μ , α_1 , and α_2 possibly depending on the norm of $\mathbf{A}_1$, was first obtained for a special class of *flows* by Criminale, Ericksen, & Filbey (1958), who showed that (1.1) sufficed to give the stress exactly in *any* fluid of differential type which is undergoing

*See, for example, Truesdell & Noll (1965) or Chadwick (1976).

what are now known as *steady, viscometric flows*. Later, in work that is often taken as justifying the use of (1.1) in *all* “slow enough” flows for a special class of *materials*, Coleman & Noll (1960) showed that (1.1) also emerged as the second order truncation of an *expansion* of the stress in a simple fluid with a particular kind of fading memory whenever a fixed, given deformation history is sufficiently *retarded*[†] — just as, they showed, the Navier-Stokes form for the stress arises as the first order truncation of such a *retardation expansion*. Neither result, of course, confers any special status on (1.1) when it is taken as a constitutive equation describing the stress-deformation response of a particular hypothetical material in general motions. However, since the form (1.1) is properly frame invariant, the above results can be (and certainly have been) taken as *motivating* the study of (1.1) as an exact model of material behavior. Viewing (1.1) as an exact model is, after all, precisely the approach adopted for its major special subcase, the Navier-Stokes fluid (given by $\alpha_1 = \alpha_2 = 0$). Indeed, in this context, Truesdell & Noll (1965) remark “... works on hydrodynamics generally regard the Navier-Stokes formula, not as approximate” or as restricted to some special class of flows, “but rather as an exact definition of a particular fluid, flows of which are not restricted to any approximating sequence” or special class. “... The fluid of second grade being defined likewise by a frame indifferent constitutive equation may be approached in the same way, not as an approximation, but rather as a special case offering particular interest.” Unfortunately, at least as regards the signs of its coefficients, (1.1) was not in the main fated to be so approached.

Working within a special constitutive theory that included classical linear viscoelasticity, Coleman & Markovitz (1964) established an interesting link between the material constant α_1 of (1.1) and the *shear relaxation modulus* $G(\cdot)$ which arises upon linearizing any one of their constitutive equations. What they showed is that, *for materials with a certain, special type of fading memory*, if the form (1.1) is the result of the expansion procedure of Coleman & Noll then the number α_1 is given by

$$\alpha_1 = - \int_0^\infty s G(s) ds. \quad (1.4)$$

Since $G(\cdot)$ is normally thought to be positive[‡], Coleman & Markovitz thus concluded that α_1 should be negative *whenever* (1.1) *occurs as an expansion of their special constitutive*

[†]Although it is physically rather more special than seems desirable, the Coleman & Noll notion of retardation corresponds to a mathematically precise way in which a given deformation history may be progressively more and more “slowed down”.

[‡]We know of no proof that $G(\cdot)$ (or, more to the point, its first moment) must be positive. Coleman & Markovitz (1964) make an honest, if slightly unsatisfying, appeal to “thermodynamic intuition” on this crucial point.

equations to terms of second order in a retardation parameter. While (1.4) is important within the particular theory of fading memory studied by Coleman & Markovitz, we will see in a moment that, when (1.1) is taken as exact, their analysis offers far less guidance for the sign of α_1 than at first might appear.

Now consider a (steady) *simple shearing flow* which in Cartesian coordinates x_1, x_2, x_3 has a velocity field of the form $v_1 = v(x_2)$, $v_2 = v_3 = 0$. When a general simple fluid[§] undergoes such a motion it can be shown that the constitutive response of the stress is completely characterized by three functions, $N_1(\cdot)$, $N_2(\cdot)$, and $\tau(\cdot)$, of the *shear rate* $\kappa \equiv v'(x_2)$. In terms of the Cartesian components of $\mathbf{T}$, these are, respectively, the *first and second normal stress differences*

$$N_1 = N_1(\kappa) \equiv T_{11} - T_{22} \quad ,$$

$$N_2 = N_2(\kappa) \equiv T_{22} - T_{33} \quad ,$$

and the *shear stress function*

$$\tau = \tau(\kappa) \equiv T_{12} \quad .$$

Collectively, we refer to the three functions $N_1(\cdot)$, $N_2(\cdot)$, and $\tau(\cdot)$ as the *viscometric* functions of our material, and we note that, when it is applied to a simple fluid, frame indifference requires $\tau(\cdot)$ to be an odd function and requires $N_1(\cdot)$ and $N_2(\cdot)$ to be even functions that vanish at 0. Further, since only the shear stresses do work in a simple shearing flow, the condition that the flow dissipate mechanical energy is easily seen to require that $\tau(\kappa)$ always be of the same sign as κ . The general laws of mechanics impose no known additional restrictions on either $N_1(\cdot)$, $N_2(\cdot)$, or $\tau(\cdot)$. Suppose, however, that $\tau(\cdot)$ is continuously differentiable at the origin, and suppose also that $N_1(\cdot)$ and $N_2(\cdot)$ are twice continuously differentiable at the origin. For such fluids, we can define the *first normal stress coefficient*, the *second normal stress coefficient*, and the *zeroth order viscosity*; these are just the respective numbers

$$\Psi_1 = \lim_{\kappa \rightarrow 0} \frac{N_1(\kappa)}{\kappa^2}, \quad \Psi_2 = \lim_{\kappa \rightarrow 0} \frac{N_2(\kappa)}{\kappa^2}, \quad \mu^0 = \lim_{\kappa \rightarrow 0} \frac{\tau(\kappa)}{\kappa}.$$

Set $\Phi_1 \equiv -\frac{1}{2}\Psi_1$ and $\Phi_2 \equiv \Psi_1 + \Psi_2$. With some abuse of language, we also will refer to Φ_1 and Φ_2 as, respectively, the first and second normal stress coefficients. Of course, in terms of Φ_1 , Φ_2 , and μ^0 , our assumptions of differentiability are precisely the assertions that

$$N_1 = N_1(\kappa) = -2\Phi_1\kappa^2 + o(\kappa^2) \quad ,$$

$$N_2 = N_2(\kappa) = (2\Phi_1 + \Phi_2)\kappa^2 + o(\kappa^2) \quad , \tag{1.5}_{1,2,3}$$

[§]See Truesdell & Noll (1965) for an exact definition. This concept includes all the fluids of Coleman & Markovitz as well as all those, like (1.1) when it is taken as exact, which are of differential type.

$$\tau = \tau(\kappa) = \mu^0 \kappa + o(\kappa).$$

These three equations are, as we say, no more than a re-writing of our *assumptions* of differentiability, assumptions that a given simple fluid may or may not satisfy. Indeed, not only do such assumptions exclude certain yield-like phenomena, they also embody a special ordering of effects as $|\kappa| \rightarrow 0$ that, as we shall see, may well be inappropriate for modelling the response of certain real materials.

Now, for the form (1.1), it is easy to calculate that

$$\begin{aligned} N_1 &= N_1(\kappa) = -2\alpha_1 \kappa^2, \\ N_2 &= N_2(\kappa) = (2\alpha_1 + \alpha_2) \kappa^2, \\ \tau &= \tau(\kappa) = \mu \kappa. \end{aligned} \tag{1.6}_{1,2,3}$$

Hence, for a grade 2 fluid, $\Phi_1 = \alpha_1$, $\Phi_2 = \alpha_2$, $\mu^0 = \mu$, and there are no higher order terms in either $N_1(\cdot)$, $N_2(\cdot)$, or $\tau(\cdot)$. In a grade 2 fluid, the three material moduli μ , α_1 , and α_2 completely determine the numbers μ^0 , Φ_1 , and Φ_2 , as they in fact completely determine the functions $N_1(\cdot)$, $N_2(\cdot)$, and $\tau(\cdot)$. In general, of course, nothing like this is to be expected: $N_1(\cdot)$ and $N_2(\cdot)$ need not be quadratic, $\tau(\cdot)$ need not be linear, and the numbers Φ_1 , Φ_2 , and μ^0 , although of course *determined* by the response function for the stress, may not themselves even *appear* in the response function for the stress (let alone appear in the simple, direct way they do in a grade 2 fluid).[¶]

Now, of course, a given simple fluid may fail to meet the smoothness hypotheses required by the Coleman & Noll theory of fading memory. Indeed, there are simple fluids — there are even fluids of differential type — that fail to satisfy the even weaker smoothness assumptions which are needed in order to just prove that (1.1) holds in the limit of sufficiently retarded deformation histories. For such materials, the standard retardation theorem fails to be applicable to their stress response in sufficiently “slow” flows. On the other hand, such materials may have very smooth viscometric functions $N_1(\cdot)$, $N_2(\cdot)$, and $\tau(\cdot)$ that govern their response in steady, simple shearing motions. There is no mystery in this: the retardation theorem requires the existence of *differentials* of the stress response function whereas only the existence of certain of its *directional derivatives* along steady, simple shearing deformation histories are required for the smoothness of $N_1(\cdot)$, $N_2(\cdot)$, and $\tau(\cdot)$. Thus, even though a form like (1.1) may fail to occur in a retardation expansion of the stress in certain materials, the numbers Φ_1 , Φ_2 , and μ^0 of (1.5)_{1,2,3} may well exist. On the other hand, for a material

[¶]As will be clear in a moment, this last point is vividly illustrated by the materials studied by Coleman & Markovitz.

that meets the hypotheses of the retardation theorem of Coleman & Noll, the expression (1.1) holds to terms of second order in a retardation parameter. Further, the family of steady, simple shearings is so special that it is now not hard to prove that for such materials the triad $(\alpha_1, \alpha_2, \mu)$ in the *truncation* (1.1) will be equal to the triad (Φ_1, Φ_2, μ^0) appearing in the differentiability assumptions $(1.5)_{1,2,3}$. Thus, for certain simple fluids, the lowest order terms in the viscometric functions serve to determine the lowest order terms in the retardation expansion. For all these fluids therefore, as with fluids of second grade, we would be justified in once and for all identifying the triad (Φ_1, Φ_2, μ^0) appearing in $(1.5)_{1,2,3}$ with the triad $(\alpha_1, \alpha_2, \mu)$ appearing in (1.1). Nevertheless, we shall not do this: not only would the resulting abbreviated notation then suggest an entanglement between (1.1) and $(1.5)_{1,2,3}$ that, as we have seen, is not general, it would also fail to address — indeed, it would even go towards blurring — the fact that the conceptual settings that typically surround (1.1) and $(1.5)_{1,2,3}$ are entirely different. Indeed, even when it is taken as “just an approximation” to some other fluid model,^{||} we usually have in mind — and this seems basic to the notion of a “constitutive equation” — that (1.1) delivers the stress (or some close enough approximation to the stress) for a much bigger (if not always well-specified) class of flows than just steady, simple shearings. In contrast, given the general setting of simple fluids, it is clear that the equations $(1.5)_{1,2,3}$ refer and are limited to just motions that are steady, simple shearings (or to motions that are, like steady, viscometric flows, constitutively equivalent to steady, simple shearings). Put another way: whether the *form* (1.1) be taken as exact or as in some sense an “approximation”, its use usually brings with it, if only tacitly, far reaching assumptions about material response, *i.e.*, about modelling; contrariwise, the *differentiability assumptions* $(1.5)_{1,2,3}$ — indeed, even the complete functional forms $N_1(\cdot)$, $N_2(\cdot)$, and $\tau(\cdot)$ — *by themselves* make no statement about material response *away* from steady, simple shearing flows, involve far milder assumptions about modelling, and certainly give nothing like (1.1) for more general flows.

For the materials with fading memory studied by Coleman & Markovitz, which have (1.1) as just the truncation to terms of second order of a retardation expansion, we have then that $(\Phi_1, \Phi_2, \mu^0) = (\alpha_1, \alpha_2, \mu)$. Thus, by (1.4), $\Phi_1 = \alpha_1 = -\int_0^\infty sG(s)ds$. Moreover, paralleling (1.4), Coleman & Markovitz (1964) also demonstrated that $\mu^0 = \mu = \int_0^\infty G(s)ds$, and that $\Phi_2 = \alpha_2$ is the double integral over $(0, \infty)$ of a certain other kernel function.** Coleman &

^{||}We in fact think that this is a highly problematic move. Although we pass over it for now, we will return to this point in Section 4.

**In an interesting note Joseph (1980) extended the procedure of Coleman & Markovitz. For their special materials, he found a hierarchy of inequalities which govern the coefficients that occur in retardation expansions. See also the abbreviated discussion in Joseph (1981).

Markovitz's materials are therefore somewhat special in that both of the numbers μ^0 and Φ_1 are completely determined by the shear relaxation modulus $G(\cdot)$. Moreover, it is obvious that in these materials the numbers Φ_1 , Φ_2 , and μ^0 generally will *not* appear in any explicit or direct way in the response function for the stress. Lastly, we see from $(1.5)_1$ that if Coleman & Markovitz's argument for the sign of α_1 is accepted then, at low enough shear rates, N_1 must be positive. This, along with a positive value for N_2 (see, however, below), is exactly what was found experimentally by Markovitz & Brown (1963) for a polyisobutylene-cetane solution over the range of shear rates they studied. With this seemingly perfect union of theory and experiment, the impression or idea arose — or so it seems from the distant vantage point of 25 years later — that the form (1.1) should be studied *only* when α_1 is negative.

However, the matter is not (and did not for long remain) so simple. In the first place — and these are merely two points of logic — while the result (1.4) of Coleman & Markovitz is important in delimiting the structure of (1.1) when it is viewed as arising from a process of flow retardation applied to a very special class of materials, it is clear, first, that the result (1.4) says nothing about the sign of α_1 when (1.1) arises in a retardation expansion for some *other* class of materials*, and, second, that, since (1.1) itself embraces no idea of fading memory (and so fails to even have associated with it the notion of a shear relaxation modulus $G(\cdot)$), the result (1.4) gives “no guidance at all for the sign of α_1 in any real second grade fluid”†, *i.e.*, in any material model for which (1.1) holds *exactly* in each given fluid flow. Illustrations of this last point were not long in coming. Indeed, a full year before the work of Coleman & Markovitz, Ting (1963) had studied several unsteady flows of a fluid modeled by (1.1) and had found that, while none of the problems he considered had a bounded solution when $\alpha_1 < 0$, the flows with $\alpha_1 > 0$ were in general much better behaved than their Navier-Stokes counterparts. A subsequent study by Coleman, Duffin, & Mizel (1965) examined in

*For example, Dunn (1982) has studied (1.1) when μ , α_1 , and α_2 are allowed to be arbitrary, smooth functions of temperature and $\mathbf{A}_1$. He finds that thermodynamics allows α_1 to depend on no more than the temperature and the *norm* of $\mathbf{A}_1$, *i.e.*, $\alpha_1 = \bar{\alpha}_1(|\mathbf{A}_1|^2)$, where $\bar{\alpha}_1(\cdot)$ must meet

$$\int_0^z \bar{\alpha}_1(\xi) d\xi \geq 0 \quad (\star)$$

for all $z > 0$, and where the temperature dependence has been suppressed. Clearly, when a process of flow retardation is applied to *this* fluid model, the constant modulus α_1 which will appear in (1.1) is just the number $\bar{\alpha}_1(0)$ which by $(\star)$ can only be *non-negative*. This, of course, is the *exact opposite* of the sign choice which Coleman & Markovitz felt was thermodynamically appropriate for the (different) materials they studied.

†Dunn & Fosdick (1974).

detail laminar shearing flow of a grade 2 fluid in a channel; in accord with the results of Ting, they found drastic instability and nonexistence results for the case $\alpha_1 < 0$.

The work of Ting (1963) and Coleman, Duffin, & Mizel (1965) thus established that the form (1.1), when taken as exact, must (at least) have $\alpha_1 \geq 0$. Or, put a little differently, the stability analyses of these authors demonstrated more than 25 years ago that physically realistic grade 2 fluid models must at least have $\alpha_1 \geq 0$ (and so have $N_1(\cdot) \leq 0$). However, as was the case with the fact that the result (1.4) says nothing about the sign of α_1 in an actual second grade fluid, there seems to have been little appreciation of this at the time. Instead, when they were appraised in the (blinding?) light cast off by both the experimental work of Markovitz & Brown (1963) and the mathematical analysis of Coleman & Markovitz (1964), the work of Ting, who focused on the case $\alpha_1 > 0$, seems to us to have been subsequently slighted, while the analysis of Coleman, Duffin, & Mizel seems to us to have elicited a remarkable number of false summations and confused “resolutions” of its core point. Thus, to pick but one example[‡], Metzner, White, & Denn (1966 a) assert, without qualification, that “Coleman et al. use the second-order Rivlin-Ericksen fluid as a constitutive equation and find no solution is possible,” (page 865), and that the “solution to the Coleman-Duffin-Mizel (CDM) paradox lies in the ... realization that the Rivlin-Ericksen formulation is an asymptotic approximation valid only for times of deformation which are large as compared to θ , a characteristic relaxation time of the fluid,” (page 865). In addition to completely ignoring the work of Ting while simultaneously impugning all fluids of second grade, irrespective of the nature of their coefficients, the first of these two quotes is just not true: Coleman, Duffin, and Mizel find that no *bounded* solution is possible *for certain channel widths if (and only if, as it turns out) $\alpha_1 < 0$* . This last qualification is absolutely critical since solutions both exist and are well-behaved for arbitrary channel widths if $\alpha_1 > 0$. The second quote, as we shall be arguing in our final section, is also false in that it does not really provide an explanation of the “(CDM) paradox”. Here, however, we especially wish to draw attention to the position advanced that “the Rivlin-Ericksen formulation is an asymptotic approximation ...”. As we discuss in Section 4, this position seems to be rather commonly held. Unfortunately, it is also anti-historical: the differential type fluids of Rivlin and Ericksen were laid down and formulated, *without any idea of approximation*, 5 years (8 years, according to informal remarks of Rivlin,) before Coleman and Noll found that, when the stress is expanded in a retardation parameter, materials with a certain type of fading memory possess *partial stress forms* which are identical to the stress forms which characterize certain, highly special fluids of differential type. Moreover, by relegating

[‡]Others are discussed in Section 4.

fluids of differential type to mere, approximate stand-ins for (unspecified) “truer” models, this position of Metzner, White, & Denn automatically undercuts the status of fluids of differential type as models in their own right. Indeed, all by itself this position strikes down (and these are far and away the majority) all those fluids of differential type which do *not* arise as a result of some expansion or another.

What is especially striking in the position staked out by Metzner, White, & Denn is the completeness of their dismissal of fluids of differential type in general and of fluids of second grade in particular. Because, we suspect, of a misunderstanding of the significance of (1.4), because of their finding a “paradox” where there is none in the work of Coleman, Duffin, & Mizel, because the fluids examined up through 1965 all had $N_1(\kappa) > 0$ over the range of shear rates investigated, and, as we will discuss more fully in Section 4, because of a too hasty appeal to a fairly imprecise concept of approximation, Metzner, White, & Denn appear to us to have been maneuvered into a stance that has them denying* even the *possibility* of real materials which might be modelled by fluids of grade 2 or by more general fluids of differential type. Unfortunately, this stance is by no means unique to these authors: as we shall see, it has been the peculiar fate of these models to be whipsawed between experimental facts of frequently dubious relevancy and theoretical results of overblown and/or misstated significance. The question here is not whether materials exist somewhere in the universe that are well modelled by, say, fluids of second grade; nor is the question one of whether a clever enough chemist could make such a material. The key issues here are, in fact, not existential at all. They are conceptual. What, we suggest, ought to be rethought are the usual *reasons* adduced for the dismissal of fluids of differential type. Are those reasons, supposedly rooted in both theory and experiment, as logically compelling as is commonly thought? Do they lead to so complete a dismissal as folklore would have it?

The second grade fluid model has had as significant an interplay with experiment as it has had with matters of concept and theory. Indeed, not too long after Markovitz & Brown (1963) had found N_1 and N_2 to both be positive, Broadbent, Kaye, Lodge, & Vale (1968) suggested that there was a systematic error in the data reduction schemes of most experimenters of that time owing to the practice of measuring fluid pressure by means of pressure-hole taps. The reality of this so-called pressure-hole error is now commonly accepted, and ways to account for and/or quantify it have been much studied. Two early studies, Tanner & Pipkin (1969) and Kearsley (1970), both analyzed the problem by assuming the fluid to be of second grade. As a result of these and other analyses that involve even milder constitutive assumptions, the

*With the exception of (what must appear to them to be the singular case of) the fluid of Navier and Stokes.

second normal stress difference N_2 for most polymeric fluids is now thought to be *negative* with $|N_2| \approx \frac{1}{10}|N_1|$. (Indeed, Tanner (1970) studied a polyisobutylene-cetane solution and found that N_1 was positive but, in contrast to the conclusions of Markovitz & Brown, N_2 was negative.)[†]

Now, assuming that a solution of polyisobutylene and cetane is a simple fluid, a casual glance at $(1.5)_1$ might suggest that the positivity Tanner found for N_1 still ensures that the modulus Φ_1 of $(1.5)_{1,2}$ is negative[‡], in complete accord with the theoretical result (1.4) of Coleman & Markovitz since for their materials $\Phi_1 = \alpha_1$. But here we come to something strange: While Markovitz & Brown took care to mention that their data of 1963 was consistent with the quadratic dependence of $N_1(\cdot)$ and $N_2(\cdot)$ on κ which is demanded by $(1.5)_{1,2}$, *the 1970 data of Tanner does not support such a dependence*. Rather, the data of Tanner suggests that for low shear rates both $N_1(\kappa)$ and $N_2(\kappa)$ go like $|\kappa|^{1.7}$ for a 6.8% polyisobutylene-cetane solution and like $|\kappa|^{.86}$ and $|\kappa|^{1.1}$, respectively, for a 1% polyethylene-oxide water solution. Moreover, while there are indeed materials that appear to evince a quadratic dependence of $N_1(\cdot)$ and $N_2(\cdot)$ on κ at low shear rates, *Tanner's findings are not in any way anomalous*. Indeed, an extremely large number of experimental studies have produced data that suggests that at low shear rates $N_1(\kappa)$, say, goes like $|\kappa|^m$, $m < 2$ (and, frequently, $m < 1$). A presentation of many of these experimental findings will be found in Dunn, Fosdick, & Rajagopal (1992) where a compelling case is thus made for the non-quadratic variation of $N_1(\cdot)$ and $N_2(\cdot)$ with κ for many polymeric fluids at low shear rates. Here let it suffice to note that, for example, Meister & Biggs (1969) present data that shows that $m = .9$ for a .50% hydroxyethyl cellulose solution and for both a .25% and a .50% polyacrylamide solution; Pritchard (1971) records data that gives $m \leq 1$ for polyisobutene solution; Miller & Christiansen (1972) studied both a 1.49% polyacrylamide water-glycerine solution and a 3.54% polyethylene oxide in water solution, for the first their data gives $m = .4$, for the second their data gives $m = .9$. The data of Chen & Bogue (1972) shows that at low shear rates $m = 1.2$ for low density polyethylene, the same power that data of Walters (1975, page 116) yields for a 1% Oppanol B100 in decalin solution. Finally, mention might be made of the work of Joseph, Beavers, Cers, Dewald, Hoger, & Than (1984) who present data for a 6.0% Vistanex L-100 in decalin solution and for a 6.5% Vistanex L-140 in decalin solution. For both solutions their data yields $m = .66$.

[†]Following a suggestion by Wineman & Pipkin (1966), Tanner utilized in his study a novel experimental method which allowed him to get at the sign (even value) of N_2 independently of any pressure-hole taps.

[‡]Indeed, it is frequently argued in exactly this way that, for example, the 1971 data of Pritchard showing a positive N_1 "proves" $\Phi_1 < 0$. This line of reasoning, sometimes explicit, sometimes only tacit, seems to be quite common.

According to experiment then, at the lowest shear rates to which they were subjected, the above materials failed to evince the quadratic dependence of $N_1(\cdot)$ on κ which is required by (1.5)₁. Or, put differently: even if $N_1(\cdot)$ has a subdomain over which it varies quadratically, the experiments listed above have failed to access it. As a consequence, these experiments stand mute when it comes to evaluating the modulus Φ_1 of (1.5)_{1,2}.[§] Not only is the easy step $N_1 > 0 \Rightarrow \Phi_1 < 0$ no longer secure, one cannot even conclude that Φ_1 exists, and, even if it exists, the experimental data is consistent with $\Phi_1 < 0$, with $\Phi_1 > 0$, with $\Phi_1 = 0$, and with $\Phi_1 = +\infty$ or $-\infty$.

Much of the experimental data on $N_1(\cdot)$ says absolutely nothing at all then about Φ_1 . Indeed, when $N_1(\kappa)$ is found to go like $|\kappa|^m$, $m < 2$, for low shear rates, a healthy regard for experiment would, it seems to us, cause one to question not only the applicability of the grade 2 fluid model (1.1) with its constant modulus α_1 but also the utility of theorems like the retardation theorem which, in its present form, fails to account for the asymptotic behavior suggested by the data. Of course, like asymptotic formulae generally, the retardation theorem of Coleman & Noll does not tell us how small their retardation parameter needs to be for any given term in their expansion scheme to be the dominant one. One could therefore always claim that, no matter what the data says about the form of $N_1(\cdot)$, for still lower (yet unattained) shear rates, $N_1(\kappa)$ would be indeed quadratic-like in κ .[¶] This position, however, seems rather unsatisfying: Quite apart from the act of faith it requires, in what way is

[§]The small sampling of the experimental literature we have given here is not complete or even totally consistent, *i.e.*, we have not listed every fluid studied in each of these reports, nor do all of the reports give data over the same range of shear rates. Thus, a low shear rate for the data of Walters is 100 s^{-1} , while for some of the other workers it is $.1 \text{ s}^{-1}$. Neither point, of course, bears on our observation that for many (but not all) materials, over the range of shear rates studied, the given data is *not* quadratic in κ and so is silent as to the value (existence) of Φ_1 .

[¶]Here it is perhaps useful to draw attention to the somewhat odd remark by Pipkin & Tanner (1972) that "it is difficult to attain shear rates so low that the second order approximation has any relevance". Besides containing a tacit rejection of (1.1) as an exact model, it is not clear what to make of this comment. Again putting aside for later a discussion of whether (1.1) is a "second order approximation" to anything, we must note that the authors do not explain how they know what they claim to know nor do they indicate what shear rate would be low enough for the "second order approximation" to be "relevant". The matter is potentially of some importance since, as we have already indicated, techniques of data reduction (*e.g.*, the pressure hole correction) frequently are based on the second grade fluid model. What is the validity of such techniques if the material is not a second grade fluid, or if a second grade fluid model is not "relevant" to the material's observed response? If, for example, $N_1(\kappa)$ is like $|\kappa|^{.66}$ as κ goes to zero? Additionally, if the experimenter has not achieved shear rates low enough for the "second order approximation" to be "relevant", are we to conclude that nothing is known for sure on the value of α_1 , even for those fluids that do appear to have $N_1(\kappa)$ going like κ^2 at low shear rates? Given a value for Φ_1 , do we have any assurance that α_1 in the "second order approximation" even exists?

an expansion scheme useful if it fails to give forms that match data over the experimental regimes of interest and/or attainability? A more interesting idea might be to study material models, of differential type or not, that allow $N_1(\kappa)$ to go like $|\kappa|^m$, m any positive number. There is, of course, good precedent for this in the generalized Newtonian power law models where $\tau(\kappa)$ of (1.5)₃ is of the form $\mu_0 \text{sgn}(\kappa)|\kappa|^n$ with n usually less than one.^{||} Such models, stripped as their response functions will be of the usual easy assumptions of differentiability, will almost surely be a bit subtle. Corresponding to their potentially non-standard ordering of effects, they may be expected to give rise to both more intricate retardation theorems and more involuted thermomechanics. A preliminary study of such matters will be found in Dunn, Fosdick, & Rajagopal (1992). In the present work we confine ourselves to the more familiar fluids of differential type of which (1.1) is paradigmatic.

We have seen above that the broad sweep of the original (circa 1964) arguments for α_1 in (1.1) being negative cannot be maintained. This is true whether (1.1) is viewed as the result of an expansion arising in a process of flow retardation or as a model in its own right: First, with regards to the structure of (1.1) when it occurs as the result of a retardation expansion, we have discussed how the result (1.4) of Coleman & Markovitz fails to give the most general form for α_1 across the class of simple fluids; consequently, it *cannot* motivate a sign for α_1 across that class. Second, (1.4) and the special concepts of fading memory that underpin (1.4) are not even applicable to (1.1) when (1.1) is taken as an exact model in its own right. Third, the catastrophic instability results of Ting and of Coleman, Duffin, & Mizel all cut against $\alpha_1 < 0$ when (1.1) is taken as a model in its own right. Fourth, unlike the early study of Markovitz & Brown, later experimental data on many materials* fails to even support the quadratic form for $N_1(\cdot)$ required by (1.5)₁ if the form (1.1) is to hold merely asymptotically. This data thus says nothing about α_1 (besides calling its very existence into question.) These simple facts have all been available from the early 70's onwards. It seems particularly unfortunate therefore that the belief that arose in the early years that (1.1) *must* hold in the limit of "slow enough" flows and that the number α_1 *must* be negative still flourishes and draws in new acolytes. Accordingly, even though this paper is in the main concerned with matters theoretical, it is perhaps worth remarking that experimental work on non-Newtonian fluids reveals even richer behavior than we have so far noted. Here we have in mind the data of Pritchard (1971) which both shows that the sign of N_2 is a function

^{||}Chi-Sing Man has shown us interesting, nonclassical results for the model (1.1) when μ is replaced with $\mu_0|\mathbf{A}_1|^p$, $p \in (-1, 0)$. Some of his results are to be found in Man (1992). Man, Shields, Kjartanson, & Sun (1985) have used such a model to analyze creep data on polycrystalline ice. Significantly, they found that their data gave $\alpha_1 > 0$. See also Man & Sun (1987).

*Including the same mixture of polyisobutylene and cetane examined by Markovitz & Brown.

of shear rate and also suggests, in defiance of $(1.5)_2$, that $N_2(0^\pm) \neq N_2(0)(=0)$, *i.e.* N_2 exhibits a yield-like behavior. We also have in mind the data of Kiss & Porter (1980), Lem & Han (1983), and Gotsis & Baird (1986) which reveals that in some liquids the sign of N_1 is also a function of shear rate.[†] By $(1.6)_{1,2}$, such materials are clearly too complex to be adequately described in all flows by a model like (1.1) with its constant moduli μ , α_1 , and α_2 . At most (1.1) might be useful in describing such materials over *certain ranges* of flow behavior, and then, if the flow is unsteady or if the material meets the thermodynamic postulates we lay down here, only if (see (1.7) below) $\alpha_1 \geq 0$, $\alpha_1 + \alpha_2 = 0$, and $\mu \geq 0$. More generally, of course, response as complex as that evinced by the fluids employed in the above studies suggests the desirability of having a good understanding of materials of differential type when the coefficients in the stress form (like μ , α_1 , and α_2 in (1.1)) are permitted to be *functions* of the motion. Some analysis of such fluids, of course no longer of grade 2, will be found in the 1982 and 1990 work of Dunn.

Now, Ting and Coleman, Duffin, & Mizel limited their analyses to shearing type motions. Indeed, Coleman, Duffin, & Mizel just studied time-dependent, simple shearing flows, while Ting, in addition to these motions, also analyzed time-dependent, Poiseuille flow down a

[†]The experiments of Kiss & Porter and of Gotsis & Baird were on liquid crystals while those of Lem & Han were on suspensions. Whether one believes that for certain purposes such materials can be modelled as isotropic or not, it is important to keep in mind that, as we have already remarked, no known physical principle seems to much restrict the normal stress differences, $N_1(\cdot)$ and $N_2(\cdot)$ in general materials. Indeed, even the rather uncomplicated fluids of differential type studied by Dunn (1982) allow for $N_1(\cdot)$ and $N_2(\cdot)$ that oscillate in sign, and, more recently, Dunn (1990) has exhibited a thermodynamically consistent fluid model of differential type which is capable of accommodating almost totally arbitrary normal stress data. It thus appears to be a bit problematic to unambiguously attribute a structural property of, say, $N_1(\cdot)$ to the evolution or alteration of some one or another imputed microstructure. Is the microstructure the cause, the effect, or just a neutral companion of the more gross, isotropic phenomenology? Moreover, while it might seem quixotic to initially ignore microstructure and attempt to model such materials as isotropic, there is much precedent for doing exactly this. Indeed, as Jeffrey & Acrivos (1976) remark: "When we deal with suspensions, it is not always necessary to acknowledge that they are mixtures of particles and fluid; instead, it is often possible to regard them as homogeneous fluids and to ascribe to them certain effective fluid properties." Similarly, homogeneous, isotropic models are often employed in studying problems involving non-homogeneous, anisotropic materials. Illustrative examples of these points of view are not only found in Einstein's work on dilute suspensions of rigid spheres and in studies of polycrystalline metals, they also abound in the fields of food rheology and slurry flows. With regards to polymeric liquid crystals, while optically these materials are anisotropic, it may well be that so fine and/or random is their microstructure that, in many flow situations, their mechanical response might well be modelled as isotropic. It seems hard to be dogmatic here. Rather, the success or failure of such approaches to material modelling will, no doubt, be decided by the intended application(s) of the theory and the range(s) of different physical response elicited and studied by the experimenter.

circular pipe. General, three dimensional motions were not addressed by these authors, nor was the influence or value of the constant α_2 appearing in (1.1), since it dropped out of their analyses. In 1974 Dunn & Fosdick studied a general class of three dimensional flows of a fluid of grade 2 in a bounded domain and showed that if $\alpha_1 < 0$ and if $\alpha_1 + \alpha_2 = 0$ (see (1.7)₃ below), then such flows were, in general, unstable. Later, Fosdick & Rajagopal (1979) extended the results of Dunn & Fosdick and showed that a large class of three dimensional flows are unstable when $\alpha_1 < 0$, whatever the value of $\alpha_1 + \alpha_2$.^{*} Recently, Joseph (1981) studied those special fluids of differential type which are of *grade* $n^\dagger$, $n \geq 2$. He showed that, for a certain subclass of grade n fluids, the rest state is always unstable in the sense of linear stability theory. To describe the special grade n fluids he analyzed, let ϕ_n and ϕ_{n-1} denote the (constant) coefficients of $\mathbf{A}_n$ and $\mathbf{A}_{n-1}$, respectively, in the constitutive equation for the stress of a grade n fluid. Then Joseph's instability result holds for each of those fluids of grade n for which the ratio ϕ_{n-1}/ϕ_n is defined and negative. Since the viscosity μ must be non-negative, Joseph's result immediately implies that *if* $\alpha_1 < 0$ then the rest state of the fluid of grade 2 is unstable, thus echoing for grade 2 fluids the results of Ting (1963), of Coleman, Duffin, & Mizel (1965), of Dunn & Fosdick (1974), and of Fosdick & Rajagopal (1979). Joseph's result, of course, goes much further than the work of these authors inasmuch as it suggests as unreasonable a fairly large, if special, class of materials of differential type.

While the mathematical end result of Joseph's work, which he expresses in a formal theorem, is correct[†], we are not able to say as much about the *conclusions* he seems to draw from it. It seems worthwhile discussing Joseph's paper at some length both because it is relatively current (indeed, key portions of it are repeated verbatim in his 1990 textbook) and because he offers much of the same — we think flawed — argumentation that has come to surround the grade n fluid model. While it is not our intention to indulge in pettifoggery, it does seem to us that there are matters of concept, having to do with both experimental

^{*}In 1990 Galdi, Padula, & Rajagopal studied the stability of flows of second grade fluids in unbounded domains. Provided only that both α_1 and μ are positive, they showed that the rest state is always *conditionally stable*, *i.e.*, the rest state is always stable to small enough disturbances. This result also turns out to be true for flows in bounded domains. Further, Galdi, Padula, & Rajagopal also showed that in an unbounded domain, irrespective of the value of $\alpha_1 + \alpha_2$, the rest state is unstable if $\alpha_1 < 0$ and $\mu > 0$. We emphasize that the work on instability by Galdi, Padula, & Rajagopal is different from that of Dunn & Fosdick: Galdi, Padula, & Rajagopal formulate conditions which ensure that for *all* (unbounded) domains there are *some* (small enough) disturbances which blow up, Dunn & Fosdick prove that for $\alpha_1 < 0$, $\alpha_1 + \alpha_2 = 0$, and $\mu > 0$ there are *some* domains in which *all* disturbances blow up.

[†]See our (2.6)_{1,2} for a formal definition.

[‡]Professor P. Galdi (private communication) has pointed out to us that, while the proof given in Joseph (1981) is flawed, the theorem stated there is in fact true.

interpretation and contending ideas of fluidity, that have been consistently ill treated and/or misstated in the lore that has grown up around grade n fluids. For reasons we do not profess to completely understand, the usual, self-correcting mechanisms that help shape scientific investigation seem to have broken down. The result, as we have already suggested, has been all too frequent misreadings of experimental facts and misstatements of the significance of theoretical results. Fluids of differential type have been impugned time and again, as has in some cases (see below) the whole field of thermomechanics. This state of affairs seems to us both regrettable and avoidable, and it is with this in mind that we seek to examine, perhaps a little more closely than is common, the conceptual undergirding of some of our colleagues' positions. Moreover, the stocktaking of ideas that our program has required raises an assortment of issues in the modelling of non-Newtonian fluids that are of independent interest and which are frequently given rather less attention than we think they deserve.

Now, in the very title he selects: *Instability of the Rest State of Fluids of Arbitrary Grade Greater than One*, not: *Instability of the Rest State of **Certain** Fluids of Arbitrary Grade Greater than One* — word and emphasis added by us but needed by Joseph — we see once more the hand of loose overstatement closing over these models. Further, when we turn to Joseph's text for amplification, we meet straight off the assertion that

“I am going to prove that the rest state of fluids of grade n , any $n \neq 1$, is unstable in the spectral sense of linearized theory when the ratio of the coefficients of $\mathbf{A}_n$ and $\mathbf{A}_{n-1}$ in the constitutive equation is negative. Negative ratios, and only negative ratios, are implied by integral expansions of the stress. Moreover, in the only case ($n = 2$) which has been checked in experiments with polymeric liquids, the ratio is negative.”

There are several problems packed into these few lines. To take the least serious first: What exactly is the main point or idea which Joseph wishes his readers to carry away from these three sentences? Is the hypothesis of the first sentence (that the ratio ϕ_{n-1}/ϕ_n is negative) supposed to gain the stamp of “correct” or of “physically sound” from the final two sentences? Both the structure and placement of these two sentences would suggest such an interpretation, and, if this was Joseph's intention, one might understand why he felt it unnecessary, maybe even redundant, to insert the critical qualifier “certain” into his title. However, to begin to grant such an interpretation is to see why it must be denied: For all the fluids typically studied by rheologists, the rest state is an amazingly dull, stable affair; thus, the first sentence, with its assertion of instability, suffices by itself to deny a certification of “correct” or of “physically sound” to its own hypothesis, *i.e.*, the only grade n fluid models that could *possibly* be labelled “physically sound” for unsteady flows near the rest state are,

by Joseph's own results, those that *fail* his hypothesis. Since the first sentence thus argues for ϕ_{n-1}/ϕ_n positive (or undefined), we are left wondering what to make of the second and third sentences. If Joseph merely means to tell us that some polymeric liquids — as we have seen, a rather shorter list than he would seem to think — yield a value of μ^0/Φ_1 which cannot be taken on within the class of “physically sound” grade n fluid models, he adopts a strange way to do so. Moreover, except in those very special fluids of differential type (like the fluid of second grade) where the value of this limit determines the sign of $N_1(\kappa)$ for *all* κ , the significance of such a fact for constitutive modelling is unclear.

This brings us to a more serious problem of Joseph's introductory paragraph: Observe that the full paragraph derives its force and apparent coherency from the seeming strength of the connecting concept “ratio” which is the only idea serving to link the three sentences together. But this coherency is an illusion since, in Joseph's hands, far from being a sturdy and unchanging idea, the concept “ratio” suffers a drastic metamorphosis. With the progression of each sentence, it is used in altered, ever more different, and ever less constitutively immediate ways. These shifts in meaning are so complete that, far from providing mutual reinforcement, the three sentences seem to us to scarcely speak to one another.[§]

[§]What, for example, does the “ratio” mentioned in sentence one have to do with the “ratios” mentioned in sentence two? For $n = 2$, the first sentence refers to the number μ/α_1 determined by the multipliers of $\mathbf{A}_1$ and $\mathbf{A}_2$ in the form (1.1) for the stress in a second grade fluid; the second sentence speaks to a fluid model in which hypotheses of fading memory explicitly forbid the constitutive equation for the stress from registering any sensitivity on general deformation histories to the current values of $\mathbf{A}_1$ and $\mathbf{A}_2$. From this perspective, two more different fluid models would be difficult to imagine. Of course, like all formulae involving real numbers, they each will offer a ratio hunter ample game. The *significance* of two given ratios having the same sign is, however, the critical issue. Unfortunately, it is not an issue that Joseph's allusion to integral expansions adequately addresses. In his confounding of these two different notions of ratio Joseph has fallen into exactly the same trap we described earlier of mistaking (1.4) as guidance for the sign of α_1 in (1.1) when (1.1) is to be taken as a model in its own right. It does no good here to argue, as we will later see Joseph doing, that fluids of grade n “arise” as a result of flow retardation or integral expansion. All this is doubly, even triply, irrelevant: Not only do these particular artifices or procedures fail to constitute *derivations* of material models, even a valid derivation of, say, grade n fluids (of which we know of none) would at best yield only a *range* of values of the material moduli that is appropriate to that derivations premises. It would say nothing about the range of values of those moduli that might be implied by some *other* derivation. Moreover, regardless of how Joseph or anyone else feels they “arise”, by his entering grade n fluids (more exactly, their linearizations) into the (linearized) law of momentum balance, Joseph is effectively treating them as models in their own right, unencumbered by the dross he insists they rise through. This done, mathematics and ideas physical (in Joseph's case, stability considerations) then speak to the appropriate range of values of the moduli in these materials. If we listen closely, we may even hear them speak to the soundness of any particular purported derivation of the model.

In his third sentence, meanings are shifted on us yet again: a real, *i.e.*, actual, polymeric liquid, all by

For a variety of reasons then, we have never been able to completely understand Joseph's first paragraph. If there is a single, coherent message that can be drawn from his three sentences, we have been unable to find it. The implication of his first sentence (and content of his paper), that the grade n fluid model is unrealistic when ϕ_{n-1}/ϕ_n is defined and negative, is true enough, echoes the stability work that went before him, and is echoed in the thermodynamic analysis we present here. So much is fine. We sense, however, that somehow Joseph wants us to carry away a much stronger message, a message that, as his title suggests, *all* fluids of grade n are unrealistic or, at least, unnatural. Although we think the reasons for it lie elsewhere, we are in fact not unsympathetic to a judgment of unnatural (in fact, the higher the grade, perhaps the more unnatural). What is clear, however, is that neither Joseph's mathematics nor his discussion of modelling and physics even *motivates* such a conclusion, let alone *proves* it.

The results of Joseph (1981) were extended by Renardy (1983) to cover those fluids of grade n , $n \geq 5$, for which the ratio of the coefficient of $\mathbf{A}_{n-1}$ to that of $\mathbf{A}_n$, *i.e.*, ϕ_{n-1}/ϕ_n , is a non-zero real number. Due, perhaps, to a certain looseness in his writing and to his unfortunate choice of terminology, Renardy's results, like Joseph's before him, are easily (and, indeed, have been) misconstrued to imply that all fluids of grade n are physically unsound, irrespective of the nature of the coefficients. This is false, as is made clear by Dunn & Fosdick (1974) for $n = 2$ and by Fosdick & Rajagopal (1980) for $n = 3$.[¶]

itself, does not even have a "ratio of the coefficients of $\mathbf{A}_n$ and $\mathbf{A}_{n-1}$ " for an experimenter to find negative or positive. One won't get a "ratio" unless and until one posits a more or less explicit *model* for the material in question. What Joseph is thus almost surely alluding to here is that, in general, experimenters find the first normal stress difference, N_1 , to be positive over the range of shear rates they investigate. But, absent a model, one can draw no conclusions about Joseph's "ratio". One can, of course, try to compute the number $-2 \lim_{\kappa \rightarrow 0} (\kappa \tau / N_1)$ which may (but need not) correspond to $\mu^0 / \Phi_1 (\equiv \phi_1 / \phi_2 ?)$ in the putative forms (1.5)_{1,3} but, as we have seen, the low shear rate data for many fluids appears inconsistent with the existence of Φ_1 . In any case, the existence of the limit alone suggests no obvious model and, hence, no obvious connection between it and either of the "ratios" of Joseph's first two sentences.

[¶]False also is the second sentence of the final paragraph of his Introduction, where Renardy tells us that "no fluid of grade higher than four can have a stable rest state". Now, in fact, Renardy refines this remark two pages later where he states that "it is necessary to call attention to the fact that we assume a 'fluid of grade n ' to be non-degenerate, *i.e.*, the coefficient α_{n-1} of $\mathbf{A}_n$ is not zero. In principle there may be materials for which the higher-order Rivlin-Ericksen tensors enter only in the nonlinear terms ...". Unfortunately, this refinement, which is crucial, probably comes too late to help a casual reader. Additionally, the second sentence of the refinement offers up a somewhat skewed interpretation of Renardy's own result: for $n \geq 5$, Renardy in fact establishes the nice and rather strong result that, "in principle", realistic fluids of grade n — that is, realistic as counted by linear stability analysis — are to be found *only* among the materials he (unfortunately) dubs "degenerate". For fluids of differential type, the analysis we present here extends

In the present paper, we demonstrate that if a fluid of grade n is consistent with thermodynamics, then the conditions required by Renardy (1983) for instability will never be met. Here, by "consistent with thermodynamics", we mean that the material both satisfies a rather familiar dissipation principle and meets the requirement that the specific Helmholtz free energy be a minimum when the material is locally at rest. Such thermodynamic consistency turns out to, in fact, guarantee that the ratio ϕ_{n-1}/ϕ_n of Joseph and Renardy is not even defined for $n > 2$, being necessarily always of the form $0/0$ for $n \geq 4$ and of the form $\phi_2/0$ for $n = 3$. Moreover, contrary to the instability results of Joseph and Renardy, one can in fact prove that certain fluids of grade n , *e.g.*, fluids of grade 2 and 3, under reasonable flow situations, exhibit asymptotic stability whenever they are rendered consistent with the general principles of thermodynamics.

In their work of 1974 Dunn & Fosdick assumed (1.1) to be an exact model and studied the thermodynamics and stability of such a fluid in great detail. They showed that if the model is to be consistent with thermodynamics then it is necessary that

$$\mu \geq 0, \quad \alpha_1 \geq 0, \quad \text{and} \quad \alpha_1 + \alpha_2 = 0. \quad (1.7)_{1,2,3}$$

In direct opposition then to the suggestion of (1.4), Dunn & Fosdick thus found with (1.7)₂ a physical basis for the sign choice that Ting had adopted 11 years earlier. They also showed that if (1.7) is met with the inequalities being strict then the rest state is asymptotically stable, as are indeed all members of a very large class of nonzero flows in bounded domains. Further, as Dunn & Fosdick noted, the condition (1.7)₃ allows (1.1) to be put into an interesting form of some historical interest. Indeed, let $\mathbf{P}$ denote the *Jaumann* or *co-rotational* time rate of a second order tensor $\mathbf{P}$, *i.e.*,

$$\overset{\circ}{\mathbf{P}} \equiv \dot{\mathbf{P}} + \mathbf{P}\mathbf{W} - \mathbf{W}\mathbf{P},$$

where $\mathbf{W}$, the spin tensor, is just the skew part of the velocity gradient $\mathbf{L}$. Then, with (1.7)₃, the constitutive expression (1.1) becomes

$$\mathbf{T} = -p\mathbf{1} + \mu\mathbf{A}_1 + \alpha_1\overset{\circ}{\mathbf{A}}_1,$$

which is exactly the stress form for the *co-rotational Jeffreys material* when the two *natural times* of that model, the *time of relaxation* λ_1 and the *time of retardation* λ_2 , are set equal

Renardy's result and establishes for it a physical basis in the principle of energy dissipation.

to 0 and to α_1/μ , respectively. When $(1.7)_1$ is strict, we thus see that $(1.7)_2$ ensures that the time of retardation α_1/μ is non-negative.^{||}

The study of Dunn & Fosdick was followed by the work of Fosdick & Rajagopal (1980) on fluids of grade 3 where we get the first inkling that the conditions required by Renardy (1983) might be at odds with thermodynamics. The Cauchy stress $\mathbf{T}$ in an incompressible fluid of grade 3 has the constitutive structure:

$$\mathbf{T} = -p\mathbf{1} + \mu\mathbf{A}_1 + \alpha_1\mathbf{A}_2 + \alpha_2\mathbf{A}_1^2 + \beta_1\mathbf{A}_3 + \beta_2[\mathbf{A}_1\mathbf{A}_2 + \mathbf{A}_2\mathbf{A}_1] + \beta_3(\text{tr}\mathbf{A}_1^2)\mathbf{A}_1,$$

where β_1 , β_2 , and β_3 are additional material constants. Fosdick & Rajagopal (1980) show that if the fluid is to be consistent with thermodynamics, it is necessary that

$$\mu \geq 0, \quad \alpha_1 \geq 0, \quad |\alpha_1 + \alpha_2| \leq \sqrt{24\mu\beta_3}, \quad (1.8)_{1,2,3}$$

$$\beta_1 = 0, \quad \beta_2 = 0, \quad \beta_3 \geq 0. \quad (1.8)_{4,5,6}$$

We see at once that the ratio of the coefficient of $\mathbf{A}_2$ to that of $\mathbf{A}_3$ in the form for $\mathbf{T}$, i.e., the “ratio” $\alpha_1/0$, does not satisfy either the hypothesis of Joseph (1981) or the hypothesis of Renardy (1983). Thermodynamics is thus hinting here at the conflict it has with Joseph’s and Renardy’s materials (although Renardy’s instability result is for $n \geq 5$ and here $n = 3$). The study of Dunwoody (1981) on the linearized stability of the rest state of fluids of differential type illustrates this point further. Dunwoody shows that if fluids of grade n are thermodynamically consistent for all retarded flows, then the rest state of the

^{||}The first and third conditions of (1.7) have an interesting history: Before he and Noll formalized the use of the Clausius-Duhem inequality as a restriction on constitutive equations, Coleman (1962) gave an argument to the effect that, while the stress power $\mathbf{T} \cdot \mathbf{L}$ in an incompressible simple fluid may be of any sign in general, it must be *non-negative in all isothermal, substantially stagnant motions*. Three years later, Coleman, Duffin, & Mizel (1965) discussed this matter in an extended footnote at the end of their paper and quoted an unpublished result of Noll to the effect that, when $\mathbf{T}$ meets (1.1), the stress power $\mathbf{T} \cdot \mathbf{L}$ is non-negative in all substantially stagnant motions if and only if both $(1.7)_1$ and $(1.7)_3$ hold. Thus, albeit from a somewhat different and rather more special point of view than that of Dunn & Fosdick (1974), Coleman, Duffin, & Mizel came to the conclusion that “thermodynamics” requires the model (1.1) to meet $(1.7)_1$ and $(1.7)_3$. This conclusion was stated with what now seems surprisingly little force but was immediately followed, first, by the factual observation that $(1.7)_3$ fails for a particular class of materials (*viz.*, the polyisobutylene solutions of Markovitz & Brown) and then by the curious (and completely unsupported) remark that “We doubt that an experimenter will ever find a material obeying” (1.1) “precisely for all values of $\mathbf{A}_1$ and $\mathbf{A}_2$ ”. In spite of such a doubt requiring the rejection of even the Navier-Stokes fluid model, the unstudied fusing of these three remarks across the years seems to us to have had a confusing and unfortunately widespread effect on much subsequent work on the grade 2 fluid model.

fluid is stable to infinitesimal disturbances. Assuming the response of fluids of complexity n to be approximated by fluids of grade n , Dunwoody thus infers that the rest state of thermodynamically consistent complexity n fluids is stable to infinitesimal disturbances. An interesting consequence of Dunwoody's analysis is that the criterion which Renardy (1983) requires for instability can never be attained in any thermodynamically fit fluid of differential type. Equivalently, those fluids of grade n which Renardy shows have an unstable rest state are incompatible with thermodynamics. We can and should turn this around to conclude that, loosely speaking, *thermodynamic incompatibility implies instability*. Far from rendering vacuous works on the thermodynamics of materials of differential type, the results of Joseph (1981) and Renardy (1983) both complement and emphasize the relevance of such investigations. The mathematical content of their linear stability analyses and the thermodynamic results we give here should remind us once again of the frequent and intimate connection between thermodynamics and stability.

There is another common misunderstanding of restrictions like those of $(1.7)_{1,2,3}$ and Fosdick & Rajagopal's $(1.8)_{1-6}$ that ought be mentioned: Although the stability and/or thermodynamic analysis that leads to, say, $(1.7)_{1,2,3}$ is clear and straightforward, one sometimes encounters remarks like:

“Contrary to $(1.7)_{2,3}$, $\alpha_1 > 0$ and $\alpha_1 + \alpha_2 = 0$ are not valid physically. This must mean that thermodynamic arguments are incorrect and that thermodynamic reasoning cannot predict the correct values of α_1 and α_2 .” (Q)

The paradox or impugning of thermodynamics implied by remarks like these is really not too serious and is caused by what at base is a fairly simple misunderstanding. It is interesting, however, that the above sentiments are never, never expressed with the word “thermodynamic” replaced with the word “stability”. Although thermodynamic analyses and stability analyses speak with essentially one voice with regard to $(1.7)_{2,3}$, only thermodynamics, perhaps by some still felt to be mysterious, is ever assaulted with the negation of $(1.7)_{2,3}$ supposedly proven by experiment.

Now the first sentence of the position (Q) notwithstanding, we know from stability (and thermodynamic) considerations that, *when (1.1) is treated as exact* (and is embedded in a thermodynamics like that given here), *only* $\alpha_1 \geq 0$ and $\alpha_1 + \alpha_2 = 0$ are “*valid physically*”. What, we would suggest, that anyone holding to (Q) really had in mind with the slippery phrase[†] “not valid physically” is that, *rather than the α_1 and α_2 of the model form (1.1) for*

[†]Equally slippery can be a phrase like “physically reasonable”. Indeed, in a recent book, Larson (1988)

a grade 2 fluid, it is the numbers Φ_1 and Φ_2 of the normal stress difference forms (1.5)_{1,2} that are, for many liquids, not found from experiment to satisfy $\Phi_1 > 0$ and $\Phi_1 + \Phi_2 = 0$. This is harmless and in no way conflicts with (1.7)_{2,3}. Indeed, put a little better, (Q) might be reformulated as:

“Let Φ_1 and Φ_2 be defined by $2\Phi_1 \equiv -\lim_{\kappa \rightarrow 0} N_1(\kappa)/\kappa^2$ and $2\Phi_1 + \Phi_2 \equiv \lim_{\kappa \rightarrow 0} N_2(\kappa)/\kappa^2$, where κ is the shear rate and $N_1(\kappa)$ and $N_2(\kappa)$ are, respectively, the first and second normal stress differences in a steady, simple shearing flow. Then, while much experimental data on polymeric liquids is inconsistent with the existence of the indicated limits (and so inconsistent with an asymptotic form like (1.1) holding true), and while it is really fairly difficult to achieve slow enough shear rates in polymeric liquids to be completely secure in approximating Φ_1 and Φ_2 , nevertheless, there is no experimental data on polymeric liquids that suggests $\Phi_1 > 0$ and $\Phi_1 + \Phi_2 = 0$.” (Q’)

With the results (1.6)_{1,2} and (1.7)_{2,3} for grade 2 fluids in mind, a person originally holding to (Q) hopefully would then conclude something like: “At least as regards their normal stress differences, many (perhaps all) polymeric liquids will not be well modelled in general motions by fluids of second grade.” Drawn into a speculative frame of mind, he then might muse: “Whatever the constitutive equation for the stress is in, say, a polymeric fluid, it cannot be given by (1.1); *there must be additional terms* whose presence is critical in giving rise to *different* restrictions than those of (1.7)_{2,3} which are imposed on the second grade fluid

states that “One strange result” (of treating the grade 2 fluid model as exact) “is that for physically reasonable values of the coefficients, the second law of thermodynamics can be shown to be violated in certain flow histories³³”. Now Larson’s reference [33] is Dunn & Fosdick (1974), and their position, like ours here, is just the opposite of Larson’s, *i.e.*, as counted by either thermodynamics or stability, *the only physically reasonable values of the coefficients of a second grade fluid are values that meet* (1.7)_{1,2,3}. What Larson no doubt means by “physically reasonable values of the coefficients” in (1.1) is something like “experimentally determined values of the numbers Φ_1 and Φ_2 appearing in (1.5)_{1,2}”. His way of putting it, however, suggests a conflict between physics and the second law (else why “strange” and what else is achieved by juxtaposing “physically reasonable”, “law”, and “violated”?) and so creates the illusion of a paradox. Instead, of course, the real conflict we think Larson has in mind is between experiments on polymeric liquids and the model form (1.1) (a not necessarily “strange” conflict at all since, *a priori*, there is no particular reason why (1.1) should provide a good model of polymeric liquids in general flows; no reason, that is, why experimentally determined values of Φ_1 and Φ_2 should correspond to physically reasonable values of α_1 and α_2 in the model form (1.1)). Unfortunately, the distinction we make here is not one of just pointless hairsplitting: Larson’s way of formulating the matter leads him step by step in his next four sentences, as others have been lead, to insisting that (1.1) should be viewed *only* as a truncated expansion of a more real model, that “to avoid paradoxical results, *one should not regard the equation of the second-order fluid as a constitutive “law” describing some class of fluids...*” (emphasis in the original).

model by considerations of both thermodynamics and stability.”

There is no conflict therefore with “thermodynamic reasoning” on the one hand and experiment and, say, structural theories of liquids on the other hand. If experiment and/or some particular structural theory of liquids leads us to values of Φ_1 and Φ_2 in $(1.5)_{1,2}$ that fail $(1.7)_{2,3}$ when Φ_1 is identified with α_1 and Φ_2 is identified with α_2 then this is fine. It merely means that (1.1) *cannot* be the *complete* expression for the stress: the sample material of the experiment and/or the model material of the structural theory *must* have a stress form that departs in some way from (1.1). Thus, if only in a negative way, thermodynamic and stability analyses of grade 2 fluids are still of value and ought be of interest to our experimenter and our structural theoretician.[§]

The real culprit in the misunderstandings we have just discussed is language, language whose precision has not kept pace with our ability to create ever more sophisticated theories of material response, language insufficiently sharp to at once embrace experiment and at the same time delineate in an evenhanded way between competing theories that may model it. Expressions like “valid physically” and “physically sound” are relatively harmless when we

[§]The reader may think that only the straw men of our imagination could ever confuse or mix up the constants α_1 and α_2 in the second grade model with the numbers Φ_1 and Φ_2 appearing in the differentiability assumptions $(1.5)_{1,2}$. It may be of interest therefore that, while we had others in mind when we originally drew up the position (Q), Müller & Wilmanski (1986) make exactly this mistake. The third sentence of their paper describes the results $(1.7)_{1,2,3}$ of Dunn & Fosdick, the fourth sentence states that Dunn & Fosdick were well aware that $(1.7)_{2,3}$ are contradicted by experiment, and their fifth sentence goes on to state that “experiments indicate that we must have $\alpha_1 < 0$, $\alpha_1 + \alpha_2 \neq 0$.” As we have tried to make clear, not only have Müller & Wilmanski completely misread Dunn & Fosdick’s position, they have also completely mistaken Φ_1 and Φ_2 for α_1 and α_2 . It is just not true that experiments indicate that the α_1 and α_2 of the model form (1.1) must meet $\alpha_1 < 0$, $\alpha_1 + \alpha_2 \neq 0$. When experiments (on polymeric liquids) suggest they exist at all, those experiments do indeed indicate that Φ_1 and Φ_2 of $(1.5)_{1,2}$ meet $\Phi_1 < 0$ and $\Phi_1 + \Phi_2 \neq 0$. But, by itself, this says nothing about the model form (1.1), says nothing about the “right” sign of α_1 in (1.1), and, when put alongside the theoretical analyses leading to $(1.7)_{2,3}$, it says only that the fluid being tested is not well-modelled in general flows by (1.1). While this (mis)use of experimentally determined numbers to first delimit moduli in a theoretical model and then discredit the model is repeated in Müller (1987), Müller & Wilmanski go even further than this and suggest (first sentence after their (6.6)) that “ordinary thermodynamics” is itself discredited because it predicts “the wrong sign for α_1 ”. In spite of this, it seems unlikely that Müller & Wilmanski really mean to assert or imply that the “right” sign for α_1 is the one that leads to the instability results of Coleman, Duffin, & Mizel, of Dunn & Fosdick, and of Joseph. (Both of the above two papers were interested in motivating a treatment of non-Newtonian constitutive models by the methods of Müller’s “extended thermodynamics”. While such a treatment is worthwhile and of interest, we ought not lose sight of the fact that, ensnared as it will be in the iteration schemes of extended thermodynamics, the *exact* form (1.1) is not what is being studied. Thus, Müller & Wilmanski never really deliver on the promise of their first sentence to present “a thermodynamic theory of ... a second order fluid”.)

are talking, either in general or for a specific material, of a shear modulus or a viscosity: the theory underlying each modulus, be it linear elasticity on the one hand or Navier-Stokes fluids on the other, is clear and has no obvious competitors. This is not the case when we move to more complex theories of material behavior. Indeed, as we have seen, in non-Newtonian fluid mechanics, even though the real fluid may be immediate enough, we have an abundance of competing models of fluidity each of whose most significant properties are often far from immediate. Thus, for example, in some of these models the normal stress coefficient Φ_1 of (1.5)₁ will necessarily be negative for the model to make “physical sense”, *i.e.*, to be “physically reasonable” (*e.g.*, the materials of Coleman & Markovitz (1964)), in other models Φ_1 will have to be zero or larger for the model to make “physical sense” (*e.g.*, grade 2 fluids or the more general fluids studied by Dunn (1982)). Here the notions of “physical sense” and “physically reasonable” are based on considerations so general as to have had theoretical structures built up around them (*e.g.*, the web of ideas that make up thermodynamics and/or stability theory). And, as it turns out, the “physically reasonable” values of Φ_1 appropriate to one class of models are not those that are “physically reasonable” for the other class. The actual *measurement* of Φ_1 in (1.5)₁ for a real material can hardly impugn either class of models *qua* models. Such *experimentally found* (as opposed to “physically reasonable”) values[¶] of Φ_1 can, at most, impugn the *applicability* of one class of models for modelling the given, real material. To put it only a little too simply: Experiments carried out on a material well-modelled over a certain range of response by the equations of linear elasticity cannot be used to impugn the theory of Navier-Stokes fluids.^{||}

Now, while they are connected, thermodynamics and stability are, of course, not equivalent. In this regard it is worth noting that, although Joseph’s (Renardy’s) results certainly shows that those grade n fluids with ϕ_{n-1}/ϕ_n defined and negative, $n \geq 2$, (defined and

[¶]For the purpose of making our point, we are being more than a trifle absolute here. We really do not take exception to and there is really nothing wrong with referring to a “physically reasonable value” of, say, Φ_1 for polyisobutene, or for polyethylene, or for *any other actual fluid*. Such a usage, tied as it is to a real material, is completely equivalent to our “experimentally determined”. The trouble comes (or can come) when we lose sight of the fact that such a “physically reasonable” actual value typically brings with it no particular *model* and therefore no warrant that a particular model will make “physical sense” when that value is entered into it.

^{||}For a related discussion concerning models and experiments, see the extended first footnote in the work of Fosdick & Rajagopal (1978) on drag in second grade fluids. Setting aside their too easy to misconstrue remarks about “experimental evidence” supporting (or, by implication, not supporting) “thermodynamic restrictions”, that footnote of 14 years ago expresses well some of the points we have tried to lay out here. Much subsequent misleading writing would have been avoided had the analysis of Fosdick & Rajagopal not been overlooked.

non-zero, $n \geq 5$) are bad models for describing *unsteady* flows near the rest state, it is not *a priori* impossible, from the point of view of stability analysis alone, that those special grade n fluids studied by Joseph and Renardy (more precisely, the special *forms* for the stress they studied) might be used to describe *steady* flows or, perhaps, some more limited class of flow behavior. The rheological community has always been of two minds in this regard, on the one hand being aware of the instability results we have described when α_1 in (1.1) is negative, on the other using the second grade constitutive equation to interpret, even "correct", data that arises from experiments on "slow enough", steady motions that are in some sense close to being viscometric (and that usually leads to $N_1 > 0$, requiring by (1.5)₁ that $\alpha_1 < 0$). This practice is not, in any obvious way, wrong: it is quite possible that, when they are examined on, say, steady motions alone, material models having reasonable stability properties might be indistinguishable from — that is, might meet the same reduced** field equations and boundary conditions as — a second grade fluid with $\alpha_1 < 0$ (more generally, a grade n fluid with $\phi_{n-1}/\phi_n < 0$). For an appropriately restricted class of processes (*viz.* all those that give rise to steady motions), this suggests a possible, though physically rather special and restricted, role for grade n fluid models, irrespective of whether the ratio ϕ_{n-1}/ϕ_n is positive, negative, zero, or undefined. Stability analyses, linear or not, clearly cannot address the fitness of such restricted material modelling. Here, however, our thermodynamic analysis provides some additional insight: since the results we give remain true even if the class of flows be restricted to just those that are steady, *the constitutive restrictions we find here hold for any material whatsoever as long as on steady motions its stress $\mathbf{T}$ and its stored energy w (i) are given by our (2.1) (our (2.6)_{1,2}) and our (2.2), respectively, and (ii) satisfy the dissipation inequality (2.3).* Contrariwise, even if the stress in steady motions is given by (2.1) or, more specially, by (2.6)_{1,2}, *our results can all be rendered inapplicable, if either the stored energy fails to be of the form (2.2) or the dissipation processes in the material fail to be governed by (2.3).* This last, for example, can occur both in theories with internal variables and in theories that require non-standard fluxes of either energy or entropy.

Finally, while it is clear from our remarks above that we find little merit in the usual reasons given for dismissing grade n fluid models, we should say that some support for a *selective* dismissal when n is 3 or larger can in fact be drawn from the very results we present here: Our results, along with many others we have referenced, have the character that grade n fluids, $n \geq 3$, suffer significant constitutive collapse due to the requirements of thermodynamics. Equivalently, the principles of thermomechanics themselves suffice to

**By *reduced* we here mean the forms taken by the field equations and boundary conditions *after* the material's constitutive equation has been entered into them.

dismiss all but very special grade n fluid models. Indeed, we conclude Section 3 by quoting a result of Dunn & Rajagopal (1992) which gives one more indication of the wide sweep of this dismissal. Roughly speaking, it states that an attempted too special dependence of $\mathbf{T}$ on $\mathbf{L}, \dot{\mathbf{L}}, \ddot{\mathbf{L}}, \dots, \overset{n}{\mathbf{L}}$ (*i.e.*, any grade 5 fluid) must collapse to just a polynomial dependence on $\mathbf{L}$ and $\dot{\mathbf{L}}$ (*i.e.*, must collapse to a special complexity 2 fluid). One speculation for this lack of robustness of grade n fluid models will be found at the conclusion of the next section.

2 CONSTITUTIVE SETTING

Let T , T° , and T_s denote, respectively, the set of tensors, traceless tensors, and symmetric tensors over a three dimensional inner product space. Let $T_s^\circ = T^\circ \cap T_s$. In an *incompressible homogeneous fluid of differential type*, the Cauchy stress tensor $\mathbf{T} \in T_s$ is given by a constitutive equation of the form

$$\mathbf{T} = -p\mathbf{1} + \hat{\mathbf{T}}\left(\mathbf{L}, \dot{\mathbf{L}}, \ddot{\mathbf{L}}, \dots, \overset{n-1}{\mathbf{L}}\right) , \quad (2.1)$$

where p is a constitutively indeterminate pressure due to the constraint of incompressibility, where $\dot{\mathbf{L}}, \ddot{\mathbf{L}}, \dots, \overset{i}{\mathbf{L}}$ denote, respectively, the first, the second, and the i -th material time derivative of the spatial velocity gradient $\mathbf{L}$, and where the thermal independent variables entering into $\hat{\mathbf{T}}(\cdot)$ have been suppressed. Since the material is incompressible, we see that each $\overset{i}{\mathbf{L}} \in T^\circ$.

Again suppressing thermal independent variables, such fluids conventionally have associated with them a stored energy function $\hat{w}(\cdot)$ of the form

$$w = \hat{w}\left(\mathbf{L}, \dot{\mathbf{L}}, \ddot{\mathbf{L}}, \dots, \overset{m-1}{\mathbf{L}}\right) , \quad (2.2)$$

such that in *all* motions of the body

$$\rho \dot{w} \leq \mathbf{T} \cdot \mathbf{L} , \quad (2.3)$$

where ρ is the uniform constant density of the fluid. We remark that m in (2.2) need not equal n in (2.1) and that, for simplicity of stating results, we will usually have in mind the situation $n \geq 1, m \geq 1$. Easy, limiting forms of our theorems hold if n and/or m are taken to be zero, *i.e.*, if $\hat{\mathbf{T}}(\cdot)$ and/or $\hat{w}(\cdot)$ really have no sensitivity to deformation rates. We also observe that (2.3) is just the assertion that the growth of ρw is bounded above by the stress

power $\mathbf{T} \cdot \mathbf{L}$, *i.e.*, in a purely mechanical process, the rate of growth of the stored energy ρw can never exceed the rate of working of the stresses. We call (2.3) the *dissipation inequality*; it, of course, never restricts either the pressure p or any component of $\hat{\mathbf{T}}(\cdot)$ parallel to $\mathbf{1}$ since the velocity gradient $\mathbf{L}$ is traceless.

For us, we envision (2.3) as arising out of thermodynamics in the form of the Clausius-Duhem inequality as outlined by Dunn & Fosdick (1974) and Fosdick & Rajagopal (1980). In that case, if the suppressed independent thermal variables are temperature and its spatial gradient, then w is the Helmholtz free energy, while, on the other hand, if the suppressed independent thermal variables are entropy and temperature gradient, then w is the specific internal energy. The analysis we present here is independent of such interpretations and whatever detailed considerations induce (2.3). Indeed, some of our analysis is of interest even in the case when $\hat{w}(\cdot) \equiv 0$, *i.e.*, even when (2.3) just reduces to the sometimes-asserted positivity of the stress power.

Due to frame indifference (see, for example, Truesdell & Noll (1965)), the response functions $\hat{\mathbf{T}}(\cdot)$ and $\hat{w}(\cdot)$ cannot depend on $\mathbf{L}$ and its rates in an arbitrary manner. Rather, there must be isotropic functions $\tilde{\mathbf{T}}(\cdot)$ and $\tilde{w}(\cdot)$ such that

$$\begin{aligned}\mathbf{T} &= -p\mathbf{1} + \hat{\mathbf{T}}\left(\mathbf{L}, \dot{\mathbf{L}}, \ddot{\mathbf{L}}, \dots, \overset{n-1}{\mathbf{L}}\right), \\ &= -p\mathbf{1} + \tilde{\mathbf{T}}(\mathbf{A}_1, \mathbf{A}_2, \dots, \mathbf{A}_n),\end{aligned}\tag{2.4}$$

and

$$\begin{aligned}w &= \hat{w}\left(\mathbf{L}, \dot{\mathbf{L}}, \ddot{\mathbf{L}}, \dots, \overset{m-1}{\mathbf{L}}\right), \\ &= \tilde{w}(\mathbf{A}_1, \mathbf{A}_2, \dots, \mathbf{A}_m).\end{aligned}\tag{2.5}$$

By (1.2) and (1.3), the $\mathbf{A}_i$ are in T_s but, except for $\mathbf{A}_1$, they are not in general in T° .^{*} For brevity we will refer to a material model satisfying the three conditions (2.3), (2.4), and (2.5) as a fluid of *differential type of class* (m, n) , or, even more briefly, a fluid of *class* (m, n) . When $m \leq n$, we may extend the terminology of Truesdell & Noll (1965) and call n the *complexity* of the material.

Grade n fluids (more properly here, *grade* (m, n) *fluids*) are special differential type fluids

^{*}In fact, due to the recursion formula (1.3), it can be shown that in any volume preserving motion the trace of any $\mathbf{A}_i$ can be expressed as a linear combination of traces of products formed from $\mathbf{A}_1, \mathbf{A}_2, \dots, \mathbf{A}_{i-1}$. This fact makes delineation of that hyperplane of T_s occupied by a given $\mathbf{A}_i$ somewhat awkward.

of class (m, n) . For them the response function $\hat{\mathbf{T}}(\cdot)$ is of the very special form

$$\hat{\mathbf{T}}(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-1}{\mathbf{L}}) = \sum_{i=1}^n \hat{\mathbf{S}}_i(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{i-1}{\mathbf{L}}) \quad , \quad (2.6)_1$$

where each of the n symmetric-valued *partial stress* functions $\hat{\mathbf{S}}_i(\cdot)$ is defined on the Cartesian product of T^o with itself i times, is i times smooth at the origin, and is *homogeneous of degree i modulo a time scaling*, i.e., for all $\tau \in (0, 1]$ (for every lengthening of the time scale by a factor $\tau^{-1} \in [1, \infty)$) and for each $i = 1, 2, \dots, n$:

$$\hat{\mathbf{S}}_i(\tau \mathbf{L}, \tau^2 \dot{\mathbf{L}}, \dots, \tau^i \overset{i-1}{\mathbf{L}}) = \tau^i \hat{\mathbf{S}}_i(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{i-1}{\mathbf{L}}) \quad . \quad (2.6)_2$$

By differentiating (2.6)₂ i times with respect to τ and evaluating the result at $\tau = 0$, it is clear that each partial stress function $\hat{\mathbf{S}}_i(\cdot)$ is a rather special *polynomial* form in the i tensors $\mathbf{L}, \dot{\mathbf{L}}, \dots$, and $\overset{i-1}{\mathbf{L}}$. Thus, while we shall not need to exploit it in full, the stress response of grade (m, n) fluids is structured in a fairly simple and explicit way.

The conditions (2.6)_{1,2} make clear that grade (m, n) fluids stand out among fluids of differential type by embodying a strict partitioning of the stress in terms of its sensitivity to the time rates of $\mathbf{L}$. Indeed, as a formal hierarchy of models, grade (m, n) fluids were first introduced by Coleman & Noll (1960) in the context of partitioning the stress in a simple fluid with a particular type of “fading memory” according to its response to deformation histories that are more and more “slowed down” or *retarded* by a factor of τ . However conceptually appealing such an *expansion* scheme is for any *fixed* deformation history, it seems to us rather unlikely that, in general motions, any real material would meet the stringent partitioning of effects embodied in (2.6)_{1,2} when n is fixed and finite. It thus seems to us that this rigid partitioning, and not their misunderstood thermodynamics and stability properties, is the biggest reason to doubt the applicability of grade n fluid models.[†]

[†]If this suspicion — and that is all it is — is correct, then, as one goes to higher and higher values of n in (2.6)_{1,2}, the progressive *exact* models so defined ought to be *poorer* and *poorer* embodiments of real material behavior. This, of course, is just the opposite of what one would want in an approximation procedure. It would be interesting if these vague ideas could be rigorously substantiated. While we have not looked into this, the theorem described at the end of the next section, by Dunn & Rajagopal on grade 5 fluids, might be taken as offering some encouragement for such a program. However, it is our sense that the question of the applicability of grade n fluids has less to do with their thermodynamics than it does with their purely mechanical response, i.e., with the qualitative features of the solution fields coming out of momentum balance alone. Thus, an extension of the analyses of Joseph and of Renardy might be quite fruitful here.

3 THERMODYNAMIC RESTRICTIONS

We shall henceforth assume the response functions $\hat{w}(\cdot)$ and $\hat{T}(\cdot)$ are smooth enough to make meaningful all derivatives of them which appear in our analysis. Subscripts on $\hat{w}(\cdot)$ and $\hat{T}(\cdot)$ will be used to denote partial derivatives. Our first result is very simple; it shows that the dissipation inequality (2.3) significantly restricts the appearance of higher strain rates in the stored energy in terms of those rates that appear in the stress.

Theorem 1. *For every fluid of class (m, n) ,*

$$m < n \quad ,$$

i.e., the highest rate $\overset{m-1}{\mathbf{L}}$ that can actually appear in the stored energy $\hat{w}(\cdot)$ must be of lower order than the highest rate $\overset{n-1}{\mathbf{L}}$ that appears in the stress $\hat{T}(\cdot)$.

Proof: Since $\hat{w}(\cdot)$ is smooth, we have that

$$\dot{w} = \hat{w}_{\mathbf{L}} \cdot \dot{\mathbf{L}} + \hat{w}_{\dot{\mathbf{L}}} \cdot \ddot{\mathbf{L}} + \cdots + \hat{w}_{\overset{m}{\mathbf{L}}} \cdot \overset{m}{\mathbf{L}} \quad .$$

Therefore, (2.3) takes the form

$$\rho \{ \hat{w}_{\mathbf{L}} \cdot \dot{\mathbf{L}} + \hat{w}_{\dot{\mathbf{L}}} \cdot \ddot{\mathbf{L}} + \cdots + \hat{w}_{\overset{m}{\mathbf{L}}} \cdot \overset{m}{\mathbf{L}} \} \leq \hat{\mathbf{T}} \cdot \mathbf{L} \quad , \quad (3.1)$$

and this must hold for every $\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{m}{\mathbf{L}}, \dots, \overset{n-1}{\mathbf{L}}$ in T^o .

Suppose now that we attempted to take $m \geq n$. In this case $\overset{m}{\mathbf{L}}$ appears linearly in (3.1), and (3.1) is thus easily violated unless

$$\hat{w}_{\overset{m}{\mathbf{L}}} \left(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{m-1}{\mathbf{L}} \right) \equiv 0 \quad .$$

Equivalently, $\hat{w}(\cdot)$ must be independent of its last argument:

$$w = \hat{w} \left(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{m-2}{\mathbf{L}} \right) \quad .$$

This implies in turn that (3.1) is actually of the form

$$\rho \{ \hat{w}_{\mathbf{L}} \cdot \dot{\mathbf{L}} + \hat{w}_{\dot{\mathbf{L}}} \cdot \ddot{\mathbf{L}} + \cdots + \hat{w}_{\overset{m-1}{\mathbf{L}}} \cdot \overset{m-1}{\mathbf{L}} \} \leq \hat{\mathbf{T}} \cdot \mathbf{L} \quad . \quad (3.2)$$

Now, if we had originally attempted to take $m = n$ then the proof is complete: $\overset{n-1}{\mathbf{L}}$ cannot really appear in $\hat{w}(\cdot)$. If, however, we had taken $m > n$, then obviously $m - 1 > n - 1$, and so now $\overset{m-1}{\mathbf{L}}$ occurs linearly in (3.2) and we can conclude $\hat{w}_{\overset{m-2}{\mathbf{L}}} \equiv 0$, i.e.,

$$w = \hat{w} \left(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{m-3}{\mathbf{L}} \right) .$$

We can clearly continue this process until we arrive at

$$w = \hat{w} \left(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-2}{\mathbf{L}} \right) ,$$

which completes the proof. ■

Our next result shows that the energy function $\hat{w}(\cdot)$ must have a stationary value at $(0, 0, \dots, 0)$, i.e., all its partial derivatives must vanish there. However, from the dissipation inequality (2.3), we cannot determine the nature of this stationary value; it could be a minimum, a maximum, or a saddle point.

Theorem 2. *The energy function $\hat{w}(\cdot)$ of any fluid of class (m, n) must satisfy*

$$\begin{aligned} \hat{w}_{\overset{i}{\mathbf{L}}} (0, 0, \dots, 0) &= 0, \quad i = 0, \dots, m-2, \\ \hat{w}_{\overset{m-1}{\mathbf{L}}} \left(0, \dot{\mathbf{L}}, \ddot{\mathbf{L}}, \dots, \overset{m-1}{\mathbf{L}} \right) &= 0 \quad \forall \quad \dot{\mathbf{L}}, \ddot{\mathbf{L}}, \dots, \overset{m-1}{\mathbf{L}} \in T^o . \end{aligned}$$

This theorem thus establishes a bit more than just the stationary character of $\hat{w}(\cdot)$ at the point $(0, 0, \dots, 0)$: the asserted vanishing of the function $\hat{w}_{\overset{m-1}{\mathbf{L}}}(0, \cdot, \cdot, \dots, \cdot)$ is a good deal stronger than just $\hat{w}_{\overset{m-1}{\mathbf{L}}}(0, 0, 0, \dots, 0) = 0$. This stronger condition will be important below.

Proof: As we have seen, (3.1) must hold for all $\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{m}{\mathbf{L}}, \dots, \overset{n-1}{\mathbf{L}}$, where $m < n$. In general, of course, this will mean that (3.1) will involve $\overset{m}{\mathbf{L}}$ in a non-linear way. However, if we take $\mathbf{L} = 0$ in (3.1), then the resulting inequality

$$\hat{w}_{\mathbf{L}} \left(0, \dot{\mathbf{L}}, \dots, \overset{m-1}{\mathbf{L}} \right) \cdot \dot{\mathbf{L}} + \hat{w}_{\dot{\mathbf{L}}} \left(0, \dot{\mathbf{L}}, \dots, \overset{m-1}{\mathbf{L}} \right) \cdot \ddot{\mathbf{L}} + \dots + \hat{w}_{\overset{m-1}{\mathbf{L}}} \left(0, \dot{\mathbf{L}}, \dots, \overset{m-1}{\mathbf{L}} \right) \cdot \overset{m}{\mathbf{L}} \leq 0 \quad (3.3)$$

is certainly linear in $\overset{m}{\mathbf{L}}$, and thus may be violated unless $\hat{w}_{\overset{m-1}{\mathbf{L}}} \left(0, \dot{\mathbf{L}}, \dots, \overset{m-1}{\mathbf{L}} \right) \equiv 0$. This establishes the second half of the theorem.

In the now reduced form of (3.3) replace $\dot{\mathbf{L}}$ with $\gamma \dot{\mathbf{L}}$, $\ddot{\mathbf{L}}$ with $\gamma \ddot{\mathbf{L}}$, $\dots$, $\overset{m-1}{\mathbf{L}}$ with $\gamma \overset{m-1}{\mathbf{L}}$, $\gamma > 0$, divide by γ , and let $\gamma \searrow 0$. We thus find that

$$\hat{w}_{\mathbf{L}}(0,0,\dots,0) \cdot \dot{\mathbf{L}} + \hat{w}_{\ddot{\mathbf{L}}} (0,0,\dots,0) \cdot \ddot{\mathbf{L}} + \dots + \hat{w}_{\mathbf{L}^{m-1}} (0,0,\dots,0) \cdot \mathbf{L}^{m-1} \leq 0$$

for every traceless tensor $\dot{\mathbf{L}}, \ddot{\mathbf{L}}, \dots$, and $\mathbf{L}^{m-1}$. It is clear that this requires

$$\hat{w}_{\mathbf{L}}(0,0,\dots,0) = \hat{w}_{\dot{\mathbf{L}}} (0,0,\dots,0) = \dots = \hat{w}_{\mathbf{L}^{m-2}} (0,0,\dots,0) = 0 \quad . \quad \blacksquare$$

As we have seen, for any of our class (m,n) fluids, the number m cannot exceed $n-1$. In particular, then the stress is of the form

$$\mathbf{T} = -p\mathbf{1} + \hat{\mathbf{T}} \left(\mathbf{L}, \dot{\mathbf{L}}, \dots, \mathbf{L}^m, \mathbf{L}^{m+1}, \dots, \mathbf{L}^{n-1} \right) \quad .$$

Our next theorem sharply restricts the dependence of $\hat{\mathbf{T}}(\cdot)$ on the higher strain rates $\mathbf{L}^m, \dots, \mathbf{L}^{n-1}$. Indeed, we have

Theorem 3. *For any fluid of class (m,n) it is necessary that*

$$\hat{\mathbf{T}} \left(0,0,\dots,0, \mathbf{L}^m, \mathbf{L}^{m+1}, \dots, \mathbf{L}^{n-1} \right) \cdot \mathbf{L} = \rho \hat{w}_{\mathbf{L}^m} (0,0,\dots,0) \cdot \mathbf{L} \otimes \mathbf{L} \quad . \quad (3.4)$$

for every $\mathbf{L}, \mathbf{L}^m, \mathbf{L}^{m+1}, \dots, \mathbf{L}^{n-1}$ in T^o .

Proof: In (3.1) set $\dot{\mathbf{L}} = \ddot{\mathbf{L}} = \dots = \mathbf{L}^{m-1} = 0$. We thus find that

$$\rho \hat{w}_{\mathbf{L}^m} (\mathbf{L}, 0, 0, \dots, 0) \cdot \mathbf{L} \leq \hat{\mathbf{T}} \left(\mathbf{L}, 0, 0, \dots, \mathbf{L}^m, \mathbf{L}^{m+1}, \dots, \mathbf{L}^{n-1} \right) \cdot \mathbf{L} \quad .$$

In this last replace $\mathbf{L}$ with $\gamma\mathbf{L}, \gamma > 0$, divide by γ , and let $\gamma \searrow 0$. Since $\hat{w}_{\mathbf{L}^m} (0,0,\dots,0)$ vanishes by Theorem 2, we may employ L'Hospital's rule to calculate the limit of the term on the left hand side. We thereby find

$$\rho \hat{w}_{\mathbf{L}^m} (0,0,\dots,0) \cdot \mathbf{L} \otimes \mathbf{L} \leq \hat{\mathbf{T}} \left(0,0,\dots, \mathbf{L}^m, \mathbf{L}^{m+1}, \dots, \mathbf{L}^{n-1} \right) \cdot \mathbf{L} \quad ,$$

and, since $\mathbf{L}$ appears linearly in this inequality, only equality can hold. $\blacksquare$

Corollary. *For any fluid of class $(m,n), m < n-1$, it is necessary that*

$$\hat{\mathbf{T}} \left(0,0,\dots,0, \mathbf{L}^{m+1}, \mathbf{L}^{m+2}, \dots, \mathbf{L}^{n-1} \right) \cdot \mathbf{L} \equiv 0 \quad ,$$

for every $\mathbf{L}$, $\overset{m+1}{\mathbf{L}}$, $\overset{m+2}{\mathbf{L}}$, ..., $\overset{n-1}{\mathbf{L}}$ in T^o .

Besides linking certain coefficients of $\hat{\mathbf{T}}(\cdot)$ to the second derivative of $\hat{w}(\cdot)$ at the rest state, Theorem 3 tells us that $\hat{\mathbf{T}}\left(0, 0, \dots, 0, \overset{m}{\mathbf{L}}, \overset{m+1}{\mathbf{L}}, \dots, \overset{n-1}{\mathbf{L}}\right)$ (more properly, its projection on T_s^o) can be at most linear in $\overset{m}{\mathbf{L}}$ and can never have any real dependence on the tensors $\overset{m+1}{\mathbf{L}}, \dots, \overset{n-1}{\mathbf{L}}$. Thus, by thermodynamic necessity, fluids of differential type with $m < n - 1$ will *not* satisfy the assumptions laid down by Joseph (1981) or Renardy (1983): When $m < n - 1$, any coefficient of $\overset{n-1}{\mathbf{L}}$ in the stress form $\hat{\mathbf{T}}(\cdot)$ must vanish identically whenever $\mathbf{L}, \dot{\mathbf{L}}, \dots$, and $\overset{m-1}{\mathbf{L}}$ vanish, regardless of the values of $\overset{m}{\mathbf{L}}, \overset{m+1}{\mathbf{L}}, \dots$, and $\overset{n-1}{\mathbf{L}}$. *A fortiori*, there can be no constant coefficient of $\overset{n-1}{\mathbf{L}}$ in the form $\hat{\mathbf{T}}(\cdot)$. Even when $m = n - 1$, we will now show that, for $n = 3$ or larger*, thermodynamics requires physically reasonable fluids of differential type to be inconsistent with the assumptions which Joseph and Renardy deploy to show instability. That is, thermodynamically proper fluids of differential type are, of necessity, “degenerate”† in just such a way as to allow them to slip the noose of Joseph’s and Renardy’s theorems.

To demonstrate the above remarks, recall that Theorem 2 told us that the rest state $(0, 0, \dots, 0)$ must be a stationary point for $\hat{w}(\cdot)$ and that the function $\hat{w}_{\overset{\mathbf{L}}{m-1}}(0, \cdot, \cdot, \dots, \cdot)$ must vanish identically. When $m - 1 > 0$, i.e., when m is 2 or larger, this last implies that

$$\hat{w}_{\overset{\mathbf{L}}{m-1} \overset{\mathbf{L}}{m-1}}(0, 0, \dots, 0) = 0 \quad .$$

Suppose now that the stationary point possessed by $\hat{w}(\cdot)$ at $(0, 0, \dots, 0)$ is *not* a saddle point. That is, assume that $(0, 0, \dots, 0)$ is locally an extreme point for $\hat{w}(\cdot)$. It follows then that

*When $n = 1$ and the fluid admits partitioning by temporal scaling as in (2.6)_{1,2}, the fluid is just that of Navier and Stokes and no one has any qualms about the usefulness of this model. When $n = 2$ the fluid model contains second grade fluids as a special case, and (3.4) becomes the assertion that

$$\hat{\mathbf{T}}(0, \dot{\mathbf{L}}) \cdot \mathbf{L} = \rho \hat{w}_{\mathbf{L}\mathbf{L}}(0) \cdot \dot{\mathbf{L}} \otimes \mathbf{L} \quad .$$

This class of fluids has been analyzed by Dunn & Fosdick (1974) and rather more generally by Dunn (1982). A very large subclass of these fluids, viz., all those that are thermodynamically proper, have physically desirable stability characteristics.

†In the context here the term is Renardy’s, who, aware that his theorem would not apply to a large class of grade n fluids, dismissed all such materials as “degenerate”. This move is not so different from an analyst studying elastic fluids with negative specific heats and/or negative isothermal compressibilities, finding them to be unstable, and then dismissing materials with positive values for these moduli as “degenerate”.

$$\frac{d^2}{ds^2} \hat{w} \left(s\mathbf{L}, 0, 0, \dots, 0, s \mathbf{\overset{m-1}{L}} \right) |_{s=0} \geq (\leq) 0 \quad ,$$

for all $\mathbf{L}$ and $\mathbf{\overset{m-1}{L}}$ in T^o , according as the rest state locally minimizes or maximizes $\hat{w}(\cdot)$. Equivalently,

$$\begin{aligned} & \hat{w}_{\mathbf{L}\mathbf{L}}(0, 0, \dots, 0) \cdot \mathbf{L} \otimes \mathbf{L} + 2\hat{w}_{\mathbf{L}\mathbf{\overset{m-1}{L}}}^{\mathbf{\overset{m-1}{L}}}(0, 0, \dots, 0) \cdot \mathbf{\overset{m-1}{L}} \otimes \mathbf{L} + \\ & \hat{w}_{\mathbf{\overset{m-1}{L}}\mathbf{\overset{m-1}{L}}}^{\mathbf{\overset{m-1}{L}}}(0, 0, \dots, 0) \cdot \mathbf{\overset{m-1}{L}} \otimes \mathbf{\overset{m-1}{L}} \geq (\leq) 0 \quad . \end{aligned}$$

But, as we have seen, $\hat{w}_{\mathbf{L}\mathbf{\overset{m-1}{L}}}^{\mathbf{\overset{m-1}{L}}}(0, 0, \dots, 0) = 0$ whenever $m \geq 2$. Hence, this last inequality is linear in $\mathbf{\overset{m-1}{L}}$ and can therefore be true only if

$$\hat{w}_{\mathbf{L}\mathbf{\overset{m-1}{L}}}^{\mathbf{\overset{m-1}{L}}}(0, 0, \dots, 0) = 0 \quad .$$

In conjunction with Theorem 3, we see that we have just established

Theorem 4. *A fluid of class (m, n) , $m \geq 2$, for which the rest state affords $\hat{w}(\cdot)$ a local minimum or a local maximum must satisfy*

$$\hat{\mathbf{T}} \left(0, 0, \dots, 0, \mathbf{\overset{m}{L}}, \mathbf{\overset{m+1}{L}}, \dots, \mathbf{\overset{n-1}{L}} \right) \cdot \mathbf{L} \equiv 0 \quad ,$$

for all traceless tensors $\mathbf{L}, \mathbf{\overset{m}{L}}, \mathbf{\overset{m+1}{L}}, \dots, \mathbf{\overset{n-1}{L}}$.

Theorem 4 thus prohibits $\hat{\mathbf{T}} \left(0, 0, \dots, 0, \mathbf{\overset{m}{L}}, \mathbf{\overset{m+1}{L}}, \dots, \mathbf{\overset{n-1}{L}} \right)$ from depending in a non-trivial way[†] on any of the $n - m$ tensors $\mathbf{\overset{m}{L}}, \dots, \mathbf{\overset{n-1}{L}}$. Even for $m = n - 1$, Theorem 4 gives physically natural conditions[§] under which no term involving solely $\mathbf{\overset{n-1}{L}}$, $n \geq 3$, could ever appear in a non-trivial way in $\hat{\mathbf{T}}(\cdot)$. As a consequence, the instability analyses of Joseph and Renardy are rendered irrelevant for *any* fluid of differential type that obeys thermodynamics. Indeed, let ϕ_n and ϕ_{n-1} denote the scalar multipliers of, respectively, $\mathbf{\overset{n-1}{L}}$ and $\mathbf{\overset{n-2}{L}}$ which appear in the

[†]That is, in a way that is not purely spherical and which could not, therefore, be absorbed into the constitutively independent pressure, p .

[§]Recall that, depending on the suppressed thermal variables, $\hat{w}(\cdot)$ can be variously interpreted as the internal energy, the Helmholtz free energy, or even, with some trivial alterations in our equations, as the entropy. Conventionally, the first two of these are to be minimized in equilibrium, while the last is to be maximized there.

constitutive form (2.6)_{1,2} for the stress in a grade (m, n) fluid. Assume $n \geq 3$. Then, either by the Corollary of Theorem 3 if $m < n - 1$ or by Theorem 4 if $m = n - 1$, we see that ϕ_n must vanish, i.e., the ratio ϕ_{n-1}/ϕ_n which Joseph (1981) needs to be non-negative and Renardy (1983) needs to be non-zero must be of the form $\phi_{n-1}/0$ and so is undefined. Moreover, this is only half of the grief which befalls the fundamental ratio ϕ_{n-1}/ϕ_n of Joseph and Renardy: As our next two theorems will reveal, whenever $n \geq 4$, the remaining coefficient in their ratio, ϕ_{n-1} , must also vanish.

Let us first agree to say that a tensor-valued function $\mathbf{f} = \mathbf{f}(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{p}{\mathbf{L}})$ is affine in the argument $\overset{j}{\mathbf{L}}$, $0 \leq j \leq p$, if

$$\begin{aligned} \mathbf{f}(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{p}{\mathbf{L}}) &= {}^j\mathbf{f}^0(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{j-1}{\mathbf{L}}, \overset{j+1}{\mathbf{L}}, \dots, \overset{p}{\mathbf{L}}) \\ &\quad + {}^j\mathbf{f}^1(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{j-1}{\mathbf{L}}, \overset{j+1}{\mathbf{L}}, \dots, \overset{p}{\mathbf{L}})[\overset{j}{\mathbf{L}}], \end{aligned}$$

where the map ${}^j\mathbf{f}^1(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{j-1}{\mathbf{L}}, \overset{j+1}{\mathbf{L}}, \dots, \overset{p}{\mathbf{L}})[\cdot]$ is linear.

Now differentiate (2.6)₂ i times with respect to τ and set $\tau = 0$. It is then clear that each of the functions $\hat{\mathbf{S}}_i(\cdot)$ is affine in $\overset{i-1}{\mathbf{L}}$ with linear part ${}^{i-1}\hat{\mathbf{S}}_i^1(\cdot)[\overset{i-1}{\mathbf{L}}]$ independent of $\overset{i-2}{\mathbf{L}}$. Further, it is also clear that, for $i \geq 3$, each $\hat{\mathbf{S}}_i(\cdot)$ is also affine in $\overset{i-2}{\mathbf{L}}$ with linear part ${}^{i-2}\hat{\mathbf{S}}_i^1(\cdot)[\overset{i-2}{\mathbf{L}}]$ independent of $\overset{i-3}{\mathbf{L}}$ if $i > 3$. In particular therefore, the stress response function $\hat{\mathbf{T}}(\cdot)$ of a grade (m, n) fluid is affine in $\overset{n-1}{\mathbf{L}}$ with its corresponding linear part independent of $\overset{n-2}{\mathbf{L}}$. Moreover, if $n \geq 3$, then $\hat{\mathbf{T}}(\cdot)$ is also affine in $\overset{n-2}{\mathbf{L}}$ with, if $n > 3$, its corresponding linear part independent of $\overset{n-3}{\mathbf{L}}$. These two special properties[¶] of the stress form of grade (m, n) fluids are all that is required for our final two theorems. Thus, while grade (m, n) fluids are included as a special case, the next two theorems speak to a much broader class of fluids of differential type.

Consider the stationary point of $\hat{w}(\cdot)$ that occurs at the rest state. In general, we would expect this point to be not just a local extreme point for $\hat{w}(\cdot)$ but, in fact, a global extreme

[¶]Although we make no use of it here, more of course is true: First, ${}^{i-1}\hat{\mathbf{S}}_i^1(\cdot)[\overset{i-1}{\mathbf{L}}]$ is independent of every $\overset{j}{\mathbf{L}}$, $j = 0, 1, \dots, i-2$. Second, ${}^{i-2}\hat{\mathbf{S}}_i^1(\cdot)[\overset{i-2}{\mathbf{L}}]$ is independent of every $\overset{j}{\mathbf{L}}$, $j = 1, 2, \dots, i-3$, when $i > 3$. And, third, the stress response function of a grade (m, n) fluid will always be affine in any $\overset{j}{\mathbf{L}}$ with $j > \frac{n}{2} - 1$, and its corresponding linear part will be independent of $\overset{j-1}{\mathbf{L}}$ if $j > \frac{n}{2} - \frac{1}{2}$.

point. That is, we would expect $\hat{w}(\cdot)$ to take on its absolute minimum or maximum at the rest state and so be bounded either below or above. This weaker idea of one-sided boundedness suffices for

Theorem 5. *Consider those fluids of class (m, n) , $n \geq 3$, for which $\hat{w}(\cdot)$ is bounded above or below and $\hat{\mathbf{T}}(\cdot)$ is affine in $\overset{n-1}{\mathbf{L}}$ with linear part independent of $\overset{n-2}{\mathbf{L}}$. Then*

$$m < n - 1 \quad , \quad (3.5)_1$$

and

$$\overset{n-1}{\hat{\mathbf{T}}}^1 \left(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-3}{\mathbf{L}} \right) \left[\overset{n-1}{\mathbf{L}} \right] \cdot \mathbf{L} \equiv 0 \quad , \quad (3.5)_2$$

for all traceless tensors $\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-3}{\mathbf{L}}$, and $\overset{n-1}{\mathbf{L}}$.

It should be noted that Theorem 5 does *not* say that the fluids of its hypotheses are really class $(m, n - 1)$ fluids. For this we would need to have

$$\overset{n-1}{\hat{\mathbf{T}}}^1 \left(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-3}{\mathbf{L}} \right) \left[\overset{n-1}{\mathbf{L}} \right] \equiv 0 \quad ,$$

and $(3.5)_2$ is not this strong. What $(3.5)_2$ does say, however, is that, whatever the stresses associated with $\overset{n-1}{\hat{\mathbf{T}}}^1(\cdot)[\cdot]$ are, they must always be *workless*. It is also worth noting that the requirement $n \geq 3$ is crucial: the theorem fails for $n = 2$, a fact which gives class $(m, 2)$ fluids much of their charm, as can be seen, for example, in the work of Dunn & Fosdick (1974) and Dunn (1982).

Proof: Our fundamental inequality (2.3) here takes the form

$$\rho \left\{ \hat{w}_{\mathbf{L}} \cdot \dot{\mathbf{L}} + \hat{w}_{\dot{\mathbf{L}}} \cdot \ddot{\mathbf{L}} + \dots + \hat{w}_{\overset{m}{\mathbf{L}}} \cdot \overset{m}{\mathbf{L}} \right\} \leq \overset{n-1}{\hat{\mathbf{T}}}^0 \cdot \mathbf{L} + \overset{n-1}{\hat{\mathbf{T}}}^1 \left[\overset{n-1}{\mathbf{L}} \right] \cdot \mathbf{L} \quad , \quad (3.6)$$

where, by Theorem 1, $m < n$. If $m < n - 1$, then (3.6) is linear in $\overset{n-1}{\mathbf{L}}$ and so can be easily violated unless its linear part $\overset{n-1}{\hat{\mathbf{T}}}^1 \left[\overset{n-1}{\mathbf{L}} \right] \cdot \mathbf{L}$ is identically zero, *i.e.*, unless $(3.5)_2$ holds.

Now assume for contradiction that $m = n - 1$. In this case (3.6) is still linear in $\overset{n-1}{\mathbf{L}}$ but now the vanishing of its linear part means that

$$\rho \hat{w}_{\overset{n-2}{\mathbf{L}}} \left(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-2}{\mathbf{L}} \right) \cdot \overset{n-1}{\mathbf{L}} = \overset{n-1}{\hat{\mathbf{T}}}^1 \left(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-3}{\mathbf{L}} \right) \left[\overset{n-1}{\mathbf{L}} \right] \cdot \mathbf{L} \quad , \quad (3.7)$$

for all $\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-3}{\mathbf{L}}, \overset{n-2}{\mathbf{L}}, \overset{n-1}{\mathbf{L}}$ in T^o . Since ${}^{n-1}\hat{\mathbf{T}}^1(\cdot)[\cdot]$ is independent of $\overset{n-2}{\mathbf{L}}$, the right hand side of (3.7) will be independent of $\overset{n-2}{\mathbf{L}}$ whenever $\mathbf{L}$ is independent of $\overset{n-2}{\mathbf{L}}$, i.e., whenever $n-2 > 0$. In this case, an easy integration of (3.7) then gives

$$\rho\hat{w}\left(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-3}{\mathbf{L}}, \overset{n-2}{\mathbf{L}}\right) = \rho\hat{w}\left(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-3}{\mathbf{L}}, 0\right) + \\ {}^{n-1}\hat{\mathbf{T}}^1\left(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-3}{\mathbf{L}}\right)\left[\overset{n-2}{\mathbf{L}}\right] \cdot \mathbf{L} \ .$$

We see therefore that $\overset{n-2}{\mathbf{L}}$ appears linearly in $\hat{w}(\cdot)$ — but this contradicts our hypothesis that $\hat{w}(\cdot)$ is bounded above or below unless this dependence on $\overset{n-2}{\mathbf{L}}$ is illusory, i.e., unless

$${}^{n-1}\hat{\mathbf{T}}^1\left(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-3}{\mathbf{L}}\right)\left[\overset{n-2}{\mathbf{L}}\right] \cdot \mathbf{L} \equiv 0 \ ,$$

for every $\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-3}{\mathbf{L}}, \overset{n-2}{\mathbf{L}}$ in T^o . As a result

$$\hat{w}\left(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-3}{\mathbf{L}}, \overset{n-2}{\mathbf{L}}\right) = \hat{w}\left(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-3}{\mathbf{L}}, 0\right) \ .$$

The first of these last two equations is exactly (3.5)₂, the second is (3.5)₁. ■

Now the result (3.5)₂ of Theorem 5 is considerably stronger than the mere assertion that the (constant) scalar multiplier of $\overset{n-1}{\mathbf{L}}$ inside $\hat{\mathbf{T}}(\cdot)$ vanishes for a grade (m, n) fluid. Indeed, we know this last from a straightforward specialization of Theorem 4. The real significance of Theorem 5 lies in both (3.5)₁ and (3.5)₂ together: they show that a nontrivial but special dependence of $\hat{\mathbf{T}}(\cdot)$ on one of the kinematical rates $\overset{p}{\mathbf{L}}$ (viz. $\overset{n-1}{\mathbf{L}}$) can lead to further constitutive collapse both in $\hat{w}(\cdot)$ and in certain portions of $\hat{\mathbf{T}}(\cdot)$ that contribute to dissipation. Theorem 5 is thus a harbinger of both our final theorem and a result we will momentarily draw on from Dunn & Rajagopal (1992). The theorem we have in mind disposes of the remaining coefficient in the ratio of Renardy and Joseph — it too must vanish by

Theorem 6. *Consider those fluids of class (m, n) , $m \geq 2$, $n \geq 4$, for which $\hat{\mathbf{T}}(\cdot)$ is affine in both $\overset{n-1}{\mathbf{L}}$ and $\overset{n-2}{\mathbf{L}}$ with the linear part ${}^{n-1}\hat{\mathbf{T}}^1(\cdot)[\cdot]$ independent of $\overset{n-2}{\mathbf{L}}$. Suppose also that $\hat{w}(\cdot)$ is bounded above or below, and that the extreme point for $\hat{w}(\cdot)$ occurring at the rest state is not a saddle. Then*

$${}^{n-2}\hat{\mathbf{T}}^1(0, 0, \dots, 0)\left[\overset{n-2}{\mathbf{L}}\right] \cdot \mathbf{L} \equiv 0 \ , \quad (3.8)$$

for all $\mathbf{L}$ and ${}^{n-2}\mathbf{L}$ in T^o .

Proof: Our hypothesis on $\hat{\mathbf{T}}(\cdot)$ means that there are tensor-valued functions $\mathbf{A}(\cdot)$, $\mathbf{B}(\cdot)[\cdot]$, and $\mathbf{C}(\cdot)[\cdot]$ such that

$$\mathbf{T} = \mathbf{A}\left(\mathbf{L}, \dot{\mathbf{L}}, \dots, {}^{n-3}\mathbf{L}\right) + \mathbf{B}\left(\mathbf{L}, \dot{\mathbf{L}}, \dots, {}^{n-3}\mathbf{L}\right) \left[{}^{n-2}\mathbf{L}\right] + \mathbf{C}\left(\mathbf{L}, \dot{\mathbf{L}}, \dots, {}^{n-3}\mathbf{L}\right) \left[{}^{n-1}\mathbf{L}\right] .$$

Clearly, ${}^{n-1}\hat{\mathbf{T}}^1(\cdot)[\cdot] = \mathbf{C}(\cdot)[\cdot]$, ${}^{n-2}\hat{\mathbf{T}}^1(\cdot)[\cdot] = \mathbf{B}(\cdot)[\cdot]$, and, by Theorem 5,

$$\mathbf{C}\left(\mathbf{L}, \dot{\mathbf{L}}, \dots, {}^{n-3}\mathbf{L}\right) \left[{}^{n-1}\mathbf{L}\right] \cdot \mathbf{L} \equiv 0 .$$

As a consequence, our fundamental inequality (2.3) has here the form

$$\rho \left\{ \hat{w}_{\mathbf{L}} \cdot \dot{\mathbf{L}} + \hat{w}_{\dot{\mathbf{L}}} \cdot \ddot{\mathbf{L}} + \dots + \hat{w}_{\mathbf{L}^{(m)}} \cdot \mathbf{L}^{(m)} \right\} \leq \mathbf{A}\left(\mathbf{L}, \dot{\mathbf{L}}, \dots, {}^{n-3}\mathbf{L}\right) \cdot \mathbf{L} + \mathbf{B}\left(\mathbf{L}, \dot{\mathbf{L}}, \dots, {}^{n-3}\mathbf{L}\right) \left[{}^{n-2}\mathbf{L}\right] \cdot \mathbf{L} ,$$

with $m \leq n-2$. If $m < n-2$, then it is clear that the term $\mathbf{B}\left(\mathbf{L}, \dot{\mathbf{L}}, \dots, {}^{n-3}\mathbf{L}\right) \left[{}^{n-2}\mathbf{L}\right] \cdot \mathbf{L}$ must vanish identically in order for the inequality to be satisfied, and we are done. If $m = n-2$, then the inequality still contains ${}^{n-2}\mathbf{L}$ linearly, and we see that its satisfaction requires that

$$\rho \hat{w}_{\mathbf{L}^{(n-2)}} \left(\mathbf{L}, \dot{\mathbf{L}}, \dots, {}^{n-3}\mathbf{L}\right) \cdot {}^{n-2}\mathbf{L} = \mathbf{B}\left(\mathbf{L}, \dot{\mathbf{L}}, \dots, {}^{n-3}\mathbf{L}\right) \left[{}^{n-2}\mathbf{L}\right] \cdot \mathbf{L} .$$

Now $\hat{w}_{\mathbf{L}^{(n-2)}} (= \hat{w}_{\mathbf{L}^{(n-2)}})$ meets $\hat{w}_{\mathbf{L}^{(n-2)}}(0, \dot{\mathbf{L}}, \ddot{\mathbf{L}}, \dots, {}^{n-3}\mathbf{L}) \equiv 0$. Thus, if in this last we replace $\mathbf{L}$ with $\gamma\mathbf{L}$, $\gamma > 0$, divide by γ , and let $\gamma \searrow 0$, then we may use L'Hospital's rule to reach

$$\rho \hat{w}_{\mathbf{L}^{(n-2)}}(0, \dot{\mathbf{L}}, \ddot{\mathbf{L}}, \dots, {}^{n-3}\mathbf{L}) \cdot {}^{n-2}\mathbf{L} \otimes \mathbf{L} = \mathbf{B}(0, \dot{\mathbf{L}}, \ddot{\mathbf{L}}, \dots, {}^{n-3}\mathbf{L}) \left[{}^{n-2}\mathbf{L}\right] \cdot \mathbf{L} .$$

But, the extreme point for $\hat{w}(\cdot)$ occurring at the rest state is not a saddle, and $m \geq 2$. Consequently, as we have seen earlier, the partial derivative $\hat{w}_{\mathbf{L}^{(n-2)}}(0, 0, \dots, 0)$ must vanish, and so (3.8) must hold. ■

Finally, there is a result of Dunn & Rajagopal (1992) which bears on the matters we have discussed here. It asserts that the special constitutive structure of grade (m, n) fluids, $n \leq 5$, must suffer significant collapse if the dissipation inequality is required to hold in all motions. As will be clear, their result thus offers one more, particularly striking, illustration

that the underlying thermal physics makes the instability analyses of Joseph and Renardy not relevant to physically realistic grade n fluid models. Indeed, not only is the “ratio” ϕ_{n-1}/ϕ_n here of the form $0/0$, thermodynamics has several other significant and more subtle specializations that it requires of these models: Dunn & Rajagopal (1992) show that a *grade* (m, n) *fluid*, $n \leq 5$, whose stored energy function $\hat{w}(\cdot)$ is bounded above or below, with a relative minimum or maximum at the rest state, *can never in fact be more than a class* $(m, 2)$ *fluid*. Their result is thus seen to give rather general conditions under which the constitutive structure

$$\begin{aligned} \hat{\mathbf{T}}(\mathbf{L}, \dot{\mathbf{L}}, \ddot{\mathbf{L}}, \ddot{\mathbf{L}}, \ddot{\mathbf{L}}) = & \hat{\mathbf{S}}_1(\mathbf{L}) + \hat{\mathbf{S}}_2(\mathbf{L}, \dot{\mathbf{L}}) + \hat{\mathbf{S}}_3(\mathbf{L}, \dot{\mathbf{L}}, \ddot{\mathbf{L}}) \\ & + \hat{\mathbf{S}}_4(\mathbf{L}, \dot{\mathbf{L}}, \ddot{\mathbf{L}}, \ddot{\mathbf{L}}) + \hat{\mathbf{S}}_5(\mathbf{L}, \dot{\mathbf{L}}, \ddot{\mathbf{L}}, \ddot{\mathbf{L}}, \ddot{\mathbf{L}}) \quad , \end{aligned}$$

with the partial stresses $\hat{\mathbf{S}}_i(\cdot)$ as in $(2.6)_2$, must collapse to no more than

$$\hat{\mathbf{T}} = \hat{\hat{\mathbf{T}}}(\mathbf{L}, \dot{\mathbf{L}}) \quad ,$$

where $\hat{\hat{\mathbf{T}}}(\mathbf{L}, \dot{\mathbf{L}})$ is a very special polynomial in $\mathbf{L}$ and $\dot{\mathbf{L}}$. The dissipation inequality (2.3) is once again seen to be hardly an enthusiastic champion of the constitutive form $(2.6)_{1,2}$ for grade n fluids. Or, put another way: rather basic principles of thermodynamics suggest a need for deepening the program begun in Dunn (1982) where a study of the thermomechanics of the general class of complexity 2 fluids was initiated.

4 Final Remarks

We have seen that thermodynamics, through the dissipation inequality (2.3) and the requirement of one-sided boundedness on the stored energy, imposes significant restrictions on the forms $\hat{\mathbf{T}}(\cdot)$ and $\hat{w}(\cdot)$ which occur in the theory of fluids of differential type. Indeed, the dissipation inequality alone was seen to require that the stored energy $\hat{w}(\cdot)$ have a stationary value at the rest state and that m be less than n , where $\overset{m-1}{\mathbf{L}}$ and $\overset{n-1}{\mathbf{L}}$ are the highest order time derivatives of $\mathbf{L}$ to appear in $\hat{w}(\cdot)$ and $\hat{\mathbf{T}}(\cdot)$, respectively. Moreover, thermodynamic consistency was shown to require that the stress form $\hat{\mathbf{T}}(\cdot)$ must always be such that the stress $\hat{\mathbf{T}}\left(0, 0, \dots, 0, \overset{p}{\mathbf{L}}, \overset{p+1}{\mathbf{L}}, \dots, \overset{n-1}{\mathbf{L}}\right)$ is *spherical*, i.e., is a pure pressure. With $p = m + 1$,

this followed from the dissipation inequality alone whenever $m < n - 1$. It followed, with $p = m$, whenever $m \geq 2$ if both the dissipation inequality held and the rest state was not a saddle for $\hat{w}(\cdot)$. As we pointed out, an immediate consequence of this much of the stress form vanishing is that no thermodynamically sound grade (m, n) fluid, $n \geq 3$, will ever meet the hypotheses which Joseph (1981, 1990) and Renardy (1983) need to impose in order to prove the instability of the rest state. In fact, none of the special grade (m, n) fluids, $n \geq 2$, which these authors analyze are thermodynamically realistic: for $n \geq 3$, each of their particular grade (m, n) fluids violates the dissipation inequality (2.3) and/or requires a stored energy function which has a saddle at the rest state; for $n = 2$, the fluids Joseph studies are exactly those whose internal energy has no lower bound, *i.e.*, they are (rather unrealistic) reservoirs of infinite energy. The results of Joseph and of Renardy may thus be taken as demonstrating that those grade (m, n) fluids, $n \geq 2$, that are thermodynamically unrealistic are also unrealistic from the point of view of stability theory. Indeed, their work is one more illustration of the rich interplay that practice has taught us to expect between stability considerations and any well-wrought theory of thermodynamics.

In spite of the complete *mathematical* compatibility of our work with that of Joseph and Renardy, it appears to us that these authors have seriously misinterpreted their works' significance. As we discussed in Section 2, while there is no doubting the physically unrealistic instability they established for their special grade (m, n) fluids — indeed, established for any class (m, n) fluid that has the special linearization they require — both Joseph and Renardy (Joseph especially) seem to feel, if we read them right, that all *other* class (m, n) fluids are unnatural also. In both senses of the word, we have been unable to follow this odd point of view: As reckoned by thermodynamic and stability considerations, there is in fact a large variety of physically reasonable fluids of differential type. For grade 2 fluids that meet (1.6)_{1,2,3}, this was strongly suggested almost 30 years ago by the work of Ting (1963) and was made obvious by the work of Dunn & Fosdick (1974). It also was made obvious by the work of Fosdick & Rajagopal (1980) on grade 3 fluids that meet (1.7)₁₋₆ and by the work of Dunn (1982, 1990) on complexity 2 fluids. To be sure, there is good reason to doubt the *applicability* of the special grade 2 and grade 3 fluids of even these studies since, like grade (m, n) fluids in general, they embody an extremely special (indeed, we think even artificial) partitioning of the stress in terms of its sensitivity, through deformation rates, to changes in the time scale. This, however, has no bearing at all on their status as material models consistent with all known first principles of physics.* Moreover, as the work of Dunn (1990) reveals,

*All grade 2 and grade 3 fluid models that are thermodynamically sound have non-positive first normal stress differences, as do, at low shear rates, some of the special complexity 2 fluids studied by Dunn (1982).

there are close cousins of even the grade 2 fluid model that satisfy ideas of thermodynamics and stability, embody no particular notion of stress partitioning by time scale, and still, say, allow for modelling almost completely arbitrary viscometric data. Joseph, in some vague way, seems to feel all such models are discredited. No facts of which we are aware support him.

The point of view propounded by Joseph in 1981 and repeated in his 1990 textbook actually arose and has its roots in earlier writings of other authors. Indeed, this point of view has become so common that no historical analysis that failed to examine it could be complete. In singling out Joseph's work for special attention, we have endeavored to avail ourselves of a fairly current, fairly complete, and (going to one of the strengths of this author) strongly worded statement that we think not only captures the underlying ideas involved but also illustrates some of their consequences. In this regard, Joseph's concluding paragraph of 1981 is we think especially revealing. Joseph asserts that

"I say it is wrong to study stability using the constitutive expressions for 'fluids' of grade n . These expressions arise in response to slow deformations. In fact, there is no such thing as a constitutive equation without prior specification of the domain of deformations in which the constitutive equation lives. However good rigid body mechanics is for some problems, it is obviously no good for studying the deformation of strained bodies. It is equally no good to study stability with constitutive expressions which do not allow you to consider small disturbances with large frequencies. Such disturbances can never be classed as retarded and they do not lead to fluids of grade n ."

Unfortunately, little of this is satisfactory: What does the first sentence mean? The stability of *what* is wrongly studied with grade n fluids? Surely not grade n fluids themselves; this would be an absurd construction of Joseph's sentence and can scarcely be what he has in mind in concluding a 6 page paper on precisely this subject. If not grade n fluids, then perhaps Joseph means the stability of this or that real material. If so, it clearly will make all the difference in the world *which* real material one decides to study — just as it would make a difference which real material one applied the Navier-Stokes constitutive equations to. Or, perhaps, Joseph means that the stability of some *other* class of fluid *models* is wrongly studied with grade n fluids. But, if so, it would have been useful if he could have named the

While this certainly indicates that these models would fail to correctly capture normal stress effects in those real materials whose first normal stress difference is always positive, it seems a bit extreme on this basis alone, absent some more fundamental principle, to declare these models, one and all, "physically unsound". *Inapplicable* to certain real materials for certain purposes, yes; *physically unsound*, no.

class(es) he has in mind. (No doubt Joseph has at least “memory” or integral fluid models in mind. Does he have others? General fluids of differential type?) His statement, both vague and absolute, provides us with no illumination. The strength of his assertion aside, all that is really clear here — and it is highlighted by his use of quotation marks around the word *fluids* — is that Joseph just *doesn't believe* that grade n fluids, one and all, are reasonable models for material behavior. Whatever the merits of this belief, the *reasons* Joseph offers us in support of it are inadequate.

The sentiment of Joseph's second sentence is clear enough and is probably widely shared. Unfortunately, in the first place, this sentiment misses the mark for grade 0 and grade 1 fluids, *i.e.*, for perfect fluids and Navier-Stokes fluids, and it is not even strictly correct for grade 2 fluids.* Indeed, as we have seen, the entire class of fluids of differential type was introduced several years *before* Coleman & Noll introduced their retardation theorem and thereby drew attention to that extremely special subclass of differential type fluids now known as grade n fluids. More important than questions of priority, however, this sentence of Joseph seems to us to embody a major confusion: Strictly speaking — and it is probably not possible to overemphasize this — grade n fluids do not “arise” in any rigorous way from slow deformations at all.† The retardation theorem of Coleman & Noll produces a Taylor series

*True enough, as far as we know, the *name* “second-order fluid” made its first appearance in the 1960 paper of Coleman & Noll. But it is hard to imagine that anyone motivated by the 1958 work of Criminale, Ericksen, & Filbey would not find it a natural step to study the model material given by their stress form with constant material moduli, *i.e.*, our (1.1), the model material now known as a grade 2 fluid. In any case, the paper by Rajeswari & Rathna on the flow near a stagnation point of a material given by our (1.1) suggests that the point of view of Rivlin and of Rivlin & Ericksen, expressed as early as 1955, for motivating a class of “non-Newtonian, visco-elastic” fluid models had a following well before the work of Coleman & Noll. (The received date of the paper by Rajeswari & Rathna is only $7\frac{1}{2}$ months later than the received date of the work of Coleman & Noll.) Moreover, as was mentioned in Section 1, when $\alpha_1 \geq 0$ and $\alpha_1 + \alpha_2 = 0$, *i.e.*, when the conditions (1.7)_{2,3} hold, the model (1.1) is exactly the so-called co-rotational Jeffreys model with a time of relaxation equal to zero and a time of retardation equal to α_1/μ . As is well-known, this model is but one of an infinite class of frame indifferent material models that generalize to three dimensional motions and large deformations a model proposed 60 years ago by Jeffreys (1929) to describe the “imperfection of elasticity”.

†Even more so do they not “arise in *response* ...” (emphasis added). Joseph's loose approach to this issue is even more on display in his 1990 textbook. There (page 458) he asserts that the “constitutive equations” of grade n fluids “are asymptotic expressions designed only for nearly steady slow motion.” While we shall momentarily argue at length that the front end of this quote, *i.e.*, the phrase “asymptotic expressions” is wrongheaded, for now let us draw attention to just one word. *Designed?* Quite apart from the fact that no one really knows much about *designing* constitutive equations, let alone *designing* them for this or that response regime, we cannot help but wonder if Joseph also feels that lines and parabolas were *designed* by Brook Taylor. That they have no independent identity worthy of study apart from their appearance in

whose coefficients have a certain structure; it is not a theorem about, nor does it deliver, classes of materials. Of course, it may be used to *motivate* all sorts of things, including even the study of some particular class of material models. Although we do not find this a particularly compelling approach to any of the fluids of differential type, it does seem to be one of the ends that Joseph, along with many others, puts the retardation theorem to. This is fine, even unexceptional. However, having so let a class of models “rise” to one’s attention, it seems perverse to then not listen to what the canon of models, the collected laws or principles of physics, has to say about them *qua models*. Even the thermodynamic portions of the canon aside, the stability analyses of Joseph and of Renardy (and of Coleman, Duffin, & Mizel), by themselves, hardly lead to anything like what Joseph is saying, hardly lead to anything approaching the breadth of his rejection, hardly lead to any position more bold than something like

“It really doesn’t matter how they are motivated; those grade n fluids whose coefficients obey our special hypotheses, as well as even all those class (m, n) fluids whose linearization does the same thing, are physically unrealistic. Physically sound grade n fluids, as well as physically sound fluids of class (m, n) , are to be found *only* among those fluids of differential type whose linearization *fails* our hypotheses.”

Even though his mathematical analysis requires Joseph to *treat* grade n fluids (more precisely, to treat their linearizations) as exact, Joseph, like many others, never seems willing to *judge* them as exact. To judge them that is as exact models in their own right, as models unencumbered with prejudgments based on how they “arise” or how they are motivated. This, we think, is a mistake. Matters of motivation, after all, go to questions of psychology. While they may thus be of great concern and interest to scientists, they are of no concern, interest, or relevance to the laws of science. Whatever one’s motivations, surely the principles of physics should have the last word on judging the theoretical fitness (as opposed to the usefulness or the applicability) of a model.

Joseph’s third sentence is interesting only to the extent he fails to follow his own advice: In precise, mathematical language, he nowhere tells us just what he takes the constitutive domain of a grade n fluid to be. Even for grade 0 and grade 1 fluids, *i.e.*, even for perfect fluids and Navier-Stokes fluids, this is a nontrivial task that, before Joseph, nobody seems to have thought had much to do with “slowness”. In his fifth sentence, Joseph not only uses

Taylor’s Theorem.

the phrase “study stability” in the same vague way that marred his first sentence, he also then goes on to say something that, taken literally, is just plain not true: the constitutive expressions or response functions for grade n fluids, like the constitutive expressions for fluids of differential type generally, certainly *do* allow one “to consider small disturbances with large frequencies”. Indeed, the polynomial constitutive structure for the stress in grade (m, n) fluids even makes them a little nicer in this regard than general differential type fluids: for grade (m, n) fluids, the form $\hat{\mathbf{T}}(\mathbf{L}, \dot{\mathbf{L}}, \dots, \overset{n-1}{\mathbf{L}})$ is well-defined for *all* values of $\mathbf{L}$, $\dot{\mathbf{L}}, \dots$, and $\overset{n-1}{\mathbf{L}}$, *i.e.*, beyond smoothness sufficient to guarantee the existence of $\mathbf{L}$, $\dot{\mathbf{L}}, \dots$, and $\overset{n-1}{\mathbf{L}}$, *the possible motions of a grade (m, n) fluid are essentially arbitrary.*[‡] While we think Joseph is onto an important truth, his formal argument here is technically in error and is even a little distracting. The real issue is not the domain of this or that constitutive expression. The retardation theorem is not a theorem about constitutive domains at all. It is, instead, a theorem that asserts that two very distinct stress forms, each defined on its own, independent constitutive domain, are **asymptotically equivalent**, *i.e.*, give values for the stress that are equal, **to order n along certain, very special, one parameter families of deformation histories.**[§] Moreover, besides thus misreading the retardation theorem, Joseph (fortunately) does not take seriously his own counsel: Had he really believed, at the beginning of his paper, that grade n fluids cannot be used “to consider small disturbances with large frequencies”, Joseph would have never entered the putative velocity-pressure form

[‡]Those who still favor Joseph’s view might find it interesting to answer the question: Precisely how small do the “frequencies” of a motion have to be for it to either be entered into or arise from a grade n fluid model?

[§]We emphasize that this does *not* mean that grade n fluids are only “valid in the limit”, or that they are “asymptotes” or “asymptotic cases” of simple fluids with a special type of fading memory. No. Like any material of differential type, the fluids of grade n are models in their own right. The fact that the *values* of their stresses and those of some other model are asymptotically equal when examined on special families of deformation histories should not make us lose sight of the fact that typically much of the character of each fluid will involve response *off* these families, *i.e.*, in a general flow, whether we hold our attention on a given particle and consider time variations or whether we consider variations from particle to particle at a fixed time, the resulting family of deformation histories will almost surely *not* lie on (will almost surely *cut across*) the family of retardations of any single deformation history.

An analogy here might be helpful: Consider two functions defined on $\Re^{100}$ that are asymptotically equal to, say, order 2 in a curve parameter along some collection Ω of curves that terminate at the origin. Would we on this basis alone say that one of the functions (which one?) is an asymptote of the other? Notwithstanding their 2^{nd} order agreement at 0 along Ω , isn’t it generally to be expected that the most interesting properties of our two functions will probably hinge on their behavior off Ω ? What sort of confusion would we be inviting if, instead of just simply saying that at the origin the two functions are equal to order 2 on Ω , we pronounced one of the *functions* (again, which one?) as “valid only in the limit”, as “arising” due to Ω , as “designed” only for Ω .

$(\mathbf{v}, p)(\mathbf{x}, t) = e^{-\sigma t}(\zeta, \pi)(\mathbf{x})$ into his linearized balance equations, would have never considered extremely large eigenvalues to the resulting spectral problem for $\zeta(\mathbf{x})$, and so would have never found the Theorem that forms the content of his paper. To prove his theorem, Joseph has to reject his own ruminations on domain spaces.

Whatever he may have had in mind then, we think it clear that Joseph's discussion offers little real insight into either the retardation theorem or grade n fluids as models in their own right. But, while we have been unable to endorse his specific arguments, it nevertheless seems to us that Joseph senses valid issues here that are worth examining. His misguided targeting of constitutive domains aside, Joseph appears to have in mind really two ideas which, though connected, are slightly different. The first seems to be that somehow grade n fluids are irretrievably contaminated or impaired or flawed by the fact of their stress form's entanglement with and appearance in the retardation expansion. Such a feeling seems to inform his whole paper, and, when put this way, Joseph might be taken as expressing doubts not so much different than our own misgivings concerning the rigid partitioning of effects by time scale that grade n fluids embody. The trouble, of course, is that Joseph *does not* express it this way. He therefore never really names a *specific way* in which grade n fluids, *as models in their own right*, are impaired or contaminated.

Second, Joseph appears to think or sense that critical features of the response of many (all?) models can be lost with the truncation of a retardation expansion. If this is his idea, we agree with it. It is indeed "no good to study" the "stability" of flows for a model or, say, a model's thermodynamics with truncated constitutive expressions which fail to embody such aspects of the model's response. If this is a problem, then, notwithstanding the tone of Joseph's remarks, it is not a problem of grade n fluids, some of which have reasonable stability and thermodynamic characteristics, some of which do not. Rather, the "problem" lies in asking too much of (and so misreading) a retardation theorem that never laid claim to delivering truncations that capture anything so complex as the constitutive features of a model that govern the stability of its flows and/or its thermodynamics. That, despite later overly bold assertions about it in the secondary literature, never laid claim to providing anything more than asymptotic formulae for the *value* of the stress on certain, special, one parameter families of deformation histories.[¶] In any case, one can not help

[¶]When the truncations of the retardation expansion do not capture the stability and thermodynamic features of the original model, one has to wonder what else they fail to capture. In particular, something like the *gradient* of the stress may be approximated quite badly or not at all in the truncation of a retardation expansion. This point was made as long ago as 1965 by Truesdell & Noll and, of course, is crucial in assessing the outcome of entering such truncated expressions into, say, momentum balance. It is thus even a key issue for all but the most trivial steady motions, let alone the unsteady ones that Joseph (we think correctly)

but wonder if Joseph, with the hindsight provided by the stability analysis of Renardy and the thermodynamic analysis presented here, would not have put his closing paragraph a little differently, would not have made his strictures a little stronger, would not have said or endorsed something like

Unless — and this is only a necessary condition — the ratio ϕ_{n-1}/ϕ_n is of the form $0/0$ for $n \geq 4$, of the form $\phi_2/0$, $\phi_2 \geq 0$, for $n = 3$, or of the form ϕ_1/ϕ_2 , $\phi_1 \geq 0$ and $\phi_2 \geq 0$, for $n = 2$, the constitutive expressions for grade n fluids should not be used to study stability *or anything else* that entails those expressions being treated as models in their own right and, so, being entered into the laws governing momentum balance and energy dissipation. Moreover, when the ratio ϕ_{n-1}/ϕ_n fails the above criteria, as it typically will when these expressions are motivated by a retardation expansion of the Coleman-Noll type on materials with a certain, special type of fading memory, then, a priori, these expressions have no more status than the retardation theorem assigns them: for a single, fixed deformation history, to within order n in the retardation parameter, they may be used to approximate the stress on retardations of that deformation history, *if (and only if) the deformation history is already known*. In particular, there is no known or proven status for using these expressions to *find or compute* either motions or, indeed, any approximations of motions. This is only to be expected inasmuch as the retardation theorem makes no claim that its truncated expressions faithfully capture or embody any of the more complex aspects of the original model's response, makes no claim, that is, that the *processes* associated with its truncations have anything to do with the *processes* associated with the original material model. Indeed, the thermodynamics and stability of a given grade n fluid, whether its study be motivated by the retardation theorem or not, involve issues quite separate from questions of how well and in what sense(s) it might “approximate” some other model.

In the 8 or so years centered on 1960, a good number of papers were nominally devoted to the subject of how one model (more precisely, how one constitutive equation) might “approximate” the response of another. The chief effort and success of that period, the cleansing of non-essentials from the foundations of mechanics and the production of a blooming multitude of properly formulated constitutive models, made this an altogether natural next step.*

seems to be worried about.

*The idea of one *model* approximating the response of another *model* is, in principle, ultimately a question of mathematics. As such it contrasts sharply with that gathering of philosophical tinder which, when sparked, sometimes gives off smoke like “every model is ultimately only an approximation of reality” or “we can never know the constitutive equation for any real (*i.e.*, actual) material, and so everything we do is just approximate”. Unfortunately, these two very different notions of approximation are sometimes confusingly commingled.

However, in the way (if not with the precision) of Coleman & Noll, most of this work never went deeper than studying conditions under which different constitutive forms for (usually) the stress gave asymptotically equal stress *values*. No doubt the belief underlying such studies was that if the stress values of two given models were approximately equal for certain types of deformation histories then this would give rise to correspondingly close *behavior* of each model, *i.e.*, the *solutions* of the governing field equations for each model material would themselves be close. As experience with the retardation theorem and grade n fluid models reveals, this belief is at best naive, at worst wrong. If some refinement of it is true, then the terms — let alone proof — of that refinement have yet to be formulated.

While most of the early work on “approximate” response was far more informal than the retardation theorem of Coleman & Noll, much of it still survives in what is now the standard, textbook literature of rheology where it probably is intended to serve no more than a heuristic function. Unfortunately — and much of our discussion here and in Section 1 is no more than an abbreviated catalog of this — even as a heuristic it carries with it a danger, a danger that, *at the very least*, results can be entirely misread when the imputed approximation breaks down or fails to deliver “reasonable” answers. This misreading of results is aided and abetted by a curious trick our psychology can play on us: In science we are accustomed to asking little of an “approximation”; approximations can (and, if pushed hard enough, usually do) break down or give silly answers, and *this seldom bothers us*. Indeed, we usually then make up a little story that “explains” the breakdown and thereby convince ourselves that we understand the approximation all the better. This habit of absolving an “approximation” from certain select standards of reasonableness usually is fine and generally causes no harm when a process of solution perturbation (like, for example, that which leads to linear elasticity) is involved. On the other hand, precisely because approximations are not intended to be exact, the mere *naming* of something as an approximation can, all by itself, serve to mute issues that would be better vetted independently of any idea of approximation. This has occurred repeatedly with grade 2 fluids, and, of course, hinges critically on adopting the blinkered view that fluids of differential type are merely approximations to more “fundamental” or more “correct” models of fluid behavior. As we noted in Section 1, such a view is anti-historical, and yet it is surprisingly common in certain quarters of the rheological community. While it would be impossible to trace its many expressions and/or consequences, any short list of important sources where one or both occur would have to include

- Besides their already referenced paper (1966 a), Metzner, White, & Denn again write (1966 b, pages 84 and 91) as if fluids of differential type occur only as truncated expansions or “are limited asymptotes” of more exact fluid models, “valid only in flow fields characterized by

low values of the Deborah number.”[†] But this is of course wrong: the retardation theorem says absolutely nothing about when and for what one can use one of its truncations. The *truncations* are not what is limited to “slow flows”; rather, when they are each evaluated on a family of retarded histories, it is the *difference* between the value of some general stress form and the value of any one of these truncations that is limited (to some appropriate order in the retardation parameter).

- Four years later, Walters (1970) first discusses the retardation theorem of Coleman & Noll and then states correctly that “Rivlin and Ericksen (1955) formulated equations of state on the basis that the stress is a function of the velocity gradients, acceleration gradients ... $(n - 1)^{\text{th}}$ acceleration gradients. They deduced that the stress needed to be expressed as a function of the n Rivlin-Ericksen tensors ...”. Walters then immediately asserts that “The Rivlin-Ericksen fluids may therefore be considered to be approximations to the simple fluid which are valid under the conditions of retarded motions or rapidly-fading memory discussed in Section 2.” This position is repeated almost verbatim 5 years later in his book, Walters (1975). As we have seen, remarks like these create a totally false setting for the bulk of fluids of differential type since, as far as is known now (1992), most of them *cannot* be considered to be approximations to some one or another general simple fluid.
- In their often cited major review article of 1976, Petrie & Denn state (page 226) that “A rather different constitutive instability, which has no physical relevance whatsoever, is associated with the use of the second-order fluid [Equation (6)] and other differential constitutive equations.” They then discuss the result of Coleman, Duffin, & Mizel (without any reference to the decisive role played by the sign of α_1) and then state that “The basic point is that the second-order fluid is an approximation which is valid only for flows which vary slowly on a time scale associated with the fluid memory.” In this Petrie & Denn, of course, echo the discussion of Metzner, White, & Denn (1966 a) who, as we saw in Section 1, found a solution to the “(CDM) paradox” in the asymptotic status of the “Rivlin-Ericksen formulation.” But this easy appeal to the idea of approximation won’t do: Even if grade 2 fluids were not a valid fluid model in their own right, a simple appeal to their status as an “approximation” would not constitute an explanation of the instabilities and non-existence found by Coleman, Duffin, & Mizel since there are no instabilities and no unpleasant non-existence results for

[†]Like (1966 a), the paper (1966 b) contains absolutely no mention of the crucial qualifier that α_1 must be taken negative to reach the non-existence result of Coleman, Duffin, & Mizel. Having thus re-created the “CDM paradox”, which they apparently first christened in (1966 a), Metzner, White, & Denn then go on to counsel (page 91) that “To obtain a solution, Coleman, Duffin, & Mizel *should* have used” (emphasis added) a constitutive expression different than the one they chose to study! Would this be the stance of these authors if an analyst, ignoring stability and/or thermodynamic considerations, chose to study the Navier-Stokes equations with the viscosity taken negative? Would it constitute proper reportage of such an analyst’s efforts to omit any reference to the critical role played by his choice of sign for the viscosity?

perfect fluids and Newtonian fluids, materials which according to this narrow point of view are, respectively, just 0th and 1st order “approximations”. Why is the 2nd special? This is not answered by Metzner, White, & Denn, nor is it answered by Petrie & Denn.

- Although it is not what the retardation theorem in fact says, even the key 1960 paper by Coleman & Noll establishing that theorem manages to plant the idea that a function of just the first derivative of the history at zero “often” “accounts well ...in the case of slow processes” for “phenomena which one would expect to be described exactly by” a general memory functional. Now these phrases appear in the fourth paragraph of the manuscript by Coleman & Noll and are certainly offered as no more than motivation for the rigorous results to follow.[†] Unfortunately, the marriage of the two overly broad ideas “accounts well ...in the case of slow processes” and “phenomena” leads the reader to the very threshold of the universe of “approximate constitutive equations.” Indeed, in his 1985 sketch Coleman himself seems to us to step over that threshold when he tells us that the retardation theorem “makes precise the asymptotic status of rate-type *materials* within the class of materials with gradually fading memory” (emphasis added). While Coleman may have in mind a conception of *materials* sufficiently enfeebled to make his statement true, we think many others will, very naturally, endow that word with robust associations involving field equations, processes, and boundary conditions. Associations that are well outside the compass of the retardation theorem.
- Although much of the work of Pipkin and his co-workers in fact draws more on perturbation techniques rather than on any idea of one constitutive model approximating another, there are some odd remarks of Pipkin which must be discussed if only for the fact that many later researchers use them as justification for their own approach.[§]

On page 90 Pipkin (1966) states that

“It is customary and comfortable to regard a constitutive equation as defining an idealized material, whose behavior under all conditions of operation is described by that same constitutive equation. This convention is useful and for many purposes inescapable, but it should not be adopted when dealing with second- or higher-order approximations. A second-order approximation is likely to lead to absurd results if applied to flow conditions outside the range for which the equation was intended, even if the corresponding first approximation produces no absurdity.”

[†]Indeed, Coleman & Noll’s fourth paragraph is probably most naturally interpreted as a *reversal* of the true logic of discovery and proof of the theorems to follow. It is almost certainly intended not to precisely summarize those theorems but, rather, to suggest their naturalness and plausibility.

[§]As a recent example, we direct the reader to the efforts by Giesekus (1989) and by Hassager, Armstrong, & Bird (1990) to evade or refute the interesting work of Kaloni (1989) on higher-order boundary conditions.

Now in the first place, we would have to disagree that it is merely by custom and comfortability that constitutive equations are regarded as defining an idealized material. This is the very *definition* of constitutive equations, *i.e.*, they define, precisely and without equivocation, idealized materials. What else would Pipkin suggest that constitutive equations are? With the trailer about “whose behavior under all conditions of operation,” does Pipkin mean to imply that somehow, *independent of constitutive equations*, we can confront and apprehend “idealized materials?” We of course do this routinely with real materials, but we are unable to understand any way to even *think* of “idealized materials” absent constitutive equations.[¶]

Having reduced the concept of constitutive equations to the merely customary and comfortable, Pipkin proceeds to suggest that the idea be abandoned altogether “when dealing with second- or higher-order approximations.” Assuming for the moment that Pipkin really sees himself here as still talking about constitutive equations, we should take him seriously and ask a question: If a purported constitutive equation is just thrown to us over the transom, how are we to know that it is a first- or second- or higher-order approximation to anything? Of course, it may or may not be frame indifferent, may or may not be thermodynamically consistent, may or may not fail this or that criterion we set up for a “proper” constitutive equation. This is all fine. But by what means, operating on the orphaned expression alone, would we announce that it is, say, a third-order approximation to something else (which we do not know!)?

But all this is probably byplay. Pipkin, we suspect, will know that he is dealing with an approximation, and he will know at each step the order of that approximation precisely because, rather than it having been tossed over the transom, it will have been *generated* by Pipkin by a formal process of perturbation on the *solution* of this or that boundary-initial-value problem. But the iterated field equations and their solutions which are produced in this way will almost always be impossible to mistake for a constitutive equation. In spite of the way Pipkin starts off then, and in spite of the fact that the use of certain constitutive equations is what he seems to want to discourage, the entire procedure Pipkin describes is unlike any *constitutive equation* one normally works with. It certainly does not correspond to what one does when one enters, say, the constitutive equation of a grade 2 fluid into momentum balance and tries to solve the resulting non-linear equations exactly. If we read him right, Pipkin has mixed up the idea of constitutive equations with the sequence of solution iterates that emerges from a perturbation process. He then uses the sometimes singular qualities

[¶]Pipkin may mean only that certain constitutive equations, *in certain portions of their domains*, yield processes which we do not like or which we find “unnatural.” This is often the case and is unremarkable: Sometimes we use such findings to motivate our cutting out the offending portion of the constitutive domain; on other occasions we use entry into the offending subdomain as a trigger to initiate additional constitutive response that yields more “subtle” and “natural” behavior. At each turn however, we use constitutive equations to define and characterize ideal materials.

of the later to impugn the former. This seems a serious mistake: Precisely because they define ideal materials, constitutive equations rise or fall according as they comply or not with the rules that govern the realm of the ideal, the collected principles of physics.* This is a totally different set of criteria than that which mathematicians use to judge the soundness of a perturbation scheme.

Next, on page 105 Pipkin (1966) amplifies his thoughts on the distinction between first-order and all higher-order approximations:

“First-order approximations can formally be regarded as obtained by setting some parameter equal to zero, . . . Once a parameter has been set equal to zero, it no longer appears in the equation. With the parameter no longer in evidence, it is possible to forget that smallness of the parameter is required for validity of the equation. Such forgetfulness can lead to inaccurate results, but absurdities based on the fact that the parameter is large cannot occur in a solution which does not contain the parameter. For this reason, first-order approximations usually have the self-consistency desired in the definition of an idealized material.

These remarks have more point, by contrast, when second- or higher-order approximations are considered. In a second-order approximation, the perturbation parameter is still present, even if it is not made explicit, and one can usually obtain absurd results by applying the equation to flows in which the second-order term is as large or larger than the first-order term.”

We find it especially difficult to follow Pipkin’s line of reasoning here. In particular, no clear idea seems to attach to the phrase “self-consistency desired in the definition of an idealized material,” and the idea of a parameter being “present, even if it is not made explicit” in an explicit mathematical expression seems quite puzzling. It does seem clear however that, except for the reference to “idealized materials,” Pipkin here intends his remarks to bear solely on the results of perturbation schemes and not on constitutive equations. Moreover, when they are taken in this way, those portions of Pipkin’s discussion which we follow seem valid enough. It is strange therefore that Pipkin then offers as a specific illustration of his remarks the analysis of Coleman, Duffin, & Mizel (1965), an analysis which of course treats the grade 2 fluid model as exact and which, because the underlying equation turns out to be linear, requires no idea of perturbation or of approximation. While Pipkin and, we are sure, others would no doubt argue that the essential approximation is committed at the outset by taking the grade 2 fluid model as an object of study[†], we think that there is much less to

*Whether a given constitutive equation is “interesting” or “plausible” or “natural” or “applicable to a real world problem” are all valid, but different, issues.

[†]Not always an object for very complete study, however: In his discussion of the “interesting absurdity” which he feels Coleman, Duffin, & Mizel brought to light, Pipkin passes over without comment his tacit but

this point than meets the eye. To begin with, it should be noted that the fairly standard notion of “approximation” that Pipkin employs in the above passages does not really connect very well with the so-called “approximation” involved in using truncations of the retardation theorem as material models. Here again Pipkin seems to be mixing two different concepts, this time two different (probably two vastly different) notions of “approximation”.[†]

Secondly, one wonders what would become of Pipkin’s (page 106) “interesting absurdity which arises when the following particular slow-motion approximation” (*i.e.*, the grade 2 fluid) “is treated as if it were exact” if history were reversed. If, that is, the work of Coleman, Duffin, & Mizel had been published — as mathematically it could have been published — in, say, 1956, *after* fluids of differential type had been defined but *before* the retardation theorem of Coleman & Noll (1960) had even been formulated. In 1956 there would have been no “slow-motion approximations” for Pipkin to appeal to, and yet Coleman, Duffin, & Mizel would still have found the “interesting absurdity” of instability and non-existence for those grade 2 fluids which have $\alpha_1 < 0$. In truth, it is just not logically satisfying to try to tie the unrealistic behavior of those grade 2 fluids with $\alpha_1 < 0$ to the livery which they wear by virtue of some one or another scheme of “approximation” (that may not even be formulated as of any given moment). Far more appropriate it seems to us is to turn things around: With the results of Coleman, Duffin, & Mizel (and many others) in hand, we should challenge as perhaps only an “interesting absurdity” any purported scheme of “approximation” that, without sharply limiting the allowed flow regimes, truly offers up grade 2 fluid models with $\alpha_1 < 0$ as approximates of well-behaved materials.

In his widely referenced Lectures of 1972, Pipkin extends and enlarges on some of his earlier assertions. Specifically, on page 145 Pipkin (1972) tells us that

“In order to get correct answers when using slow motion approximations, it is necessary to use ordinary perturbation methods, starting from the Newtonian (Navier-Stokes) solution as the lowest-order approximation. In other words, it is necessary that the solution can be expanded in powers of a small parameter, with the Newtonian flow solution as the first term, and that the higher-order terms do indeed represent only a small perturbation on this first term. If a solution is obtained directly without using a perturbation procedure, it is valid only if it is possible to expand it in this way. It is necessary to mention this because the higher-order approximate constitutive equations contain higher derivatives, and they can give rise to formal “solutions” which are not small perturbations on Newtonian flow. A “solution” which does not satisfy the criterion of slowness used

crucial sign choice amounting to $\alpha_1 < 0$.

[†]Additionally, we cannot help but wonder where in (1.1), the constitutive equation for a grade 2 fluid, is the “perturbation parameter” “still present”, but “not made explicit”.

in formulating the constitutive equation is merely a curiosity with no meaning.”

Following the above passage, Pipkin then proceeds to offer as a concrete “curiosity with no meaning” the “interesting absurdity” that he earlier felt emerged from the work of Coleman, Duffin, & Mizel. Since we have already commented on Pipkin’s approach to that work, we confine ourselves here to a few observations concerning the approach to perturbation methods which he is now advancing. We start by assuming that by “correct answers” Pipkin means *solutions* (unique?) to field equations but we are not so sure of the intended meaning of “ordinary perturbations.” If, as Pipkin would seem to endorse, these are to embrace the successive use of retardation truncations then Pipkin’s “ordinary perturbations” means, at the very least, *singular* perturbations, and these usually give rather “non-ordinary” results. Since Pipkin clearly sees that his planned strategy of perturbation will involve singular perturbations rather than regular perturbations, we are not sure what he intends by forswearing the “non-ordinary”. This brings us to a second, more basic, issue.

Why should the lowest-order approximation be the Newtonian solution? Why not the solution for an Eulerian perfect fluid? Why a solution involving either of these two classical special cases? Pipkin gives no reason for his choice but appears to just take for granted the (on the face of it, amazing) meta-theorem that all *non-Newtonian* fluids — except of course perfect fluids, which he does not mention — are Newtonian in “slow enough” flows. While it is daunting to try to envision the general mathematical formulation of such a conjecture, it seems impossible to imagine what could count as its experimental verification. With as much reason as Pipkin offers for his position, one could decree the same status for Eulerian perfect fluids, and, had such a decree been taken seriously at the right point in time, the development of the Navier-Stokes equations and boundary layer theory would have been significantly stunted. Following just one thread of Pipkin’s ideas, it seems to us neither true nor likely to be true that a delineation of higher-order boundary conditions and near-boundary behavior for general fluids is a mere curiosity.

Lastly, while Pipkin refers to “the criterion of slowness used in formulating the constitutive equation,” he nowhere ever tells us, in mathematically precise terms, what that criterion is for each of the models he is propounding. Indeed, while we know of no instance where a criterion of slowness has actually been used in *formulating* a constitutive equation, and while Pipkin offers no concrete example of a constitutive equation being so manufactured, it seems at least possible that one could “derive” constitutive equations from a variety of different “slowness” (and other) criteria. Which does Pipkin feel are “correct”? Why? Rather than such vague, ad hoc limitations on modelling, why not hold preconceptions in abeyance and, once a specific constitutive equation has been selected and rendered consistent with the formal principles of physics, let mathematics elucidate what is and is not significant in the structure of the solution fields associated with that constitutive equation. That knowledge in hand, we might

well face experiments in a way that lets us truly add to science's copybook of nature.

Perhaps the main lesson that experience with the retardation theorem and grade n materials ought drive home is the ultimately simple one that issues of some subtlety are involved in assessing when and in what way(s) a “lower order” model might be used to approximate the response of a “higher order” model. In what sense, for example, does a perfect fluid approximate a Newtonian fluid? A Newtonian fluid approximate a second grade fluid? In these two examples — and they must be among the most simple imaginable — the lower order model represents a singular perturbation of the higher order model we wish to approximate. In the first case, the higher order model possesses a viscosity μ , a sensitivity to boundary conditions, and a mechanism of dissipation that are completely unknown to the lower order, dissipationless perfect fluid. In the second case, a grade 2 fluid brings with it a new material parameter α_1/μ with the dimensions of time, non-zero normal stress differences, and a mechanism of diffusion for velocity and vorticity completely unknown to the Newtonian model. Far from the higher order model being “approximated” by the lower order one in any obvious fashion, the higher order model in each case brings with it a much richer physics that, usually, constitutes our main reason for studying it. Hard questions thus come up that ought be asked of *any* purported scheme of “approximation” involving such physically distinct models[§].

[§]As we have already lightly touched on, a particularly subtle and difficult issue for materials of differential type is the question of higher order boundary conditions: Unless the flow kinematics are very special, the equations of motion of a complexity n fluid form a system of partial differential equations of order $n + 1$ for the velocity field. Thus, when $n \geq 2$, the usual adherence boundary condition is, in general, insufficient to render a boundary-initial value problem determinate, and we are then faced with the task of finding realistic additional conditions that model the interaction of the fluid with its boundary. This, of course, raises experimental and theoretical considerations of a microstructural nature that currently appear extremely difficult to address, let alone resolve. While these issues have been recognized and discussed in the literature, they are in fact far from resolution. For problems in unbounded domains, investigators have advocated augmenting the usual boundary conditions with *a priori* assumptions on the behavior of the solution(s) at ∞ . Unfortunately, as Rajagopal (1984) discusses, this does not always work out. Further, the perturbation approach employed by Langlois & Rivlin (1990) to reduce the order of the underlying field equations is flawed as it is tantamount to treating a singular perturbation as a regular perturbation (see Rajagopal, Kaloni, & Tao (1989)). Confounding matters are the uniqueness results of Dunn & Fosdick (1974) for grade 2 fluids and of Fosdick & Rajagopal (1980) for grade 3 fluids which require of the competing fields only equality of the boundary conditions familiar from Navier-Stokes theory.

Until such questions of higher order boundary conditions are addressed and muted the general theory of fluids of differential type will remain stunted when compared to its first and still most vital special case, the Navier-Stokes fluid.

Indeed, it is worth contrasting this idea of one *model* “approximating” another *model* with the more familiar perturbation procedures used to build better and better approximations to the *solution* fields of a fixed, given model. In this latter case, we typically introduce a one parameter family of *solutions*, say $s(\cdot, \epsilon)$, to a given (system of) differential equation(s) governing a *single* model and seek to obtain (*i.e.*, approximate) some particular solution — call it $s(\cdot, 1)$ — in terms of, say, a power series $s(\cdot, 0) + \epsilon \frac{d}{d\epsilon} s(\cdot, 0) + \dots$ expansion in the parameter ϵ . At each step in this procedure, the original differential equation(s) and the earlier solutions $\frac{d^i}{d\epsilon^i} s(\cdot, 0)$, $i = 0, 1, 2, \dots, n$, completely determine the (linear) differential equation governing $\frac{d^{n+1}}{d\epsilon^{n+1}} s(\cdot, 0)$, and, at least in principle, it is clear how to take any given approximation to $s(\cdot, 1)$, say $p_n(\cdot) \equiv s(\cdot, 0) + \epsilon \frac{d}{d\epsilon} s(\cdot, 0) + \dots + \frac{1}{n!} \epsilon^n \frac{d^n}{d\epsilon^n} s(\cdot, 0)$, and use it to obtain an improved approximation to $s(\cdot, 1)$. While typically none of the iterates $p_n(\cdot)$ will meet exactly any of the underlying field equations, those field equations serve to structure and determine each $p_n(\cdot)$, just as they serve (at least in elementary problems) to determine a bound for the *error* between $s(\cdot, 1)$ and each $p_n(\cdot)$.

In the former case, however, when one model material is somehow expected to “approximate” another model material, everything is different. Now each member of an entire class of solutions to the basic field equations for one of our models, $\mathcal{M}$, *i.e.*, each member of a whole class of its *processes*, $\{s(\cdot)\}$, is expected to be approximated in some sense by an appropriate member of another class of processes, $\{s^0(\cdot)\}$, which describes the exact response of some (“zeroth-order”) approximating model, $\mathcal{M}_0$. Besides the underlying differential equations, complex questions concerning the *constitutive structures* of $\mathcal{M}$ and $\mathcal{M}_0$ will now enter into any rigorous discussion of the “error” made in “approximating” some particular process $s(\cdot)$ of $\mathcal{M}$ by some other “appropriate” process $s^0(\cdot)$ of $\mathcal{M}_0$. Moreover, contrary to the usual perturbation procedures, if we decide we want an “improved” approximation, then the zeroth-order process class $\{s^0(\cdot)\}$ is of no help to us. Indeed, rather than building on it to find an improved class of approximating processes, we cast $\{s^0(\cdot)\}$ aside and go to *another* (the “first-order”) approximating model material, $\mathcal{M}_1$, and determine *its* associated special class of processes, $\{s^1(\cdot)\}$, each member of which solves the (non-linear) differential equation(s) that constitute the basic field equations governing the response of this new model. Dogging us at each step of the way in any rigorous iteration scheme based on this idea will be subtle issues concerning the relation between the constitutive structure of $\mathcal{M}$ and the constitutive structure of each of the models $\mathcal{M}_0, \mathcal{M}_1, \dots, \mathcal{M}_n$, n finite or not.

This scheme of “iteration-by-model” can clearly be rather inefficient since none of the preceding iterates, $\{s^0(\cdot)\}, \{s^1(\cdot)\}, \dots, \{s^n(\cdot)\}$, are necessarily used in determining $\{s^{n+1}(\cdot)\}$.

Nevertheless, science would hardly be imaginable if such an iteration process was not oftentimes in fact valid. Indeed, the very act of selecting a model of a real material involves a fundamental appeal to an approximation scheme of this sort. What else do we have in mind when we *abstract* from the buzzing complexity of an actual material’s response those (very few) features we hope to approximate and study? In more mundane settings, without usually bothering to render it precise, we trust to such a scheme of approximation every time we use just the “approximately” known pressure function of a compressible gas or use just the “approximately” known viscosity of a Newtonian fluid. Such approximating hierarchies of models are thus inevitable. However, anyone who wishes to argue that the hierarchy of (grade n) models “suggested” by the retardation theorem is an approximating hierarchy in the above sense should keep in mind that there are no theorems to support this, no theorems to support approximating the behavior of a model with a “memory” with models that have no “memory” at all, no theorems to support or guide us in the use of such a hierarchy of singular perturbations. Such theorems would take an important step toward deepening the science of constitutive equations.

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