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# A Lagrangian formulation for the Oldroyd B fluid and the second principle of thermodynamics

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## Abstract

We show that the Oldroyd B fluid model is the Eulerian form of a Lagrangian model with an internal variable that satisfies the second principle of thermodynamics under some conditions on the initial value of the internal variable. We similarly derive a compressible version of the Oldroyd B model and several nonlinear versions thereof. We also derive Lagrangian formulations of the Zaremba-Jaumann and Oldroyd A fluid models. We discuss whether or not these new models satisfy the second principle.

## 1 Introduction

In [8], we introduced a fairly general framework for thermo-visco-elastic materials with or without internal variables that are frame-indifferent, may have all kinds of material symmetries, and satisfy the second principle of thermodynamics. Our work is based on the Coleman-Noll approach [1]–[2]. Among several other examples, we showed that the Oldroyd B fluid model, an incompressible complex fluid model introduced by Oldroyd in [9], could be made part of our framework by considering part of the Cauchy stress, the so-called polymer stress  $\sigma_p$ , as an internal variable. After having shown numerically that the Eulerian naive dissipation  $\sigma : d$ , which does not involve any internal variable, does not stay nonnegative, which means that this form of the second principle is violated by the Oldroyd B fluid, and by many other similar complex fluid models, we proceeded to show that there is no choice of Helmholtz free energy in the internal variable that would alleviate this violation. As a consequence, we cast some doubts on the thermodynamic validity of the Oldroyd B model, a question that we had not seen addressed in the literature.

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In the present paper, we show that our concerns were partially ill-founded by rewriting the Oldroyd B model as a Lagrangian model with an a priori more physical choice of internal variable and Helmholtz free energy, and by showing that in this form, it actually satisfies an adapted form of the second principle of thermodynamics. We take an altogether completely different route, inspired by a Lagrangian approach by Francfort and Lopez-Pamies, [4], who were motivated by the same thermodynamical concerns about the Oldroyd B fluid. They derived a quadratic Oldroyd B-like fluid model, with a slightly different objective derivative than the usual Oldroyd B derivative, starting from a standard generalized material point of view, see [6], which is meant to ensure that the second principle of thermodynamics is satisfied, albeit not necessarily in a very transparent manner. Their model is quadratic in the sense that the ordinary differential equation in time satisfied by the Cauchy stress  $\sigma$ , used in such models as a replacement for a constitutive law, features a square term  $\sigma^2$  in place of  $\sigma$  as in the usual Oldroyd B model. Francfort and Lopez-Pamies did not however obtain the classical, linear (with respect to  $\sigma$ ) Oldroyd B model.

We realized that the quadratic Oldroyd B-like model of [4] can alternatively be seen as fitting within our general framework of [8]. This prompted us to try and derive the classical Oldroyd B model in a Lagrangian formulation with an internal variable. It however turns out that this Lagrangian formulation is actually equivalent, with a specific choice of free energy, to the Eulerian one we introduced in [8], and thus cannot satisfy the second principle of thermodynamics for all possible evolutions.

We therefore propose here a conditional version of the second principle of thermodynamics, which is adequate for general models with internal variables, beyond the present Oldroyd B Lagrangian rewriting. Internal variables satisfy some differential constraint, here in the form of an ordinary differential equation, and the question of initial values for such an equation seems to be quite overlooked in the literature. It is actually crucial for frame-indifference and material symmetry issues, see [8], and it is absolutely crucial here for the second principle.

We thus say that the second principle is satisfied for an initial value of the internal variable if, given any admissible deformation, the corresponding dissipation remains nonnegative at all subsequent times. If the set  $\mathcal{C}$  of such initial values is nonempty, then we say that the second principle is conditionally satisfied under condition  $\mathcal{C}$ . Note that the set  $\mathcal{C}$  does not need to be invariant under the evolution of the internal variable, so it is not just a question of restricting the set of internal variables.

In the case of the Oldroyd B model, we entirely characterize the set of initial conditions that allow for the conditional second principle to hold. In Eulerian terms, this set corresponds to initial polymer Cauchy stresses that are positive semidefinite. Our main conclusion is therefore that the Oldroyd B fluid satisfies the conditional second principle of thermodynamics for these initial conditions and these initial conditions alone. Moreover, this is a case when the condition set  $\mathcal{C}$  is not invariant, thus making this result a rather subtle one.

This article is as follows. We first recall the notation of [8] and review briefly the notion of objective derivative and the classical Oldroyd B model. We then do a quick rundown of the thermo-visco-elastic theory with internal variables described in [8], in a simplified form appropriate for our purposes here. Next we show how the idea of [4] can be modified in order to obtain the

classical Oldroyd B fluid as a specific instance of such a visco-elastic material. We then show that this instance is equivalent to our previous Eulerian rewriting of Oldroyd B in [8], both in terms of internal variable and of internal dissipation.

At this point, we introduce the notion of conditional second principle as an adequate replacement of the Coleman-Noll view of the second principle, in the case of models with an internal variable. We identify the set of initial conditions that make the Oldroyd B fluid satisfy this version of the second principle, namely symmetric positive semidefinite matrices, in both Lagrangian and Eulerian descriptions.

We conclude the article by various generalizations of the Oldroyd B model and their connection with the conditional second principle, that are obtained by the same Lagrangian approach. These include a compressible version of the Oldroyd B fluid, incompressible nonlinear versions among which is the quadratic one obtained by Francfort and Lopez-Pamies, and the Zaremba-Jaumann and Oldroyd A fluids.

## 2 General notation

As in [8], we use the convention of denoting any quantity pertaining to the Lagrangian description with an uppercase letter and the corresponding Eulerian quantity with the corresponding lowercase letter. We also differentiate between a given quantity and a constitutive law for that same quantity by using a hat or a tilde for the latter, *e.g.*  $T_R$  for the first Piola-Kirchhoff stress tensor as opposed to  $\hat{T}_R$  for a constitutive law for this tensor. On the Lagrangian side of things, starting from a deformation mapping  $(X, t) \mapsto \phi(X, t)$ , we will use as thermodynamic variables the deformation gradient  $F(X, t) = \nabla_X \phi(X, t)$  and the deformation rate  $H(X, t) = \nabla_X V(X, t) = \frac{\partial F}{\partial t}(X, t)$ , where  $V$  is the velocity of particules. We will ignore thermal effects, that can easily be added. On the Eulerian side, and following the above convention, we let  $h(x, t) = \nabla_x v(x, t) = \nabla_X V(X, t)F^{-1}(X, t)$ , with the understanding that  $(x, t) = \Phi(X, t) = (\phi(X, t), t)$ , for the Eulerian velocity gradient,  $d(x, t) = \frac{1}{2}(h(x, t) + h(x, t)^T)$  for the stretching tensor and  $w(x, t) = \frac{1}{2}(h(x, t) - h(x, t)^T)$  for the spin tensor.

We denote the first Piola-Kirchhoff stress tensor by  $T_R$  and the Cauchy stress tensor by  $\sigma$ . Even though the latter is an Eulerian quantity, it will sometimes be expedient to look at it from the Lagrangian point of view. In particular, in the context of the Oldroyd B fluid, we will need to compute its material derivative  $\dot{\sigma} = \frac{\partial \sigma}{\partial t} + v_i \frac{\partial \sigma}{\partial x_i}$ , which is just  $\dot{\sigma} = \frac{\partial}{\partial t}(\sigma \circ \Phi)$ . We use the Helmholtz free energy with specific density  $A_m$ , and set the reference volumic mass to 1 for simplicity. We will most of the time work in an incompressible setting, expressed by the fact that  $\det F = 1$  in the Lagrangian description and  $\operatorname{div}_x v = 0$  in the Eulerian description. In this case, it is worth noticing that  $\sigma(x, t) = T_R(X, t)F^T(X, t)$  at all corresponding points.

We denote the set of  $3 \times 3$  matrices by  $\mathbb{M}_3$ . We let  $\mathbb{M}_3^+$  be the subset of matrices with strictly positive determinant,  $\operatorname{Sym}_3$  the set of symmetric matrices,  $\operatorname{Sym}_3^+$  the set of symmetric positive-definite matrices,  $\operatorname{Skew}_3$  the set of skew-symmetric matrices,  $\operatorname{SL}(3)$  the set of matrices with determinant 1,  $\operatorname{sl}(3)$  the set of trace-free matrices and  $\operatorname{SO}(3)$  the set of rotation matrices. For any  $M \in \mathbb{M}_3$ ,  $\operatorname{cof} M$  denotes the cofactor matrix of  $M$ , and  $\operatorname{Sym}(M)$  and  $\operatorname{Skew}(M)$  respectively

denote the symmetric and skew-symmetric parts of  $M$ .

### 3 Objective derivatives

Objective derivatives occur in situations when one wishes to differentiate Eulerian quantities, typically the Cauchy stress tensor, with respect to time, in a way that is compatible with frame-indifference. These are called derivatives even though they are not derivation operators in the technical sense. The earliest example of such an objective derivative is the Zaremba-Jaumann derivative, see [13].

In Lagrangian terms, we are considering two deformations  $\phi$  and  $\phi^*$  that are related via

$$\phi^*(X, t) = Q(t)\phi(X, t) + a(t), \quad (1)$$

where  $Q$  and  $a$  are arbitrary regular functions with values respectively in  $\text{SO}(3)$  and  $\mathbb{R}^3$ . We express (1) in Eulerian terms as  $(x^*, t) = (Q(t)x + a(t), t)$ .

The principle of frame-indifference, for which we refer to [12], requires that the corresponding Cauchy stresses must satisfy

$$\sigma^*(Q(t)x + a(t), t) = Q(t)\sigma(x, t)Q(t)^T, \quad (2)$$

or  $\sigma^*(x^*, t) = Q(t)\sigma(x, t)Q(t)^T$  with obvious notation.

Let us remark that the stretching tensor  $d$  is also frame-indifferent in the sense that

$$d^*(x^*, t) = Q(t)d(x, t)Q(t)^T, \quad (3)$$

whereas the velocity gradient  $h$  is not.

Loosely speaking, an objective derivative is a differential operator that is of first order in time and depends on  $h$  in such a way that it transforms as above in the same circumstances. It can thus be used for constitutive purposes to derive frame-indifferent models of differential type.

**Definition 3.1.** *A first order in time differential operator  $\hat{\diamond}$  is an objective derivative, or is objective, if*

$$\hat{\diamond}^*(x^*, t) = Q(t)\hat{\diamond}(x, t)Q(t)^T, \quad (4)$$

for all functions  $\sigma$ ,  $Q$  and  $a$  with values in  $\text{Sym}_3$ ,  $\text{SO}(3)$  and  $\mathbb{R}^3$  respectively.

It is clear that the material derivative does not satisfy (4) and is thus not an objective derivative. Actually, any derivation satisfies Leibniz's rule and therefore cannot be objective.

A constitutive differential equation for the Cauchy stress can then be assumed of the form

$$\hat{\diamond}(\sigma, t) = G(\sigma(x, t), d(x, t))$$

for instance in the simplest cases, and if  $G$  is itself frame-indifferent, *i.e.*,  $G(Q\sigma Q^T, QdQ^T) = QG(\sigma, d)Q^T$ , then the resulting differential model will satisfy the principle of frame-indifference.

Now there is no really natural standout candidate among objective derivatives. We describe below all the objective derivatives of the specific form

$$\hat{\diamond} = \dot{\sigma} + \text{Ob}(\sigma, h), \quad (5)$$

with  $\text{Ob}: \text{Sym}_3 \times \mathbb{M}_3 \rightarrow \text{Sym}_3$ .

A function  $\text{Ob}_s: \text{Sym}_3 \times \text{Sym}_3 \rightarrow \text{Sym}_3$  is said to be objective if

$$\text{Ob}_s(Q\sigma Q^T, QdQ^T) = Q\text{Ob}_s(\sigma, d)Q^T, \quad (6)$$

for all  $(\sigma, d) \in \text{Sym}_3^2$  and  $Q \in \text{SO}(3)$ . We note that the set of such objective functions is entirely characterized, see [11]. In particular, it contains all symmetric-valued polynomials in  $(\sigma, d)$ . The following result is stated in [5].

**Proposition 3.2.** *An operator of the form (5) is objective if and only if*

$$\text{Ob}(\sigma, h) = \sigma w - w\sigma + \text{Ob}_s(\sigma, d). \quad (7)$$

with  $w = \text{Skew}(h)$ ,  $d = \text{Sym}(h)$ , and  $\text{Ob}_s: \text{Sym}_3 \times \text{Sym}_3 \rightarrow \text{Sym}_3$  is an objective function.

*Proof.* Let  $\hat{\diamond}$  be an operator of the form (5). For all  $\text{SO}(3)$ -valued functions of time  $Q$ , we have

$$\begin{aligned} \hat{\diamond}\sigma^* &= \dot{\sigma}^* + \text{Ob}(\sigma^*, h^*) \\ &= \dot{Q}\sigma Q^T + Q\sigma\dot{Q}^T + Q\dot{\sigma}Q^T + \text{Ob}(Q\sigma Q^T, QhQ^T + \dot{Q}Q^T). \end{aligned}$$

equality being understood at corresponding space-time points  $(x^*, t)$  and  $(x, t)$ .

The operator  $\hat{\diamond}$  is thus objective if and only if

$$Q\text{Ob}(\sigma, h)Q^T = \dot{Q}\sigma Q^T + Q\sigma\dot{Q}^T + \text{Ob}(Q\sigma Q^T, QhQ^T + \dot{Q}Q^T), \quad (8)$$

for all functions  $\sigma$ ,  $h$ , and  $Q$ .

Let us first derive a series of necessary conditions for  $\hat{\diamond}$  to be objective. For all  $Q \in \text{SO}(3)$  and  $z \in \text{Skew}_3$ , we let  $Q(t) = e^{tz}Q$ , so that  $\dot{Q}(0) = zQ$ . Considering constant values for  $\sigma$  and  $h$ , we see that (8) implies that

$$Q\text{Ob}(\sigma, h)Q^T = zQ\sigma Q^T - Q\sigma Q^T z + \text{Ob}(Q\sigma Q^T, QhQ^T + z), \quad (9)$$

for all  $\sigma \in \text{Sym}_3$ ,  $h \in \mathbb{M}_3$ ,  $Q \in \text{SO}(3)$  and  $z \in \text{Skew}_3$ .

Taking  $z = 0$ , we obtain a first necessary condition,

$$Q\text{Ob}(\sigma, h)Q^T = \text{Ob}(Q\sigma Q^T, QhQ^T), \quad (10)$$

for all  $Q$ ,  $\sigma$  and  $h$ .

Taking next  $Q = I$  and  $z = -\text{Skew}(h)$ , we obtain a second necessary condition,

$$\text{Ob}(\sigma, h) = -\text{Skew}(h)\sigma + \sigma\text{Skew}(h) + \text{Ob}(\sigma, \text{Sym}(h)), \quad (11)$$

for all  $\sigma \in \text{Sym}_3$  and  $h \in \mathbb{M}_3$ . Defining  $\text{Ob}_s$  to be the restriction of  $\text{Ob}$  to  $\text{Sym}_3 \times \text{Sym}_3$ , we see that (7) holds. Moreover, by (10),  $\text{Ob}_s$  is an objective function from  $\text{Sym}_3 \times \text{Sym}_3$  to  $\text{Sym}_3$ .

Conversely, let us assume  $\text{Ob}$  to be of the form (7) with an objective function  $\text{Ob}_s$ , and see if the resulting operator  $\hat{\diamond}$  is an objective derivative. It is enough to check that (8) holds for all functions  $\sigma$ ,  $h$  of  $(x, t)$ , and  $Q$  of  $t$ .

Since  $Q$  is  $\text{SO}(3)$ -valued, we can write  $\dot{Q}(t) = z(t)Q(t)$  where  $z$  is a  $\text{Skew}_3$ -valued function. Let us compute  $\text{Ob}(Q\sigma Q^T, QhQ^T + \dot{Q}Q^T)$  using (6), omitting the time and space variables. We will need the following expressions

$$\begin{aligned} \text{Sym}(QhQ^T + \dot{Q}Q^T) &= QdQ^T, \\ \text{Skew}(QhQ^T + \dot{Q}Q^T) &= Q\text{Skew}(h)Q^T + z. \end{aligned}$$

Replacing them in (7), we obtain

$$\begin{aligned}
\text{Ob}(Q\sigma Q^T, QhQ^T + \dot{Q}Q^T) &= -(Q \text{Skew}(h)Q^T + z)Q\sigma Q^T \\
&\quad + Q\sigma Q^T(Q \text{Skew}(h)Q^T + z) + \text{Ob}_s(Q\sigma Q^T, QdQ^T) \\
&= -Q \text{Skew}(h)\sigma Q^T + Q\sigma \text{Skew}(h)Q^T \\
&\quad - zQ\sigma Q^T + Q\sigma Q^T z + Q\text{Ob}_s(\sigma, d)Q^T \\
&= Q(-\text{Skew}(h)\sigma + \sigma \text{Skew}(h) + \text{Ob}_s(\sigma, d))Q^T \\
&\quad - \dot{Q}\sigma Q^T - Q\sigma \dot{Q}^T \\
&= Q\text{Ob}(\sigma, h)Q^T - \dot{Q}\sigma Q^T - Q\sigma \dot{Q}^T,
\end{aligned}$$

that is to say (8).  $\square$

Many classical objective derivatives are of the form of Proposition 3.2, with  $\text{Ob}_s$  a simple symmetric-valued polynomial in  $(\sigma, d)$ .

- The Zaremba-Jaumann derivative  $\overset{\square}{\sigma} = \dot{\sigma} + \sigma w - w\sigma$ , with  $\text{Ob}_s = 0$ , which is thus the simplest objective derivative in this sense. Moreover, all other objective derivatives under consideration are of the form  $\overset{\hat{\square}}{\sigma} = \overset{\square}{\sigma} + \text{Ob}'_s(\sigma, d)$  for some other objective function  $\text{Ob}'_s$  (more generally, the difference between any two such objective derivatives is an objective function of  $(\sigma, d)$ ).
- The Oldroyd A or lower convected derivative  $\overset{\Delta}{\sigma} = \dot{\sigma} + h^T\sigma + \sigma h$  with  $\text{Ob}_s(\sigma, d) = d\sigma + \sigma d$ .
- The Oldroyd B or upper convected derivative  $\overset{\nabla}{\sigma} = \dot{\sigma} - h\sigma - \sigma h^T$  with  $\text{Ob}_s(\sigma, d) = -d\sigma - \sigma d$ .
- The Truesdell derivative  $\overset{\circ}{\sigma} = \overset{\nabla}{\sigma} + \text{tr}(h)\sigma$  with  $\text{Ob}_s(\sigma, d) = -d\sigma - \sigma d + \text{tr}(d)\sigma$ .

Note that the notation is not universal, and neither is the vocabulary, with corotational, covariant and contravariant rates also being in use for the first three objective derivatives.

We will encounter more objective derivatives of the same general form in Section 8.2. Some classical objective derivatives are not of this form, such as the Green-Naghdi derivative, which mixes Lagrangian and Eulerian elements. Unfortunately, even though it is apparently used in commercial software, it can be shown that this derivative gives different results with different choices of reference configuration. This sort of disqualifies it since arbitrary reference configuration choices for the same material must produce the same Eulerian behavior.

As we already mentioned, objective derivatives are too numerous, and there is no obvious way of choosing among them. Two objective derivatives however stand out in this respect. Indeed, it is well known that the Truesdell and Oldroyd B derivatives are produced by time differentiation of the Cauchy stress expressed with the second Piola-Kirchhoff stress in the compressible case and incompressible case respectively, with the simple formulas below.

**Proposition 3.3.** *Let  $\Sigma = F^{-1}\sigma \operatorname{cof} F$  be the second Piolà-Kirchhoff stress. For all deformations, we have*

$$\overset{\circ}{\sigma} = \frac{1}{\det F} F \frac{\partial \Sigma}{\partial t} F^T. \quad (12)$$

*In particular, in the incompressible case, we have*

$$\overset{\nabla}{\sigma} = F \frac{\partial \Sigma}{\partial t} F^T. \quad (13)$$

*Proof.* The relation between the Cauchy stress and the second Piolà-Kirchhoff stress reads

$$\sigma(\phi(X, t), t) = \frac{1}{\det F(X, t)} F(X, t) \Sigma(X, t) F^T(X, t).$$

Therefore, omitting  $(X, t)$  in the right-hand side for brevity,

$$\begin{aligned} \frac{\partial}{\partial t} (\sigma(\phi(X, t), t)) &= -\frac{1}{(\det F)^2} \frac{\partial}{\partial t} (\det F) F \Sigma F \\ &\quad + \frac{1}{\det F} \left( H \Sigma F^T + F \Sigma H^T + F \frac{\partial \Sigma}{\partial t} F^T \right) \\ &= -\frac{1}{\det F} \frac{\partial}{\partial t} (\det F) \sigma + H F^{-1} \sigma + \sigma F^{-T} H^T \\ &\quad + \frac{1}{\det F} F \frac{\partial \Sigma}{\partial t} F^T. \end{aligned}$$

We have  $\frac{\partial}{\partial t} (\det F) = \det F \operatorname{div}_x v$ , so that reinterpreting the above in (almost) Eulerian terms, we obtain

$$\dot{\sigma} = -\operatorname{tr}(h)\sigma + h\sigma + \sigma h^T + \frac{1}{\det F} F \frac{\partial \Sigma}{\partial t} F^T,$$

which is precisely equation (12).

If  $\det F(X, t) = 1$  for all  $(X, t)$ , then  $\operatorname{tr}(h) = 0$  and formula (12) reduces to formula (13).  $\square$

This result may induce a slight preference for the above derivatives in the incompressible and compressible cases respectively. Moreover, the second Piolà-Kirchhoff stress is especially well suited to the study of the Oldroyd B fluid model as we will see below.

## 4 The Oldroyd B complex fluid model

We give a very brief introduction to the Oldroyd B fluid model. We refer to [7] for historical and physical insights and [10] for a review of mathematical results pertaining to this model. The Oldroyd B model is a model for an incompressible viscoelastic fluid that is supposed to be a dilute suspension of polymer molecules in a Newtonian fluid solvent. It is a differential type model in the sense that the Cauchy stress is not given by a constitutive law, but by a first order differential equation in time that involves the Oldroyd B derivative

$$\sigma + \lambda_1 \overset{\nabla}{\sigma} = 2\eta(d + \lambda_2 \overset{\nabla}{d}), \quad (14)$$



where  $\eta > 0$  is a global viscosity coefficient and  $\lambda_1, \lambda_2 > 0$  are relaxation times. For the model to be physically relevant, it is assumed that  $\lambda_2 \leq \lambda_1$ . It is frame-indifferent by construction and by formula (3). Of course, the tensor  $\sigma$  above does not include the indeterminate pressure term  $pI$  that is the Lagrange multiplier of the incompressibility constraint  $\text{tr}(d) = 0$ . In the sequel, by Cauchy stress, we will mean Cauchy stress modulo the indeterminate pressure as long as no initial value is specified for the differential equation (14).

There is a classical additive decomposition of the Cauchy stress that simplifies equation (14), namely  $\sigma = \sigma_s + \sigma_p$ , obtained by letting  $\eta_s = \frac{\lambda_2}{\lambda_1}\eta$  and  $\eta_p = (1 - \frac{\lambda_2}{\lambda_1})\eta$  and

$$\sigma_s = 2\eta_s d \text{ and } \sigma_p + \lambda_1 \overset{\nabla}{\sigma}_p = 2\eta_p d, \quad (15)$$

where  $\sigma_s$  is the Newtonian solvent stress with solvent viscosity  $\eta_s$  and  $\sigma_p$  is interpreted as a polymer stress with polymer viscosity  $\eta_p$ . Conversely, (15) implies (14) with  $\eta = \eta_s + \eta_p$  and  $\lambda_2 = \frac{\lambda_1 \eta_s}{\eta_s + \eta_p}$  and the two formulations are equivalent.

There are many different ways of deriving the Oldroyd B model from various hypotheses. We concentrate below on a phenomenological Lagrangian approach with a view to testing the compatibility of the Oldroyd B model with the second principle of thermodynamics.

## 5 Viscoelastic materials with internal variables

We place ourselves within the general thermo-visco-elastic framework developed in [8], by using the thermodynamic variables  $F$  and  $H$ , complemented by a symmetric matrix-valued internal variable  $B_i$ , without thermal effects. This framework relies heavily on the exploitation of the second principle of thermodynamics or Clausius-Duhem inequality via the Coleman-Noll procedure, see again [8].

We are given a constitutive law for the Helmholtz free energy specific density of our material  $\hat{A}_m: \mathbb{M}_3^+ \times \mathbb{M}_3 \times \text{Sym}_3 \rightarrow \mathbb{R}$ , so that the free energy density at point  $(X, t)$  is given by  $A_m(X, t) = \hat{A}_m(F(X, t), H(X, t), B_i(X, t))$ . We also are given a constitutive law for the first Piola-Kirchhoff stress  $\hat{T}_R: \mathbb{M}_3^+ \times \mathbb{M}_3 \times \text{Sym}_3 \rightarrow \mathbb{M}_3$  so that likewise  $T_R(X, t) = \hat{T}_R(F(X, t), H(X, t), B_i(X, t))$ . Finally, we are given a differential constraint for the internal variable of the form

$$\frac{\partial B_i}{\partial t}(X, t) = \hat{K}(F(X, t), H(X, t), B_i(X, t)), \quad (16)$$

where the flow rule  $\hat{K}: \mathbb{M}_3^+ \times \mathbb{M}_3 \times \text{Sym}_3 \rightarrow \text{Sym}_3$  is the last constitutive ingredient of the model.

The above constitutive ingredients are written for a compressible material. Since we are also interested in incompressible materials, it is in this case enough to restrict all the above constitutive laws to  $F \in \text{SL}(3)$ , and  $(F, H)$  in the tangent bundle to  $\text{SL}(3)$ . We just keep this implicit. In addition, we still ignore the indeterminate pressure term in the stress tensors. This indeterminate pressure must naturally be taken into account in the dynamics equation.

With the above provisos in mind, the outcomes of the Coleman-Noll procedure are first that  $\widehat{A}_m$  does not depend on  $H$ , second that there is a natural decomposition of the constitutive law for the first Piola-Kirchhoff stress

$$\widehat{T}_R(F, H, B_i) = \widehat{T}_{Rd}(F, H, B_i) + \frac{\partial \widehat{A}_m}{\partial F}(F, B_i), \quad (17)$$

and third that the internal dissipation has a constitutive law given by

$$\widehat{D}_{\text{int}}(F, H, B_i) = \widehat{T}_{Rd}(F, H, B_i) : H - \frac{\partial \widehat{A}_m}{\partial B_i}(F, B_i) : \widehat{K}(F, H, B_i). \quad (18)$$

The second principle reduces here to the mechanical part of the Clausius-Planck inequalities, see [8],  $\widehat{D}_{\text{int}}(F, H, B_i) \geq 0$  for all possible arguments, in the Coleman-Noll approach. This is a constitutive restriction. The internal dissipation is a power term which appears as a source term in the heat equation when thermal effects are also taken into consideration. We will see later on that in the case of the Oldroyd B fluid, we have to abandon part of the Coleman-Noll approach. Instead, we will identify which evolutions of the internal variables make the internal dissipation given by the above constitutive law nonnegative, see Definition 7.3 below.

## 6 A Lagrangian formulation for Oldroyd B

The original idea of [4] was to use the standard generalized materials formalism, see [6], to derive a Lagrangian material that would become an Oldroyd B model in the Eulerian description, while satisfying the second principle by construction and having a variational structure. This did not succeed, possibly because standard generalized materials are not versatile enough. However, upon translating this work into our visco-elastic framework, we realized that the latter is versatile enough.

By convention, in the sequel, when we write an equality between an Eulerian quantity and a Lagrangian quantity, it will be meant at corresponding space-time points  $(x, t) = (\phi(X, t), t)$ . This keeps the length of formulas under control. Throughout the next two sections, all deformations will be incompressible, *i.e.*,  $\det F = 1$ .

Following [4], we start with the nonlinearly elastic neo-Hookean energy

$$\widehat{W}(F) = \frac{\mu}{2} \|F\|^2, \quad \mu > 0. \quad (19)$$

This energy is frame-indifferent and can be rewritten as  $\widetilde{W}(C) = \frac{\mu}{2} \text{tr } C$ , where  $C = F^T F$  is the usual strain or Cauchy-Green tensor.

We use this stored energy function to define the constitutive law of the Helmholtz free energy specific density for our material in the Lagrangian description  $\widehat{A}_m$  by

$$\widehat{A}_m(F, B_i) = \widetilde{W}(B_i C) = \frac{\mu}{2} B_i : C, \quad (20)$$

where  $B_i$  is a symmetric-valued, dimensionless internal variable. Then

$$\frac{\partial \widehat{A}_m}{\partial F}(F, B_i) = \mu F B_i \quad \text{and} \quad \frac{\partial \widehat{A}_m}{\partial B_i}(F, B_i) = \frac{\mu}{2} C. \quad (21)$$

Note that  $B_i$  is symmetric but  $B_i C$  is not. This form of free energy actually comes from a more general choice of internal variable  $F_i \in \mathbb{M}_3$  and more general choices of  $\widehat{W}$ , see [8] for details.

We take here the simplest kinematically viscous stress possible, which is that of the Newtonian fluid with viscosity  $\eta_s$ ,

$$\widehat{T}_{\text{Rd}}(F, H, B_i) = 2\eta_s \text{Sym}(HF^{-1})F^{-T}. \quad (22)$$

It is independent from the internal variable.

The natural decomposition of the first Piola-Kirchhoff stress (17) translates as a natural decomposition of the Cauchy stress  $\sigma = \sigma_s + \sigma_p$  into a solvent part

$$\sigma_s = T_{\text{Rd}}F^T = 2\eta_s d,$$

a Newtonian viscous stress by hypothesis (22), and a remainder

$$\sigma_p = \frac{\partial \widehat{A}_m}{\partial F}(F, B_i)F^T,$$

which will turn out to exactly correspond to the polymer part in decomposition (15). By (21), we have

$$\sigma_p = \mu F B_i F^T. \quad (23)$$

Let us define  $\Sigma_p = F^{-1}\sigma_p F^{-T}$  to serve as the second Piola-Kirchhoff stress part corresponding to  $\sigma_p$ . Clearly,

$$\Sigma_p = \mu B_i. \quad (24)$$

The numerical value of the modulus  $\mu$  will become irrelevant, but we keep it for reasons of dimensional homogeneity.

To complete the identification of the Oldroyd B fluid as a visco-elastic material with an internal variable, we just need to specify the flow rule. We thus choose

$$\widehat{K}(F, H, B_i) = -\frac{1}{\lambda_1} B_i + \frac{2\eta_p}{\mu\lambda_1} F^{-1} \text{Sym}(HF^{-1})F^{-T}. \quad (25)$$

In other words, the ordinary differential equation (16) for the internal variable here assumes the form

$$\frac{\partial B_i}{\partial t} = -\frac{1}{\lambda_1} B_i + \frac{2\eta_p}{\mu\lambda_1} F^{-1} \text{Sym}(HF^{-1})F^{-T}. \quad (26)$$

It is easy to check that with these choices, the resulting incompressible material is frame-indifferent. It clearly has a symmetric Cauchy stress constitutive law. It can be shown that the corresponding material is fluid, see [8].

**Proposition 6.1.** *The visco-elastic material defined by (20), (22), and (25) is the Oldroyd fluid with material constants  $\lambda_1$ ,  $\lambda_2$  and  $\eta$ .*

*Proof.* We have already seen that  $\sigma_s = 2\eta_s d$  by (22). Moreover, due to the neo-Hookean choice (20), equation (23) holds. Proposition 3.3 applied to (23) and (24) then shows that

$$\overset{\nabla}{\sigma}_p = \mu F \frac{\partial B_i}{\partial t} F^T. \quad (27)$$

We now substitute the ordinary differential equation (26) in the above relation, and obtain

$$\begin{aligned}\overset{\nabla}{\sigma}_p &= \mu F \left( -\frac{1}{\lambda_1} B_i + \frac{2\eta_p}{\mu\lambda_1} F^{-1} \text{Sym}(HF^{-1}) F^{-T} \right) F^T \\ &= -\frac{\mu}{\lambda_1} F B_i F^T + \frac{2\eta_p}{\lambda_1} \text{Sym}(HF^{-1}) \\ &= -\frac{1}{\lambda_1} \sigma_p + \frac{2\eta_p}{\lambda_1} d,\end{aligned}$$

or in other words

$$\sigma_p + \lambda_1 \overset{\nabla}{\sigma}_p = 2\eta_p d, \quad (28)$$

which is the polymer part of the Oldroyd B fluid constitutive differential equation. As already seen in Section 4, this is equivalent to

$$\sigma + \lambda_1 \overset{\nabla}{\sigma} = 2\eta(d + \lambda_2 \overset{\nabla}{d}),$$

with  $\sigma = \sigma_s + \sigma_p$ ,  $\eta = \eta_s + \eta_p$  and  $\lambda_2 = \frac{\lambda_1 \eta_s}{\eta_s + \eta_p}$ . This is the Oldroyd B fluid model with global viscosity  $\eta$  and relaxation times  $\lambda_1$  and  $\lambda_2$ .  $\square$

*Remarks 6.2.* We have shown that the specific instance of visco-elastic materials with internal variables described above satisfies the Oldroyd B equation in the Eulerian description. Conversely, given any Oldroyd B fluid, we can manufacture such a material that reproduces its behavior. Indeed, it suffices to take  $\eta_s = \frac{\lambda_2}{\lambda_1} \eta$ ,  $\eta_p = (1 - \frac{\lambda_2}{\lambda_1}) \eta$ , and any nonzero value for  $\mu$  in the Lagrangian model.

It is a posteriori interesting that the thermodynamically motivated decomposition of the first Piola-Kirchhoff stress (17) actually corresponds to the Cauchy stress decomposition for the Oldroyd B fluid (15), which initially looked like little more than an algebraic trick.

In [8], we rephrased the standard Oldroyd B model as an Eulerian model with an internal variable. We did this by choosing  $\xi = \sigma_p$  for the internal variable, having a free energy  $\hat{a}_m$  only function of  $\xi$ , and using the differential constitutive law (28) itself as a flow rule. We deemed this choice to have little physical significance at the time. A special case of it is however equivalent to the present, much more physically grounded, Lagrangian approach. Indeed,

$$\sigma_p = \mu F B_i F^T \text{ so that } B_i = \frac{1}{\mu} F^{-1} \xi F^{-T}.$$

In terms of Eulerian flow rule, we had the ordinary differential equation for  $\xi$

$$\overset{\nabla}{\xi} = \frac{1}{\lambda_1} (-\xi + 2\eta_p d). \quad (29)$$

**Proposition 6.3.** *The differential equations (26) and (29) are equivalent, and we have*

$$\hat{A}_m(F, B_i) = \hat{a}_m(\xi) \text{ with } \hat{a}_m(\xi) = \frac{1}{2} \text{tr } \xi.$$

*Proof.* Indeed, since  $\xi = \mu F B_i F^T$ , it follows from Proposition 3.3 that  $\overset{\nabla}{\xi} = \mu F \frac{\partial B_i}{\partial t} F^T$  so that substituting (26) therein, we obtain (29) and conversely. Moreover, we have

$$\hat{A}_m(F, B_i) = \frac{\mu}{2} B_i : C = \frac{\mu}{2} \text{tr}(B_i F^T F) = \frac{1}{2} \text{tr}(\mu F B_i F^T) = \frac{1}{2} \text{tr } \xi,$$

hence the equivalence in terms of free energies as well.  $\square$

With this specific choice of Eulerian free energy, we are thus recovering in Eulerian terms our Lagrangian model, the flow rule of which is tailored to precisely reproduce the Oldroyd B differential constitutive law.

## 7 The Oldroyd B fluid and the second principle of thermodynamics

Historically, Oldroyd introduced his A and B models without any concern for thermodynamics, see [9]. The issue of whether or not the Oldroyd B fluid is compatible with the second principle does not seem to be very prominent in the Oldroyd B literature, even though it is a continuum mechanics model that should obey the rules of thermodynamics.

This is not a trivial issue. Indeed, in [8], we showed that if the Oldroyd B model is considered as an Eulerian model with no internal variable, then the internal dissipation must be the naive one  $d_{\text{int}} = \sigma : d$ . In a series of very convincing numerical experiments, we also showed that, rather generically,  $d_{\text{int}}$  can become strictly negative after some time. The idea is to impose, via adapted forces, a periodically shaking velocity field and compute the corresponding evolution of the Cauchy stress at one rest point in the fluid. Because the Oldroyd B fluid involves a differential equation in time, the stress tensor lags behind the stretching tensor in some sense, and ends up in opposition of phase with the latter, hence the strictly negative inner product, see [8] for details. This is not a mathematical proof, but the numerics are fairly simple and standard, and the numerical error should be minimal whereas the second principle violation is very large.

It turns out that the equivalence between our Eulerian formulation with the internal variable  $\xi = \sigma_p$  of [8] and our present Lagrangian formulation extends to internal dissipation and second principle issues. Let us start with the Lagrangian formulation.

**Proposition 7.1.** *The constitutive law for the Lagrangian internal dissipation is*

$$\widehat{D}_{\text{int}}(F, H, B_i) = 2\eta_s \|\text{Sym}(HF^{-1})\|^2 + \frac{\mu}{2\lambda_1} B_i : C. \quad (30)$$

*Proof.* We compute all the terms in formula (18). Consider first the Newtonian fluid term,

$$\begin{aligned} \widehat{T}_{\text{Rd}}(F, H, B_i) : H &= 2\eta_s (\text{Sym}(HF^{-1})F^{-T}) : H = 2\eta_s \text{tr}(F^{-1} \text{Sym}(HF^{-1})H) \\ &= 2\eta_s \text{tr}(\text{Sym}(HF^{-1})HF^{-1}) = 2\eta_s \text{Sym}(HF^{-1}) : (HF^{-1}) \\ &= 2\eta_s \|\text{Sym}(HF^{-1})\|^2, \end{aligned}$$

which is to be expected from the corresponding Eulerian expression. Next we look at the dissipation coming from the internal variable. It follows from (21) that

$$\begin{aligned} -\frac{\partial \widehat{A}_m}{\partial B_i}(F, B_i) : \widehat{K}(F, H, B_i) &= -\frac{\mu}{2} C : \left( -\frac{1}{\lambda_1} B_i + \frac{2\eta_p}{\mu\lambda_1} F^{-1} \text{Sym}(HF^{-1})F^{-T} \right) \\ &= \frac{\mu}{2\lambda_1} C : B_i - \frac{\eta_p}{\lambda_1} L(H), \end{aligned}$$

where  $L(H) = C : (F^{-1} \text{Sym}(HF^{-1})F^{-T})$ . This linear form vanishes since

$$\begin{aligned} L(H) &= \text{tr}(CF^{-1} \text{Sym}(HF^{-1})F^{-T}) \\ &= \text{tr}(F^T \text{Sym}(HF^{-1})F^{-T}) = \text{tr}(\text{Sym}(HF^{-1})) = 0. \end{aligned}$$

Indeed,  $\text{Sym}(HF^{-1}) = d$  and  $\text{tr} d = 0$  by incompressibility. This completes the proof of equation (30).  $\square$

In order to compare this result to the Eulerian dissipation considered in [8], we recall the notation used therein for the flow rule in the form  $\dot{\xi} = \widehat{k}(h, \xi)$  with

$$\widehat{k}(h, \xi) = h\xi + \xi h^T + \frac{1}{\lambda_1}(-\xi + 2\eta_p d), \quad (31)$$

which is a rewriting of (29), and the internal dissipation

$$\widehat{d}_{\text{int}}(h, \xi) = \sigma : d - \frac{\partial \widehat{a}_m}{\partial \xi}(\xi) : \widehat{k}(h, \xi) \text{ with } \sigma = 2\nu_s d + \xi. \quad (32)$$

The two Lagrangian and Eulerian approaches are here again equivalent for the choice  $\widehat{a}_m(\xi) = \frac{1}{2} \text{tr} \xi$ .

**Proposition 7.2.** *At all corresponding values of the thermodynamic variables, we have*

$$\widehat{D}_{\text{int}}(F, H, B_i) = \widehat{d}_{\text{int}}(h, \xi).$$

*Proof.* We have here  $\frac{\partial \widehat{a}_m}{\partial \xi}(\xi) = \frac{1}{2}I$ , so that

$$\widehat{d}_{\text{int}}(h, \xi) = 2\eta_s d : d + \xi : d - \frac{1}{2} \text{tr}(\widehat{k}(h, \xi)),$$

with

$$\text{tr}(\widehat{k}(h, \xi)) = 2\xi : h - \frac{1}{\lambda_1} \text{tr} \xi = 2\xi : d - \frac{1}{\lambda_1} \text{tr} \xi,$$

by incompressibility and the symmetry of  $\xi$ . Therefore,

$$\widehat{d}_{\text{int}}(h, \xi) = 2\eta_s \|d\|^2 + \frac{1}{2\lambda_1} \text{tr} \xi.$$

We have already noticed that  $\text{tr} \xi = \text{tr} \sigma_p = \mu B_i : C$ , from which the result follows.  $\square$

Now Proposition 5.11 of [8] states that there is no free energy  $\widehat{a}_m$  that can make the Eulerian dissipation nonnegative for all values of  $d \in \text{sl}(3)$  and  $\xi \in \text{Sym}_3$ . In particular, the choice  $\widehat{a}_m(\xi) = \frac{1}{2} \text{tr} \xi$  does not yield a nonnegative dissipation in the sense of Coleman-Noll, and the same thus holds true for our Lagrangian formulation.

We can however lower our expectations and wonder whether if we start with some initial condition that corresponds to a nonnegative dissipation, then the dissipation would remain nonnegative for all subsequent times, given an admissible deformation  $\phi$ . This makes for an acceptable version of the second principle for the Oldroyd B fluid, and more generally for materials with internal variables, albeit not strictly speaking that of Coleman and Noll as revisited in

[8], where we required the dissipation to be nonnegative for all initial conditions of the internal variables.

To be more precise, given some reference configuration  $\Omega \subset \mathbb{R}^3$ , we say that a deformation  $\phi: \bar{\Omega} \times \mathbb{R} \rightarrow \mathbb{R}^3$  is admissible if it is of class  $C^1$ ,  $\det \nabla_X \phi(X, t) = 1$  for all  $X \in \Omega$  and  $t \in \mathbb{R}_+$ , and for all  $t \in \mathbb{R}_+$ ,  $\phi(\cdot, t)$  is a diffeomorphism between  $\bar{\Omega}$  and  $\phi(\Omega, t)$ .

**Definition 7.3.** *We will say that the second principle is satisfied for an initial value of the internal variable if, given any admissible deformation, the corresponding dissipation is nonnegative for all  $t \geq 0$ . If the set  $\mathcal{C}$  of such initial values is nonempty, we say that the second principle is conditionally satisfied under condition  $\mathcal{C}$ .*

The difference with the Coleman-Noll approach of [8] is that we do not expect this to hold for any initial value of the internal variable and thus the constitutive law (30) for the internal dissipation is not required to only take nonnegative values for all possible arguments.

However, if we examine the proof of [8] in this new light, we can see that there is little change in the conclusions listed in Section 5. It is still necessary that  $\hat{A}_m$  does not depend on  $H$  when  $B_i \in \mathcal{C}$ , the decomposition of the first Piola-Kirchhoff stress is unchanged, and the internal dissipation (18) must be nonnegative for all  $F, H$ , and all  $B_i \in \mathcal{C}$ .

Conversely, if the condition set  $\mathcal{C}$  is invariant by the flow rule, then the above conditions are sufficient for the conditional second principle. These conclusions, appropriately generalized, actually hold true in the much wider thermo-viscoelastic context of [8]. We will see however that such an invariance does not hold in the case of the Oldroyd B fluid, *cf.* Proposition 7.6. The issue of whether or not the conditional second principle holds for Oldroyd B is thus somewhat subtler than this.

Since the above general statements are not proved here, we proceed in the Oldroyd B case without referring to them in the sequel, which is thus self-contained.

In the specific case of our Lagrangian formulation for the Oldroyd B fluid, we already have a free energy  $\hat{A}_m$  that does not depend on  $H$ . The constitutive law for the internal dissipation (30) clearly does not take nonnegative values for all  $F \in \mathbb{M}_3^+$ ,  $H \in \mathbb{M}_3$ , and  $B_i \in \text{Sym}_3$  without restrictions on  $B_i$ . Let us proceed to identify the set  $\mathcal{C}$  of initial conditions that will ensure that the conditional second principle holds, if any.

We need the following standard lemma.

**Lemma 7.4.** *Let  $B$  and  $C$  be two symmetric positive semi-definite  $n \times n$  matrices. Then we have*

$$B : C \geq n(\det B)^{\frac{1}{n}}(\det C)^{\frac{1}{n}}. \quad (33)$$

*In particular,  $B : C \geq 0$ .*

*Proof.* We first remark that

$$B : C = \text{tr}(BC) = \text{tr}(B^{\frac{1}{2}}B^{\frac{1}{2}}C^{\frac{1}{2}}C^{\frac{1}{2}}) = \text{tr}(C^{\frac{1}{2}}B^{\frac{1}{2}}B^{\frac{1}{2}}C^{\frac{1}{2}}) = \|B^{\frac{1}{2}}C^{\frac{1}{2}}\|^2.$$

Let  $M = B^{\frac{1}{2}}C^{\frac{1}{2}}$  and  $M_i \in \mathbb{R}^n$  be its column vectors. We have

$$B : C = \sum_{i=1}^n \|M_i\|^2 \geq n \left( \prod_{i=1}^n \|M_i\|^2 \right)^{\frac{1}{n}} \geq n(\det M)^{\frac{2}{n}} = n(\det B)^{\frac{1}{n}}(\det C)^{\frac{1}{n}},$$

by the inequality between the arithmetic and geometric means and by the Hadamard inequality.  $\square$

Note that inequality (33) is sharp.

**Proposition 7.5.** *It is necessary that  $B_i(X, 0)$  be positive semidefinite for all  $X \in \Omega$  for the second principle to hold conditionally.*

*Proof.* Here and in the ensuing proofs, we fix a Lagrangian point  $X_0 \in \Omega$  throughout and write  $B_i(X_0, t) = B_i(t)$  and so on, since  $X$  only plays the role of a parameter and the sole relevant variable is the time variable  $t$ .

Let us assume that  $B_i(0)$  is such that the second principle holds conditionally. Let  $F_0 \in \text{SL}(3)$  be arbitrary and take  $\phi(X, t) = F_0 X$ . This choice is admissible with  $C(t) = C_0 = F_0^T F_0$  and  $H(t) = 0$  for all  $t$ . The internal dissipation at  $t = 0$  is thus  $D_{\text{int}}(0) = \frac{\mu}{2\lambda_1} B_i(0) : C_0$  by (30). The conditional second principle then implies that

$$B_i(0) : C_0 \geq 0,$$

for all  $C_0 \in \text{Sym}_3^+ \cap \text{SL}(3)$ . Multiplying by any positive factor and by continuity, it follows that we must have  $B_i(0) : C_0 \geq 0$  for all  $C_0 \in \overline{\text{Sym}_3^+}$ . Diagonalizing  $B_i(0)$  in the form  $B_i(0) = Q\Delta Q^T$  with  $Q \in \text{SO}(3)$  and  $\Delta$  diagonal, we see that  $\Delta : C_0 \geq 0$  for all  $C_0 \in \overline{\text{Sym}_3^+}$ . Choosing  $C_0 = \text{diag}(1, 0, 0)$ , we obtain that  $\Delta_{11} \geq 0$ , and similarly for the other eigenvalues of  $B_i(0)$ .  $\square$

We thus have that  $\mathcal{C} \subset \overline{\text{Sym}_3^+}$ . Let us remark that  $\overline{\text{Sym}_3^+}$  is precisely the set of  $B_i \in \overline{\text{Sym}_3}$  such that  $\widehat{D}_{\text{int}}(F, H, B_i) \geq 0$  for all  $F \in \mathbb{M}_3^+$  and  $H \in \mathbb{M}_3$ . Indeed, if  $B_i \in \overline{\text{Sym}_3^+}$ , then  $B_i : C \geq 0$  for all  $C \in \text{Sym}_3^+$  by Lemma 7.4. If the set  $\overline{\text{Sym}_3^+}$  was invariant by the flow rule, then the internal dissipation would be nonnegative by the previous remark. This is not the case in general.

**Proposition 7.6.** *If  $\eta_p > 0$ , there is an admissible deformation such that  $B_i(X, 0) \in \overline{\text{Sym}_3^+}$  but  $B_i(X, t) \notin \overline{\text{Sym}_3^+}$  for some  $t > 0$ . If  $\eta_p = 0$ , then  $B_i(X, 0) \in \overline{\text{Sym}_3^+}$  implies that  $B_i(X, t) \in \overline{\text{Sym}_3^+}$  for all  $t \geq 0$ .*

*Proof.* From now on, we denote time differentiation with a prime since  $t$  is the only relevant variable. Assume first that  $\eta_p > 0$ . We take  $\phi(X, t) = F(t)X$  with  $F(t) = \text{diag}(e^t, e^{-t}, 1)$ . This is obviously an admissible deformation, which corresponds to a steady Eulerian flow  $h(t) = d(t) = \text{diag}(1, -1, 0)$ . Assuming  $\lambda_1 = 1$  without loss of generality, equation (26) becomes in this case,

$$(e^t B_i(t))' = \frac{2\eta_p}{\mu} \text{diag}(e^{-t}, -e^{3t}, 0).$$

Integrating this between 0 and  $t$ , we obtain

$$B_i(t) = e^{-t} B_i(0) + \frac{2\eta_p}{\mu} \text{diag}\left(e^{-t} - e^{-2t}, \frac{1}{3}(e^{-t} - e^{2t}), 0\right).$$



If  $\eta_p > 0$ , for any  $B_i(0)$  including all positive semidefinite ones, the smallest eigenvalue of  $B_i(t)$  thus goes to  $-\infty$  when  $t \rightarrow +\infty$ . Consequently,  $B_i(t)$  exits  $\text{Sym}_3^+$  in finite time.

If, on the contrary,  $\eta_p = 0$ , then for all admissible deformations,  $B_i(t) = e^{-\frac{t}{\lambda_1}} B_i(0)$  for all  $t$ . Therefore, the flow rule obviously preserves positive semi-definiteness in this particular case.  $\square$

See also another example, albeit a numerical one, of evolution that does not preserve positive semi-definiteness in Section 8.3, Figure 2.

It is quite remarkable that, when  $\eta_p > 0$  and in spite of Proposition 7.6, the conditional second principle still holds.

**Proposition 7.7.** *For any  $\eta_p \geq 0$ , the Oldroyd B fluid satisfies the second principle of thermodynamics conditionally if and only if  $B_i(X, 0)$  is positive semi-definite for all  $X$ .*

*Proof.* The necessity was proved in Proposition 7.5. Let us thus assume that  $B_i(0)$  is positive semi-definite. It is well known that

$$C'(t) = 2F^T(t)d(t)F(t).$$

Let us rewrite the second term in the flow rule (25), which only depends on  $t$ , with this remark:

$$F^{-1} \text{Sym}(HF^{-1})F^{-T} = \frac{1}{2}F^{-1}F^{-T}C'F^{-1}F^{-T} = \frac{1}{2}C^{-1}C'C^{-1} = -\frac{1}{2}(C^{-1})'.$$

The ordinary differential equation (26) for  $B_i$  is linear with continuous right-hand side, therefore the Cauchy problem is well-posed on  $\mathbb{R}_+$  for any initial value  $B_i(0) \in \text{Sym}_3$ . Moreover, it has constant coefficients, thus the Duhamel formula provides an expression for  $B_i$ ,

$$B_i(t) = e^{-\frac{t}{\lambda_1}} B_i(0) - \alpha e^{-\frac{t}{\lambda_1}} \int_0^t e^{\frac{s}{\lambda_1}} (C^{-1})'(s) ds,$$

where  $\alpha = \frac{\eta_p}{\mu\lambda_1} \geq 0$ . Integrating the second term by parts, we obtain

$$e^{\frac{t}{\lambda_1}} B_i(t) = B_i(0) + \alpha \left( C^{-1}(0) - e^{\frac{t}{\lambda_1}} C^{-1}(t) + \frac{1}{\lambda_1} \int_0^t e^{\frac{s}{\lambda_1}} C^{-1}(s) ds \right).$$

Consequently,

$$\begin{aligned} e^{\frac{t}{\lambda_1}} B_i(t) : C(t) &= B_i(0) : C(t) + \alpha C^{-1}(0) : C(t) - 3\alpha e^{\frac{t}{\lambda_1}} \\ &\quad + \frac{\alpha}{\lambda_1} \int_0^t e^{\frac{s}{\lambda_1}} C^{-1}(s) : C(t) ds \\ &= B_i(0) : C(t) + \alpha (C^{-1}(0) : C(t) - 3) \\ &\quad + \frac{\alpha}{\lambda_1} \int_0^t e^{\frac{s}{\lambda_1}} (C^{-1}(s) : C(t) - 3) ds. \end{aligned}$$

By Lemma 7.4, we first have  $B_i(0) : C(t) \geq 0$  for all  $t \geq 0$  since  $B_i(0)$  and  $C(t)$  are both symmetric positive semi-definite. Secondly,  $C^{-1}(s)$  and  $C(t)$  are symmetric positive-definite and belong to  $\text{SL}(3)$ , by incompressibility. Therefore, also by Lemma 7.4, we have  $C^{-1}(s) : C(t) \geq 3$  for all  $s \geq 0$  and consequently  $B_i(t) : C(t) \geq 0$  for all  $t \geq 0$ . The nonnegativity of the internal dissipation then follows from Proposition 7.1.  $\square$

*Remark 7.8.* In the case of the example of Proposition 7.6, we obtain that

$$B_i(t) : C(t) = e^{-t} B_i(0) : C(t) + \frac{2\eta_p}{\mu} \left( e^t + \frac{1}{3} e^{-3t} - \frac{4}{3} \right) \geq 0$$

for all  $t$ .

Note that when  $\eta_p = 0$ , the result of Proposition 7.7 is trivial in view of Lemma 7.4 and the second part of Proposition 7.6.

The following is then a direct consequence of Proposition 7.2.

**Corollary 7.9.** *The Eulerian dissipation  $d_{\text{int}}$  remains nonnegative for all times and all given velocity fields if and only if  $\sigma_p(0)$  is positive semi-definite.*

Of course, the naive dissipation  $\sigma : d$  will still change sign for some velocity fields.

The question now is whether it is legitimate to only consider initial values for the internal variable that ensure the conditional second principle. For a general model, the physical meaning of a given internal variable may be unclear. It can be a question of principle to only pick such initial conditions, if they exist. In the particular case of the Oldroyd B fluid, we actually have

$$B_i(0) = \frac{1}{\mu} \Sigma_p(0) = \frac{1}{\mu} F^{-1}(0) \sigma_p(0) F^{-T}(0),$$

or directly in the Eulerian description

$$\xi(0) = \sigma_p(0).$$

Now of course, the polymer stress is not entirely well defined from the constitutive point of view, unless an initial value is chosen for it. Furthermore, since the model is incompressible, the actual physical Cauchy stress is of the form

$$\sigma(0) = 2\eta_s d(0) + \sigma_p(0) + p(0)I,$$

where  $p$  is the indeterminate pressure (that we have ignored up to now). So the quantity that is physically meaningful is  $\sigma_p(0) + p(0)I = \sigma(0) - 2\eta_s d(0)$ . From the constitutive point of view, we are at liberty to incorporate some of the indeterminate pressure into  $\sigma_p(0)$  in such a way that it becomes positive semi-definite without changing the right-hand side, and thus consider the second principle to be satisfied in all situations. This also holds true in the Eulerian formulation in view of Corollary 7.9.

If we were considering an initial-boundary value problem that was well-posed, then the pressure would be determined by the problem data and the above liberty would no longer be available.

*Remark 7.10.* A natural question is whether or not the present formulation of the Oldroyd B model admits a dissipation potential in the sense of standard generalized materials, [4]-[6], or in the somewhat different sense introduced in [8].

In the case  $\eta_p > 0$ , the answer is clearly negative, otherwise the second principle would hold unconditionally. The case  $\eta_p = 0$  can be considered as unconditional if we restrict the internal variable to  $\text{Sym}_3^+$ , which is then invariant

under the flow rule. This is a convex set and the function  $\widehat{P}_{\text{diss}}: \mathbb{M}_3^+ \times \mathbb{M}_3 \times \overline{\text{Sym}}_3^+ \times \overline{\text{Sym}}_3^+ \rightarrow \mathbb{R}$  defined by

$$\widehat{P}_{\text{diss}}(F, H, B_i, \Lambda) = \eta_s \|\text{Sym}(HF^{-1})\|^2 + \frac{1}{\lambda_1} B_i : \Lambda.$$

is clearly convex with respect to  $(H, \Lambda)$ , takes nonnegative values, and is such that  $\widehat{P}_{\text{diss}}(F, 0, B_i, 0) = 0$ . This function is a dissipation potential for  $\widehat{T}_{\text{Rd}}^1$  and  $\widehat{K}$ , in a slightly generalized sense compared to [8] allowing for more flexibility in the arguments of the potential, namely that we have  $\widehat{T}_{\text{Rd}}(F, H, B_i) = \frac{\partial \widehat{P}_{\text{diss}}}{\partial H}(F, H, B_i, \frac{\partial \widehat{A}_m}{\partial B_i}(F, B_i))$  and  $\widehat{K}(F, H, B_i) = -\frac{\partial \widehat{P}_{\text{diss}}}{\partial \Lambda}(F, H, B_i, \frac{\partial \widehat{A}_m}{\partial B_i}(F, B_i))$ , thus yielding a nonnegative dissipation. Of course, this remark adds very little insight into this particular situation compared to our direct approach.

## 8 A few variants of the Oldroyd B model

In this section, we develop a few generalizations of the Oldroyd B model based on Lagrangian formulations and discuss their relationship with the second principle.

### 8.1 Compressible Oldroyd B

Before we start with the modeling, let us draw a small list of useful identities that relate  $C = F^T F$  and  $d$ , the first of which was already noted earlier, namely that

$$\frac{\partial C}{\partial t} = 2F^T dF, \quad \frac{\partial C^{-1}}{\partial t} = -2F^{-1} dF^{-T}, \quad (34)$$

and

$$\text{tr}(d) = \frac{1}{2} C^{-1} : \frac{\partial C}{\partial t} = -\frac{1}{2} \frac{\partial C^{-1}}{\partial t} : C, \quad (35)$$

at all corresponding Lagrangian and Eulerian space-time points. Moreover,

$$\frac{\partial J}{\partial t} = J \text{tr}(d). \quad (36)$$

Let us now see what kind of model can be obtained by removing the incompressibility assumption. In the compressible case, there is no indeterminate pressure, and the whole Cauchy stress, including the polymer stress, will be entirely determined by the constitutive laws plus initial conditions, as opposed to what happens in the incompressible case.

We start with a free energy density of the form

$$\widehat{A}_m(F, B_i) = \widehat{w}(J) + \frac{\mu}{2} B_i : C, \text{ with } J = \det F, \quad (37)$$

with  $\mu > 0$ . In addition to the neo-Hookean term featuring the internal variable, there is an elastic fluid term  $\widehat{w}: \mathbb{R}_+^* \rightarrow \mathbb{R}$ . Differentiating (37) with respect to  $F$ , we obtain

$$\frac{\partial \widehat{A}_m}{\partial F}(F, B_i) = \widehat{w}'(J) \text{cof } F + \mu F B_i.$$

---

<sup>1</sup>The dissipation potential for the compressible Newtonian fluid in Lagrangian form we wrote in Remark 3.9 of [8] is incorrect and should read  $\widehat{P}_{\text{diss}}(F, H) = \nu \det F \|\text{Sym}(HF^{-1})\|^2$ .

According to equation (17), we thus obtain a standard decomposition of the first Piola-Kirchhoff stress as

$$\widehat{T}_R(F, H, B_i) = \widehat{T}_{Rd}(F, H) + \widehat{w}'(J) \operatorname{cof} F + \mu F B_i,$$

and of the Cauchy stress as

$$\widehat{\sigma}(F, H, B_i) = \frac{1}{J} \widehat{T}_{Rd}(F, H) F^T + \widehat{w}'(J) I + \frac{\mu}{J} F B_i F^T.$$

Similarly as before, we let  $\sigma = \sigma_s + \sigma_p$  with

$$\widehat{\sigma}_s(F, H) = \frac{1}{J} \widehat{T}_{Rd}(F, H) F^T \quad \text{and} \quad \widehat{\sigma}_p(F, B_i) = \widehat{w}'(J) I + \frac{\mu}{J} F B_i F^T. \quad (38)$$

Without loss of generality, we assume the reference configuration to be homogeneous and with mass density equal to 1. In the spirit of the Oldroyd B fluid, we assume that the solvent is a compressible Newtonian fluid, the stress of which assumes the Eulerian form

$$\sigma_s = 2\bar{\eta}_s(\rho) d + \bar{\lambda}_s(\rho) \operatorname{tr}(d) I,$$

where  $\rho = \frac{1}{J}$  is the Eulerian mass density, and the viscosities  $\bar{\eta}_s, \bar{\lambda}_s$  are given functions defined on  $\mathbb{R}_+^*$  such that  $\bar{\eta}_s \geq 0$  and  $2\bar{\eta}_s + 3\bar{\lambda}_s \geq 0$  in order for the viscous Newtonian dissipation to be nonnegative for all  $d$ . In Lagrangian terms, this reads

$$\widehat{T}_{Rd}(F, H) = (2\eta_s(J) \operatorname{Sym}(HF^{-1}) + \lambda_s(J) \operatorname{tr}(HF^{-1}) I) \operatorname{cof} F, \quad (39)$$

where  $\eta_s(J) = \bar{\eta}_s(J^{-1})$  and  $\lambda_s(J) = \bar{\lambda}_s(J^{-1})$ .

We again define a polymer second Piola-Kirchhoff stress by  $\Sigma_p = JF^{-1}\sigma_p F^{-T}$ . Letting  $\widehat{z}(J) = J\widehat{w}'(J)$ , we obtain from (38)

$$\widehat{\Sigma}_p(F, B_i) = \widehat{z}(J) C^{-1} + \mu B_i. \quad (40)$$

Of course, in the compressible case, we expect the Truesdell derivative to play a leading role.

**Proposition 8.1.** *We have*

$$\overset{\circ}{\sigma}_p = \widehat{z}'(J) \operatorname{tr}(d) I - \frac{2\widehat{z}(J)}{J} d + \frac{\mu}{J} F \frac{\partial B_i}{\partial t} F^T. \quad (41)$$

*Proof.* From (40) and (34)–(36), we deduce that

$$\begin{aligned} \frac{\partial \Sigma_p}{\partial t} &= \frac{\partial}{\partial t} (\widehat{z}(J) C^{-1}) + \mu \frac{\partial B_i}{\partial t} \\ &= \widehat{z}'(J) J \operatorname{tr}(d) C^{-1} - 2\widehat{z}(J) F^{-1} d F^{-T} + \mu \frac{\partial B_i}{\partial t}. \end{aligned}$$

We then appeal to Proposition 3.3 to conclude.  $\square$

In order to derive an Oldroyd B-like equation for  $\sigma_p$ , we choose three functions  $\lambda_1, \eta_*$  and  $\lambda_*$  from  $\mathbb{R}_+^*$  to  $\mathbb{R}$ , with  $\lambda_1$  strictly positive, then consider the following flow rule

$$\widehat{K}(F, H, B_i) = -\frac{1}{\lambda_1(J)} B_i - \eta_*(J) (C^{-1})' + \frac{\lambda_*(J)}{2} (C^{-1} : C') C^{-1}. \quad (42)$$

Here,  $\lambda_1$  plays again the role of a relaxation characteristic time and  $\eta_*$  and  $\lambda_*$  somewhat that of Lamé viscosity coefficients. The notations  $(C^{-1})'$  and  $C'$  are shorthand for  $-2F^{-1} \text{Sym}(HF^{-1})F^{-T}$  and  $2F^T \text{Sym}(HF^{-1})F$  respectively, *viz.* identities (34).

**Proposition 8.2.** *The polymer stress satisfies the equation*

$$\sigma_p + \bar{\lambda}_1(\rho) \overset{\circ}{\sigma}_p = 2\bar{\eta}_p(\rho)d + \bar{\lambda}_p(\rho) \text{tr}(d)I - \bar{p}(\rho)I, \quad (43)$$

where  $\bar{\lambda}_1(\rho) = \lambda_1(\rho^{-1})$ ,  $\bar{\eta}_p(\rho) = \eta_p(\rho^{-1})$ ,  $\bar{\lambda}_p(\rho) = \lambda_p(\rho^{-1})$ , with

$$\eta_p(J) = \frac{\lambda_1(J)}{J} (\mu\eta_*(J) - \widehat{z}(J)), \quad (44)$$

$$\lambda_p(J) = \frac{\lambda_1(J)}{J} (\mu\lambda_*(J) + J\widehat{z}'(J)), \quad (45)$$

and finally,  $\bar{p}(\rho) = -\rho\widehat{z}(\rho^{-1})$ .

*Proof.* Let us rewrite the flow rule (42) by mixing Eulerian and Lagrangian quantities, using identities (34)–(36) and omitting the variable  $J$  as argument in the various functions for brevity,

$$\frac{\partial B_i}{\partial t} = -\frac{1}{\lambda_1} B_i + 2\eta_* F^{-1} d F^{-T} + \lambda_* \text{tr}(d) C^{-1},$$

and replace it in equation (41), which yields

$$\overset{\circ}{\sigma}_p = \widehat{z}' \text{tr}(d)I - \frac{2\widehat{z}}{J} d - \frac{1}{\lambda_1} \frac{\mu}{J} F B_i F^T + \frac{\mu}{J} (2\eta_* d + \lambda_* \text{tr}(d)I),$$

hence the result since  $\sigma_p = \frac{1}{J} (\widehat{z}I + \mu F B_i F^T)$ .  $\square$

*Remarks 8.3.* The right-hand side of (43) assumes the form of a compressible Newtonian Cauchy stress with viscosities  $\bar{\eta}_p(\rho)$  and  $\bar{\lambda}_p(\rho)$ , and elastic pressure  $\bar{p}(\rho)$ . We consider this to be a compressible generalization of the expression of the Oldroyd B model in terms of  $\sigma_p$ , *i.e.*, the second part of (15). The polymer viscosities and elastic pressure are arbitrary functions at this point.

The equation can be equivalently rewritten in terms of  $\sigma$  as (omitting  $\rho$  for brevity)

$$\sigma + \bar{\lambda}_1 \overset{\circ}{\sigma} = 2\bar{\eta}d + \bar{\lambda} \text{tr}(d)I - \bar{p}I + \bar{\lambda}_1 \overbrace{(2\bar{\eta}_s d + \bar{\lambda}_s \text{tr}(d)I)}^{\circ}, \quad (46)$$

with  $\bar{\eta}(\rho) = \bar{\eta}_s(\rho) + \bar{\eta}_p(\rho)$  and  $\bar{\lambda}(\rho) = \bar{\lambda}_s(\rho) + \bar{\lambda}_p(\rho)$ . Equation (46) is in turn a compressible generalization of equation (14).

Let us now turn to second principle considerations for this compressible model.

**Proposition 8.4.** *The constitutive law for the internal dissipation is given by*

$$\begin{aligned} \widehat{D}_{\text{int}}(F, H, B_i) = & J \left( 2\eta_s(J) \|\text{Sym}(HF^{-1})\|^2 + \lambda_s(J) (\text{tr}(HF^{-1}))^2 \right) \\ & - \frac{\mu}{2} (2\eta_*(J) + 3\lambda_*(J)) \text{tr}(HF^{-1}) + \frac{\mu}{2\lambda_1(J)} B_i : C \end{aligned} \quad (47)$$

in Lagrangian form.

*Proof.* This follows directly from equation (18), relation (39), the definition (42) and the fact that  $\frac{\partial \widehat{A}_m}{\partial B_i} = \frac{\mu}{2}C$  as before.  $\square$

Note that the only viscosity coefficients that play a role in the constitutive law for the internal dissipation are  $\eta_s$ ,  $\lambda_s$ ,  $\eta_*$ , and  $\lambda_*$ .

Let us first write a necessary condition.

**Proposition 8.5.** *If the compressible Oldroyd B fluid satisfies the second principle of thermodynamics conditionally, then  $B_i(X, 0)$  is positive semi-definite.*

*Proof.* Same proof as Proposition 7.5.  $\square$

*Remarks 8.6.* The question now is is this condition sufficient? We already know that it is sufficient for incompressible deformations, by the results of Section 7. For a general compressible deformation however, the third term in equation (47) is clearly problematic. Consider for instance  $\phi(X, t) = e^{\alpha t}F_0X$ , with  $F_0 \in \mathbb{M}_3^+$ , for which  $HF^{-1} = \alpha I$ . For  $B_i(0) = 0$ , the initial dissipation is

$$D_{\text{int}}(0) = 3J(2\eta_s(J) + 3\lambda_s(J))\alpha^2 - \frac{3\mu}{2}(2\eta_*(J) + 3\lambda_*(J))\alpha,$$

with  $J = \det F_0$ . For any  $F_0$  such that  $2\eta_*(J) + 3\lambda_*(J) \neq 0$ , there are small values of  $\alpha$  that make it strictly negative. The second principle thus cannot be satisfied with the condition  $B_i(0)$  positive semi-definite in this case.

If we assume  $2\eta_*(J) + 3\lambda_*(J) = 0$  for all  $J$ , this problem disappears. This assumption is thus made from now on.

We have the following negative result in the case of constant coefficients.

**Proposition 8.7.** *Assume that  $\lambda_1$ ,  $\eta_s$ ,  $\lambda_s$  and  $\eta_*$  are constant functions and  $\lambda_* = -\frac{2}{3}\eta_* \neq 0$ . Then the compressible Oldroyd B fluid does not satisfy the second principle of thermodynamics conditionally for any  $B_i(X, 0)$ .*

*Proof.* Without loss of generality, we may assume that  $\lambda_1 = 1$  by rescaling the time variable. The ordinary differential equation for  $B_i$  thus assumes the form

$$\frac{\partial B_i}{\partial t} = -B_i - \eta_* \left( (C^{-1})' + \frac{1}{3}(C^{-1} : C')C^{-1} \right).$$

Now it is easily checked that

$$\frac{1}{3}(C^{-1} : C') = \frac{1}{3}(\ln(\det C))' = (\ln((\det C)^{1/3}))'.$$

This suggests a decoupling of the form

$$f(t) = (\det C(t))^{-1/3} \text{ and } M(t) = \frac{1}{f(t)}C^{-1}(t),$$

with  $f(t) \in \mathbb{R}_+^*$ ,  $M(t) \in \text{Sym}_3^+ \cap \text{SL}(3)$ , where  $f$  and  $M$  are independent of each other. This considerably simplifies the equation as

$$\frac{\partial B_i}{\partial t} = -B_i - \eta_* f M'.$$

By Duhamel's formula, it follows that

$$B_i(t) = e^{-t}B_i(0) - \eta_*e^{-t} \int_0^t e^s f(s)M'(s) ds,$$

and the internal dissipation becomes

$$\begin{aligned} D_{\text{int}}(t) &= f(t)^{-3/2}(2\eta_s\|d(t)\|^2 + \lambda_s(\text{tr } d(t))^2) \\ &\quad - \frac{\mu\eta_*e^{-t}}{2f(t)} \int_0^t e^s f(s)M'(s) : M^{-1}(t) ds + \frac{\mu e^{-t}}{2}B_i(0) : C(t). \end{aligned} \quad (48)$$

We are assuming that  $\eta_* \neq 0$ . There are two cases depending on the sign of  $\eta_*$ . We start with the simplest case,  $\eta_* < 0$ . A simple incompressible deformation  $f(t) = 1$  will be sufficient. Let  $\alpha \in \mathbb{R}$  and define

$$F(t) = \text{diag}(e^{-\alpha t/2}, e^{\alpha t/2}, 1).$$

Let us compute all the relevant quantities for the deformation  $\phi(X, t) = F(t)X$ :

$$\begin{aligned} H(t) &= \frac{\alpha}{2} \text{diag}(-e^{-\alpha t/2}, e^{\alpha t/2}, 0), \quad d(t) = \frac{\alpha}{2} \text{diag}(-1, 1, 0), \\ C(t) &= M^{-1}(t) = \text{diag}(e^{-\alpha t}, e^{\alpha t}, 1), \quad M'(s) = \alpha \text{diag}(e^{\alpha s}, -e^{-\alpha s}, 0). \end{aligned}$$

With these formulas in hand, we obtain that for  $\alpha \neq \pm 1$ ,

$$e^{-t} \int_0^t e^s f(s)M'(s) : M^{-1}(t) ds = \frac{2\alpha^2}{\alpha^2 - 1} + \alpha \left( \frac{e^{(\alpha-1)t}}{1-\alpha} - \frac{e^{-(\alpha+1)t}}{\alpha+1} \right),$$

and

$$\begin{aligned} 2\eta_s\|d(t)\|^2 + \lambda_s(\text{tr } d(t))^2 &= \eta_s\alpha^2, \\ e^{-t}B_i(0) : C(t) &= B_i(0) : \text{diag}(e^{-(\alpha+1)t}, e^{(\alpha-1)t}, e^{-t}). \end{aligned}$$

We now choose  $|\alpha| < 1$  so that

$$D_{\text{int}}(t) \rightarrow \alpha^2 \left( \eta_s - \frac{\mu\eta_*}{\alpha^2 - 1} \right) \text{ when } t \rightarrow +\infty,$$

which is strictly negative for  $|\alpha|$  close enough to 1. This holds for any initial value  $B_i(0)$ .

Assume now that  $\eta_* > 0$ . By Proposition 7.7, incompressible deformations cannot be used to construct a counter-example. For any initial value  $B_i(0)$ , we construct a deformation for which  $D_{\text{int}}(1) < 0$ , which is sufficient for our purpose, as follows. We start with

$$F(t) = f(t)^{-1/2} \text{diag}(e^{t(1-t)/2}, e^{t(t-1)/2}, 1),$$

and will adjust  $f$  later on. We again compute the relevant quantities,

$$\begin{aligned} d(t) &= \left( \frac{1}{2} - t \right) \text{diag}(1, -1, 0) - \frac{1}{2}f'(t)f(t)^{-1}I, \\ C(t) &= f(t)^{-1} \text{diag}(e^{t(1-t)}, e^{t(t-1)}, 1), \quad M(t) = \text{diag}(e^{t(t-1)}, e^{t(1-t)}, 1) \\ M'(s) &= (2s - 1) \text{diag}(e^{s(s-1)}, -e^{s(1-s)}, 0). \end{aligned}$$

For  $t = 1$ ,  $M^{-1}(1) = I$  so that  $M'(s) : M^{-1}(1) = 2(1 - 2s) \sinh(s(1 - s))$ . We will take  $f$  such that  $f'(1) = 0$  so that  $d(1) = -\frac{1}{2} \text{diag}(1, -1, 0)$ . In addition, we will also take  $f(1) = 1$ . We substitute all these values in (48) for  $t = 1$  and obtain

$$D_{\text{int}}(1) = \eta_s - \frac{\mu\eta_*}{e} \int_0^1 f(s)e^s(1 - 2s) \sinh(s(1 - s)) ds + \frac{\mu}{2e} \text{tr}(B_i(0)). \quad (49)$$

Let us now take  $\alpha > 0$  and let  $f(s) = \alpha$  on  $[0, 1/2]$ :

$$\int_0^1 f(s)e^s(1 - 2s) \sinh(s(1 - s)) ds = \alpha I_0 + \int_{1/2}^1 f(s)e^s(1 - 2s) \sinh(s(1 - s)) ds$$

with

$$I_0 = \int_0^{1/2} e^s(1 - 2s) \sinh(s(1 - s)) ds > 0.$$

Since  $e^s|(1 - 2s) \sinh(s(1 - s))| \leq e \sinh(1/4) \leq 1$  on  $[1/2, 1]$ , we see that

$$\left| \int_0^1 f(s)e^s(1 - 2s) \sinh(s(1 - s)) ds - \alpha I_0 \right| \leq \int_{1/2}^1 f(s) ds.$$

We now extend  $f$  to  $[1/2, 1]$  in a  $C^1$ -fashion, with  $f'(1) = 0$ ,  $f(1) = 1$  and  $\int_{1/2}^1 f(s) ds \leq \frac{\alpha I_0}{2}$ , which is clearly possible. It follows that

$$D_{\text{int}}(1) \leq \eta_s - \frac{\alpha\mu\eta_* I_0}{2e} + \frac{\mu}{2e} \text{tr}(B_i(0)) < 0$$

for  $\alpha$  large enough. □

*Remark 8.8.* As a side remark, we notice that

$$-\int_0^t e^{\frac{s}{\lambda_1}} f(s) M'(s) ds = C^{-1}(0) - e^{\frac{t}{\lambda_1}} C^{-1}(t) + \int_0^t (e^{\frac{s}{\lambda_1}} f(s))' M(s) ds$$

by integration by parts. Therefore,

$$\begin{aligned} -\int_0^t e^{\frac{s}{\lambda_1}} f(s) M'(s) ds : M^{-1}(t) &= C^{-1}(0) : M^{-1}(t) - 3e^{\frac{t}{\lambda_1}} f(t) \\ &\quad + \int_0^t (e^{\frac{s}{\lambda_1}} f(s))' M(s) : M^{-1}(t) ds. \end{aligned}$$

Now  $M$  is  $\text{SL}(3)$ -valued so that  $C^{-1}(0) : M^{-1}(t) \geq 3f(0)$  by Lemma 7.4, and if  $(e^{\frac{s}{\lambda_1}} f(s))' \geq 0$ , that is to say if  $\frac{1}{\lambda_1} f + f' \geq 0$ , it follows that

$$-\int_0^t e^{\frac{s}{\lambda_1}} f(s) M'(s) ds : M^{-1}(t) \geq 3f(0) - 3e^{\frac{t}{\lambda_1}} f(t) + 3 \int_0^t (e^{\frac{s}{\lambda_1}} f(s))' ds = 0.$$

In the case when  $\eta_* > 0$ , we deduce from this that if  $\frac{1}{\lambda_1} f + f' \geq 0$ , then the dissipation remains nonnegative at all positive times when  $B_i(0)$  is positive semidefinite. This condition is equivalent to  $J(t) \leq e^{\frac{3}{2\lambda_1}(t-s)} J(s)$  for all  $s \leq t$ , or in Eulerian terms  $\text{div}_x v \leq \frac{3}{2\lambda_1}$ , *i.e.*, what we could call moderately expansive deformations, which include of course incompressible deformations. The problem with the second principle in the compressible case with constant coefficients thus occurs when deformations expand faster than this, which is the case of the second counter-example above.



There is one case when we can prove that the second principle is conditionally satisfied.

**Proposition 8.9.** *If  $\eta_* = \lambda_* = 0$ , the compressible Oldroyd B fluid satisfies the second principle of thermodynamics conditionally when  $B_i(X, 0)$  is positive semi-definite.*

*Proof.* In this case, the flow rule reduces to  $\frac{\partial B_i}{\partial t} = -\frac{1}{\lambda_1(J)}B_i$ , so that  $B_i(X, t) = e^{-\int_0^t \frac{ds}{\lambda_1(J(s))}} B_i(X, 0)$ . If  $B_i(X, 0)$  is positive semidefinite, then so is  $B_i(X, t)$  and  $C(X, t) : B_i(X, t) \geq 0$  for all  $t$ .  $\square$

*Remarks 8.10.* This result is a little disappointing. It would be worthwhile to find more general conditions with non constant coefficients under which the compressible Oldroyd B model satisfies the second principle conditionally, with the possible inclusion of thermal effects.

## 8.2 Nonlinear Oldroyd B

We return to the incompressible case. By using the same Lagrangian framework and more elaborate flow rules, we can define a whole family of nonlinear incompressible Oldroyd B models. Specifically, given  $k \in \mathbb{N}$ , we set

$$\widehat{K}_k(F, H, B_i) = -\frac{\mu^k}{\lambda_1 \mu_k} B_i (C B_i)^k + \frac{2\eta_p}{\mu \lambda_1} F^{-1} \text{Sym}(H F^{-1}) F^{-T}. \quad (50)$$

For  $k = 0$  and  $\mu_0 = 1$ , this is the linear flow rule (25) which gives rise to the usual Oldroyd B model in the Eulerian formulation. The constants in the nonlinear flow rule above have the same interpretation as before as far as  $\mu$ ,  $\lambda_1$ ,  $\eta_p$  are concerned. The constant  $\mu_k > 0$  is a physical parameter that is homogeneous to a pressure to the power  $k$ . Indeed,  $C$  and  $B_i$  are dimensionless whereas the shear modulus  $\mu$  is homogenous to a pressure.

**Proposition 8.11.** *The Eulerian polymer stress corresponding to the data (20), (22), and (50) satisfies*

$$\overset{\nabla}{\sigma}_p = -\frac{1}{\lambda_1 \mu_k} \sigma_p^{k+1} + \frac{2\eta_p}{\lambda_1} d. \quad (51)$$

*Proof.* We proceed exactly as in the proof of Proposition 6.1 by substituting the flow rule (50) into equation (27). The second term in the flow rule gives rise to the second term in the right-hand side of (51) as before. The first term becomes

$$\mu F \left( -\frac{\mu^k}{\lambda_1 \mu_k} B_i (C B_i)^k \right) F^T = -\frac{1}{\lambda_1 \mu_k} (\mu F B_i F^T)^{k+1} = -\frac{1}{\lambda_1 \mu_k} \sigma_p^{k+1}$$

by equation (23).  $\square$

We thus obtain nonlinear incompressible Oldroyd B models by writing  $\sigma = \sigma_s + \sigma_p$  with  $\sigma_s = 2\eta_s d$ .

**Corollary 8.12.** *The Eulerian form of the above Lagrangian model reads*

$$\frac{1}{\mu_k} (\sigma - 2\eta_s d)^{k+1} + \lambda_1 \overset{\nabla}{\sigma} = 2(\eta_p d + \lambda_1 \eta_s d). \quad (52)$$

*Remark 8.13.* These models are frame-indifferent by construction. This also follows from the fact that the left-hand side of (52) features an objective derivative of  $\sigma$  and the right-hand side an objective derivative of  $d$

$$\lambda_1 \overset{\diamondsuit_1}{\sigma} = 2\eta\lambda_2 \overset{\diamondsuit_2}{d}, \quad (53)$$

where  $\eta = \eta_s + \eta_p$  and  $\lambda_2 = \lambda_1\eta_s/\eta$  as before.

There is no nice general expression for  $\overset{\diamondsuit_1}{\sigma}$  because of the noncommutative binomial expression. It however yields a symmetric-valued polynomial in  $(\sigma, d)$ , which is an objective function. Let us expand a couple of examples for small  $k$ .

For  $k = 1$ , we obtain

$$(\sigma - 2\eta_s d)^2 = \sigma^2 - 2\eta_s(\sigma d + d\sigma) + 4\eta_s^2 d^2,$$

so that (52) can be rearranged as

$$\sigma^2 - 2\eta_s(\sigma d + d\sigma) + \mu_1\lambda_1 \overset{\nabla}{\sigma} = 2\left(\mu_1\eta_p d - 2\eta_s^2 d^2 + \mu_1\lambda_1\eta_s \overset{\nabla}{d}\right).$$

or

$$\lambda_1 \left( \overset{\square}{\sigma} - \left( \frac{2\eta\lambda_2}{\mu_1\lambda_1^2} + 1 \right) (\sigma d + d\sigma) + \frac{\sigma^2}{\mu_1\lambda_1} \right) = 2\eta\lambda_2 \left( \overset{\square}{d} + \frac{\lambda_1 - \lambda_2}{\lambda_1\lambda_2} d - \left( \frac{2\eta\lambda_2}{\mu_1\lambda_1^2} + 2 \right) d^2 \right),$$

where  $\overset{\square}{\sigma}$  denotes the Zaremba-Jaumann derivative, in order to exhibit the objective derivatives  $\overset{\diamondsuit_1}{\sigma}$  and  $\overset{\diamondsuit_2}{d}$ , see the remarks following Proposition 3.2. These objective derivatives are not unique because we could actually distribute the terms in  $d$  and  $d^2$  at will in either side of the equation. All terms containing  $\sigma$  must however remain in the left-hand side.

When  $\eta_p = 0$ , *ie.*,  $\lambda_1 = \lambda_2$ , and  $\mu_1 = 1$ , this is precisely the quadratic model obtained by [4].

For  $k = 2$ , we obtain a cubic Oldroyd B model as follows

$$\begin{aligned} \sigma^3 - 2\eta_s(\sigma^2 d + \sigma d\sigma + d\sigma^2) + 4\eta_s^2(\sigma d^2 + d\sigma d + d^2\sigma) + \mu_2\lambda_1 \overset{\nabla}{\sigma} \\ = 2\left(\mu_2\eta_p d + 4\eta_s^3 d^3 + \mu_2\lambda_1\eta_s \overset{\nabla}{d}\right). \end{aligned}$$

All these models involve nonlinear differential equations for  $B_i$  or  $\sigma_p$  as soon as  $k \geq 1$ , whereas the corresponding differential equations are linear for  $k = 0$ . Consequently, given a prescribed, smooth enough deformation  $\phi(X, t)$  and an initial value for  $B_i$  or  $\sigma_p$ , the existence of the internal variables is a priori only ensured locally in time when  $k \geq 1$ . It is not clear that they exist globally in time.

More generally, by taking linear combinations of flow rules of the form (50), we obtain models of the form

$$\overset{\nabla}{\sigma}_p = -\frac{1}{\lambda_1}\sigma_p P(\sigma_p) + \frac{2\eta_p}{\lambda_1}d, \quad (54)$$

where  $P \in \mathbb{R}[X]$  is any polynomial. For instance,  $P(X) = 1 + \varepsilon X$ ,  $\varepsilon > 0$  small, would correspond to a quadratic correction of the classical linear Oldroyd B model.

Let us now discuss second principle issues for these nonlinear models. We just consider the monomial models above.

**Proposition 8.14.** *The constitutive law for the internal dissipation corresponding to the flow rule  $\widehat{K}_k$  is given by*

$$\widehat{D}_{\text{int}}(F, H, B_i) = 2\eta_s \|\text{Sym}(HF^{-1})\|^2 + \frac{\mu^{k+1}}{2\lambda_1\mu_k} \text{tr}((CB_i)^{k+1})$$

in Lagrangian form and

$$\widehat{d}_{\text{int}}(h, \sigma_p) = 2\eta_s \|d\|^2 + \frac{1}{2\lambda_1\mu_k} \text{tr}(\sigma_p^{k+1})$$

in Eulerian form.

*Proof.* Many terms are the same as before, we only focus on the one that is different, namely

$$\frac{\mu}{2} C : \left( \frac{\mu^k}{\lambda_1\mu_k} B_i (CB_i)^k \right) = \frac{\mu^{k+1}}{2\lambda_1\mu_k} \text{tr}((CB_i)^{k+1}).$$

To translate this into Eulerian terms, we notice that

$$\text{tr}((CB_i)^{k+1}) = \text{tr}((FB_iF^T)^{k+1})$$

hence the result since  $\sigma_p = \mu FB_iF^T$ .  $\square$

**Proposition 8.15.** *For  $k$  odd, the model satisfies the second principle unconditionally, i.e., in the sense of Coleman-Noll, as long as the internal variables exist.*

*Proof.* Indeed,  $k+1$  is then even, therefore  $\text{tr}(\sigma_p^{k+1}) \geq 0$  for any  $\sigma_p \in \text{Sym}_3$ .  $\square$

*Remark 8.16.* For  $k \geq 2$  even, we do not have an explicit formula for  $B_i$  to work with as in the case  $k = 0$ . For the conditional second principle to hold, the initial dissipation must still be positive for all  $C(0) = C \in \text{Sym}_3^+ \cap \text{SL}(3)$ . Since  $B_i(0)$  is symmetric, it is orthogonally diagonalizable with  $B_i(0) = Q^T \Delta Q$ ,  $\Delta = \text{diag}(v_j)$ .

$$\begin{aligned} \text{tr}((CB_i(0))^{k+1}) &= \text{tr}((CQ^T \Delta Q)^{k+1}) \\ &= \text{tr}(Q(CQ^T \Delta Q)^{k+1} Q^T) = \text{tr}((QCQ^T \Delta)^{k+1}). \end{aligned}$$

Now, as in the proof of Proposition 7.5,  $Q^T C Q$  can in fact be any matrix in  $\text{Sym}_3^+$ , not just those in  $\text{SL}(3)$ , in particular  $C = \text{diag}(c_j)$ ,  $c_j > 0$ . It follows that we must have  $\sum_{j=1}^3 c_j^{k+1} v_j^{k+1} \geq 0$ , hence  $v_j^{k+1} \geq 0$  for all  $j$ . Since  $k+1$  is odd, it is necessary that  $v_j \geq 0$ , so that  $B_i(0)$  must be positive semi-definite.

Conversely, for any  $C \in \text{Sym}_3^+$ ,  $\text{tr}((CB_i(0))^{k+1}) = ((CB_i(0))^k C) : B_i(0)$ . Since  $k$  is even, it is clear that for all  $B \in \text{Sym}_3$ ,  $(CB)^k C \in \overline{\text{Sym}_3^+}$ . Therefore, if  $B_i(0)$  is positive semi-definite, then  $\text{tr}((CB_i(0))^{k+1}) \geq 0$ .

If  $B_i(0)$  is positive definite, then it will remain so at least for some time, and the dissipation is initially strictly positive. It is not clear that the dissipation stays nonnegative for as long as  $B_i$  exists.

### 8.3 The Zaremba-Jaumann and Oldroyd A fluids

We finally consider complex fluid models based on two other objective derivatives, the Zaremba-Jaumann fluid, see for instance [3], and the Oldroyd A fluid, [7]-[9]-[10], both models being considerably less prominent in the literature than the Oldroyd B fluid.

Expressed in terms of the polymer stress, they simply read

$$\sigma_p + \lambda_1 \overset{\square}{\sigma}_p = 2\eta_p d, \quad (55)$$

for Zaremba-Jaumann and

$$\sigma_p + \lambda_1 \overset{\Delta}{\sigma}_p = 2\eta_p d, \quad (56)$$

for Oldroyd A, together with a Newtonian solvent stress.

We can derive both models from our Lagrangian formulation by adapting the flow rule while retaining (20) and (22). It is to be expected that they are slightly less natural than the Oldroyd B model, due to Proposition 3.3.

We thus use the same ingredients as for the Oldroyd B model, except for the flow rule, with

$$\begin{aligned} \widehat{K}_{ZJ}(F, H, B_i) = & -\frac{1}{\lambda_1} B_i + \frac{2\eta_p}{\mu\lambda_1} F^{-1} \text{Sym}(HF^{-1})F^{-T} \\ & - F^{-1} \text{Sym}(HF^{-1})FB_i - B_i F^T \text{Sym}(HF^{-1})F^{-T}, \end{aligned} \quad (57)$$

for Zaremba-Jaumann, and

$$\begin{aligned} \widehat{K}_A(F, H, B_i) = & -\frac{1}{\lambda_1} B_i + \frac{2\eta_p}{\mu\lambda_1} F^{-1} \text{Sym}(HF^{-1})F^{-T} \\ & - 2F^{-1} \text{Sym}(HF^{-1})FB_i - 2B_i F^T \text{Sym}(HF^{-1})F^{-T}. \end{aligned} \quad (58)$$

for Oldroyd A. It is a simple computation to check that

**Proposition 8.17.** *The Lagrangian models produced by the above choices are the Zaremba-Jaumann fluid and Oldroyd A fluid models respectively.*

Note that the correction applied to Oldroyd B in order to obtain Oldroyd A is twice that applied to obtain Zaremba-Jaumann. Indeed,  $\overset{\square}{\sigma}_p = \frac{1}{2}(\overset{\nabla}{\sigma}_p + \overset{\Delta}{\sigma}_p)$ , see Section 3. We can also compute their internal dissipations. For the Zaremba-Jaumann fluid, we obtain

**Proposition 8.18.** *The internal dissipation of the Zaremba-Jaumann model is given by*

$$\widehat{D}_{\text{int}}(F, H, B_i) = 2\eta_s \|\text{Sym}(HF^{-1})\|^2 + \frac{\mu}{2} B_i : \left( \frac{1}{\lambda_1} C + C' \right) \quad (59)$$

using  $C'$  as shorthand for  $2F^T \text{Sym}(HF^{-1})F$ , in the Lagrangian formulation and

$$\widehat{d}_{\text{int}}(h, \xi) = 2\eta_s \|d\|^2 + \frac{1}{2\lambda_1} \text{tr } \xi + \xi : d \quad (60)$$

in the Eulerian formulation, with  $\xi = \sigma_p$  as before.

*Proof.* Let us just compute the dissipation stemming from the internal variable  $\widehat{D}_{\text{int,p}}$ . We still have  $\frac{\partial \widehat{A}_m}{\partial B_i} = \frac{\mu}{2} C$ . Therefore

$$\begin{aligned} \widehat{D}_{\text{int,p}}(F, H, B_i) &= \frac{\mu}{2} C : \left( \frac{1}{\lambda_1} B_i - \frac{2\eta_p}{\mu\lambda_1} F^{-1} \text{Sym}(HF^{-1}) F^{-T} \right. \\ &\quad \left. + F^{-1} \text{Sym}(HF^{-1}) F B_i + B_i F^T \text{Sym}(HF^{-1}) F^{-T} \right) \\ &= \frac{\mu}{2} \left( C : \frac{1}{\lambda_1} B_i + 2F^T \text{Sym}(HF^{-1}) F : B_i \right) \end{aligned}$$

since  $\text{tr}(\text{Sym}(HF^{-1})) = \text{tr}(d) = 0$  as before. The translation in Eulerian terms is straightforward using (34) and since  $\xi = \sigma_p = \mu F B_i F^T$  still.  $\square$

It is still necessary that  $B_i(0)$  be positive semidefinite for the conditional second principle to hold. However, we have  $\widehat{d}_{\text{int}}(h, \xi) = \sigma : d + \frac{1}{2\lambda_1} \text{tr} \xi$ , where  $\sigma : d$  is the naive dissipation. In [8], we showed numerically that the naive dissipation also changes sign for the Zaremba-Jaumann fluid, even with positive semidefinite initial conditions. It is not too difficult to see that  $\text{tr} \xi$  is in this case a decreasing exponential in time, so that  $\widehat{d}_{\text{int}}(h, \xi)$  is going to change sign as well for the same evolutions, see Figure 1 below.

Also in [8], we showed that the choice  $\widehat{a}_m(\xi) = \frac{\lambda_1}{4\eta_p} \|\xi\|^2$  as free energy makes it satisfy the second principle unconditionally. Unfortunately, it does not seem to be possible to recover this Eulerian free energy from a Lagrangian free energy based on some adequate nonlinearly elastic stored energy function, thus giving it some physical grounding. The present approach seems to require the use of the neo-Hookean energy to work.

The situation is the same for the Oldroyd A fluid, based on the remark above on the corrections applied to Oldroyd B.

**Proposition 8.19.** *The internal dissipation of the Oldroyd A model is given by*

$$\widehat{D}_{\text{int}}(F, H, B_i) = 2\eta_s \|\text{Sym}(HF^{-1})\|^2 + \frac{\mu}{2} B_i : \left( \frac{1}{\lambda_1} C + 2C' \right) \quad (61)$$

*in the Lagrangian formulation and*

$$\widehat{d}_{\text{int}}(h, \xi) = 2\eta_s \|d\|^2 + \frac{1}{2\lambda_1} \text{tr} \xi + 2\xi : d \quad (62)$$

*in the Eulerian formulation.*

The same negative considerations concerning the second principle also hold, except for that on the trace of  $\xi$ . In conclusion, for both Zaremba-Jaumann and Oldroyd A models, positive semidefiniteness of the initial condition is a necessary condition for the conditional second principle. It does not seem to be sufficient however, based on numerical evidence, see Figure 1. This does not rule out a potentially smaller set of adequate initial values that was not empty and not numerically tested on the one hand, and on the other hand, other physically motivated choices of free energies for which the second principle could be satisfied by these models.

In Figure 1 below, we show the results of the same numerical simulation for both Oldroyd models and for the Zaremba-Jaumann model, with the same

data, already described in [8]. Namely, an Eulerian computation with  $h(x, t) = \cos(\omega t)m$ , where  $m$  is a randomly chosen  $3 \times 3$  traceless matrix with coefficients between  $-1$  and  $1$ , and  $\omega = 0.75$ . The material constants are  $\eta = 2$ ,  $\lambda_1 = 10$ ,  $\lambda_2 = .5$ . The initial value for  $\sigma_p = \xi$  is  $\xi(0) = .1I_3$ , a positive-definite matrix. We plot the internal dissipation  $d_{\text{int}}(t)$  with a solid line,  $\frac{1}{2\lambda_1} \text{tr} \xi(t)$  with a dashed line and  $\xi(t) : d(t)$  with a dotted line for  $t$  from 0 to 40 (with different vertical scales for each model). We see that the Oldroyd B dissipation (32) remains nonnegative as expected, the  $\xi : d$  plot being irrelevant in this case, whereas both Zaremba-Jaumann dissipation (60) and Oldroyd A dissipation (62) take strictly negative values in finite time, thus violating the conditional second principle.

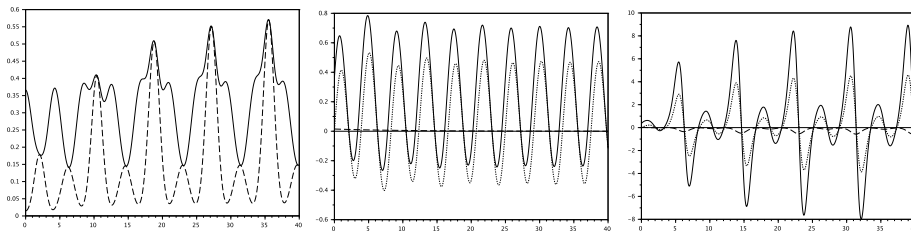


Figure 1: Left: Oldroyd B, center: Zaremba-Jaumann, right: Oldroyd A.

In Figure 2, we plot the smallest eigenvalue of  $\sigma_p$  vs. time in the above Oldroyd B case. We see that this smallest eigenvalue becomes strictly negative, hence neither  $\sigma_p$  nor  $B_i$  remain positive semidefinite for all times in this particular example either, *cf.* Proposition 7.6.

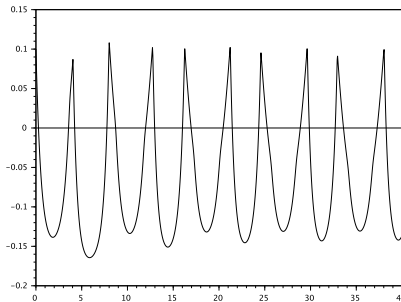


Figure 2: Smallest eigenvalue of  $\sigma_p$ , Oldroyd B case.

## References

- [1] B. D. Coleman and V. J. Mizel, Existence of caloric equations of state in thermodynamics, *J. Chem. Phys.*, **40** (1964), 1116-1125.

- [2] B. D. Coleman and W. Noll, The thermodynamics of elastic materials with heat conduction and viscosity, *Arch. Rational Mech. Anal.*, **13** (1963), 167–178.
- [3] T. Eiter, K. Hopf, and R. Lasarzik, Weak-strong uniqueness and energy-variational solutions for a class of viscoelastoplastic fluid models, to appear *Adv. Nonlinear Anal.* **12** (2023).
- [4] G. Francfort and O. Lopez-Pamies, Derivation of a kind of Lagrangian Oldroyd model using “Truesdellian thinking”, personal communication, 2021.
- [5] M. E. Gurtin, E. Fried, and L. Anand, *The Mechanics and Thermodynamics of Continua*, Cambridge University Press, Cambridge, 2010.
- [6] B. Halphen and Q.S. Nguyen, Sur les matériaux standards généralisés, *J. Mécanique*, **14** (1975), 39–63.
- [7] M. Hinch and O. Harlen, Oldroyd B, and not A?, *J. Nonnewton. Fluid Mech.*, **298** (2021).
- [8] H. Le Dret and A. Raoult, A few remarks on thermomechanics, *Discrete and Continuous Dynamical Systems - S*, doi: 10.3934/dcdss.2023116, (2023).
- [9] J. G. Oldroyd, On the formulation of rheological equations of state, *Proc. Roy. Soc. Lond. A*, **200** (1950), 523–541.
- [10] M. Renardy and B. Thomases, A mathematician’s perspective on the Oldroyd B model: Progress and future challenges, *J. Nonnewton. Fluid Mech.*, **293** (2021).
- [11] G. F. Smith, On isotropic functions of symmetric tensors, skew-symmetric tensors and vectors, *Int. J. Engng Sci.*, **9** (1971), 899-916.
- [12] C. Truesdell and W. Noll, *The Nonlinear Field Theories of Mechanics*, 3<sup>rd</sup> edition, Springer-Verlag, New York, 2004.
- [13] S. Zaremba, Sur une généralisation de la théorie classique de la viscosité, *Bull. Int. Acad. Sci. Cracovie*, (1903), 380-403.