

SOME REMARKS ABOUT THE USE OF GORDON-SCHOWALTER TIME DERIVATIVE IN RATE-TYPE VISCOELASTIC CONSTITUTIVE EQUATIONS

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Abstract. *The present paper is concerned with the analysis of the thermodynamic consistency of rate-type constitutive equations for viscoelastic fluids using the Gordon-Schowalter objective time derivative. The choice of the objective time derivative is important not only to assure a mathematically correct and physically realistic description of the viscoelastic behaviour, but also to perform a thermodynamically consistent modelling. It is shown that, for some very popular constitutive equations that adopt the Gordon-Schowalter objective derivative, it is impossible to automatically assure that the second law of thermodynamics is satisfied in any process.*

Keywords: *viscoelastic fluid behaviour; Gordon-Schowalter objective derivative; Oldroyd type fluids; second law of thermodynamics*

2. INTRODUCTION

Constitutive models for fluids are often classified as differential, rate and integral types (Dunn and Rajagopal, 1974, 1995, Rajagopal, 1993). To set up a general constitutive theory it is necessary to consider aspects of the first and second law of thermodynamics since heat transfer and dissipative behaviour must be taken into account.

The constitutive theories in which the free energy is supposed to be a function of a set of independent variables (Germain, Quoc Son and Suquet, 1983), widely used in solid mechanics (Halphen, and Quoc Son, 1975, Tigoiu and Soós, 1990) but also for fluids (Saramito, 2007, 2009, Rajagopal and Srinivasa, 2000, for instance), may provide practical tools for the modelling of non-Newtonian fluids, especially when it is necessary to account for the interference of the microstructure with the macrostructure. This is the case, for instance, of certain kinds of polymeric liquids in which the breakdown of connections between the structural units that characterize the internal (i.e. the sub macroscopic) structure may induce a decrease in the resistance to flow (Byrd, Armstrong and Hassager, 1981, Billington and Tate, 1981).

In these theories, some variables are introduced to account for dissipative mechanisms. In a phenomenological approach, such additional variables are supposed to contain micro structural information, relevant for certain classes of materials. To each of these variables is associated one constitutive equation in such a way that a local version of the second law of thermodynamics must be satisfied. The study of viscoelastic fluids cannot be adequately performed without the analysis if a given set of objective constitutive equations is thermodynamically admissible or not.

This paper is concerned with the analysis of the thermodynamic consistency of a class of (rate-type) viscoelastic constitutive equations obtained considering the Gordon-Schowalter objective derivative – the here called *Oldroyd type* constitutive equations (including the popular lower-convected, Jaumann and upper-convected *Maxwell* models). It is shown that it is impossible to assure that the SLT is automatically satisfied in any process. The analysis is based on the study of sufficient conditions for an objective and thermodynamically consistent modelling of viscoelastic fluid behaviour, developed within the framework of thermodynamics of irreversible processes by the author (da Costa Mattos, 1998, 2012). In the theory, the choice of a particular objective time derivative in rate type constitutive equations can be interpreted as a constitutive assumption. For each objective time derivative it is associated a particular measure of deformation in order to assure that a local version of the second law of thermodynamics is automatically satisfied.

2 – PRELIMINARY DEFINITIONS

Under suitable regularity assumptions it is possible to consider the following expressions as local versions of the first law (FLT) and second law of thermodynamics (SLT) (Billington and Tate, 1981, Truesdel and Toupin, 1960):

$$FLT : \rho \dot{e} = -div(\mathbf{q}) + \mathbf{T} : \mathbf{D}; \quad SLT : d = \mathbf{T} : \mathbf{D} - \rho(\dot{\psi} + s\dot{\theta}) + \mathbf{q} \cdot \mathbf{g} \geq 0 \quad (1)$$

where $\dot{(\)}$ denotes the material time derivative of $(\)$; ρ is the mass density; \mathbf{T} the Cauchy stress tensor; $\mathbf{D} = 1/2 [\mathbf{grad}(\mathbf{v}) + \mathbf{grad}(\mathbf{v})^T]$ the deformation rate tensor; e the internal energy per unity mass, θ the absolute

temperature; s the total entropy per unit mass; $\psi = (e - \theta s)$ the Helmholtz free energy per unit mass; \mathbf{q} the heat flux vector and $\mathbf{g} = -\mathbf{grad}(\log \theta)$, $\log \theta$ standing for the logarithm of the absolute temperature. d is the rate of energy dissipation per unit volume. The second law of thermodynamics makes a distinction between possible processes ($d \geq 0$) and impossible processes ($d < 0$). The possible processes may be reversible (the rate of energy dissipation d is always equal to zero) or not. This local version of the SLT does not exclude the possibility of unusual behaviours such as a decreasing temperature if heat is added to the medium. To exclude the possibility of such kind of unusual behaviour, the present study is restricted to fluids that always satisfy a further restrictive constraint:

$$d_1 = \mathbf{T} : \mathbf{D} - \rho(\dot{\psi} + s\dot{\theta}) \geq 0 \quad , \quad d_2 = \mathbf{q} \cdot \mathbf{g} \geq 0 \quad (2)$$

Obviously, if the above relations are satisfied, then the SLT constraint in (1) will also be satisfied. It is also simple to verify that (2) leads to the classical heat conduction inequality $-\mathbf{q} \cdot \mathbf{grad}(\theta) \geq 0$ since the absolute temperature θ is a positive quantity. This relation implies that heat flows in the direction of decreasing temperature when \mathbf{q} is parallel to the temperature gradient. The quantity d_1 is usually called the intrinsic dissipation and the quantity d_2 the thermal dissipation.

2. OBJECTIVE TIME DERIVATIVES

The material time derivative of an objective tensor quantity is not necessarily objective. Therefore, in order to assure objectivity, it is necessary to use some special kind of time derivative in rate type constitutive equations. A large number of definitions of objective time derivatives can be found in the literature (Jaumann, Truesdell, Cotter-Rivling, Gordon-Schowalter, etc. (Truesdell and Noll, 1965, Gordon and Schowalter, 1972, Billington and Tate, 1981, Meyers, Schieße and Bruhns, 2000)). The choice of a particular derivative is important and can be interpreted as a constitutive assumption. The present study is restricted to the Gordon-Schowalter derivative (Gordon and Schowalter, 1972), adopted in many works concerned with viscoelastic fluids (Marchal and Crochet, 1987, Johnson Jr. and Segalman, 1977, Lee and Xu, 2006, Dimakopoulos, 2010, for instance).

$$\overset{\circ}{\mathbf{A}} = \dot{\mathbf{A}} + \mathbf{A}\mathbf{W} - \mathbf{W}\mathbf{A} - a(\mathbf{A}\mathbf{D} + \mathbf{D}\mathbf{A}) \quad (3)$$

where $\overset{\circ}{\mathbf{A}}$ is the (objective) Gordon-Schowalter time derivative of an arbitrary objective symmetric tensor \mathbf{A} . $\mathbf{W} = 1/2 [\mathbf{grad}(\mathbf{v}) - \mathbf{grad}(\mathbf{v})^T]$ is the vorticity tensor and a is a scalar parameter such that $-1 \leq a \leq 1$. It is important to remark that, only if $a = 0$ in eq. (5) it is possible to assure the following properties

(i) The derivative of the unit tensor $\mathbf{1}$ vanishes: $\overset{\circ}{\mathbf{1}} = \mathbf{0}$; (ii) Any objective rate of a symmetric tensor \mathbf{A} should be symmetric: $\overset{\circ}{\mathbf{A}} = (\overset{\circ}{\mathbf{A}})^T = (\overset{\circ}{\mathbf{A}^T})$; (iii) For any scalar differentiable isotropic function ψ of a symmetric objective tensor the following generalized ‘chain rule’ holds: $\overset{\circ}{\psi} = \dot{\psi}$, with $\overset{\circ}{\psi} = \frac{\partial \psi}{\partial \mathbf{A}} : \overset{\circ}{\mathbf{A}}$ and $\dot{\psi} = \frac{\partial \psi}{\partial \mathbf{A}} : \dot{\mathbf{A}}$

Although these properties are not verified for the Gordon-Schowalter derivative if $a \neq 0$, there is not necessarily any thermodynamic restriction to the use of derivatives with $a \neq 0$ in eq. (3). When $a = 0$ we obtain the Jaumann derivative of tensors, while $a = 1$ and $a = -1$ are associated to the upper and the lower convected derivatives, respectively. For $a = 1$, the Gordon-Schowalter derivative coincides with the upper-convected Oldroyd derivative.

3. CONSTITUTIVE FRAMEWORK

In this section, it is presented an abstract framework to define a family of fluids with viscoelastic behaviour. In order to postulate a general theory of viscoelasticity, it is essential to introduce a measure of strain. The measure of strain $\boldsymbol{\varepsilon}$ is such that:

$$\overset{\circ}{\boldsymbol{\varepsilon}} = \mathbf{D} \quad (4)$$

where $\overset{\circ}{\boldsymbol{\varepsilon}}$ denotes the objective time derivative of $\boldsymbol{\varepsilon}$. Different measures of strain can be chosen depending on the definition of objective time derivative. Hence, the choice of the particular derivative can be seen as a constitutive hypothesis and the first step to establish a physically realistic set of constitutive equations.

The present work considers fluids for which the Helmholtz free energy ψ can be expressed as an isotropic and differentiable function of the absolute temperature θ , of the density ρ , of the measure of strain $\boldsymbol{\varepsilon}$ and of whatever may be relevant independent variables, which must be objective quantities. Generally, these variables are used to model the interference of the microstructure with the macrostructure and are called internal variables. Since in this theory most of the difficulties with material objectivity appear when these additional internal variables are tensor quantities, from now on only one abstract additional second order symmetric tensor variable $\boldsymbol{\gamma}$ is considered in order to simplify the presentation. Such variable is assumed to be related to irreversible changes on the microstructure caused by different possible sub macroscopic mechanisms. The Helmholtz free energy ψ is an isotropic and differentiable function of the variables $(\rho, \boldsymbol{\varepsilon}, \boldsymbol{\gamma}, \theta) \Rightarrow \psi = \psi(\rho, \boldsymbol{\varepsilon}, \boldsymbol{\gamma}, \theta)$. For now on, to simplify the presentation, the study is restricted to incompressible fluids undergoing isothermal processes, and therefore, the Helmholtz free energy ψ does not depend on the temperature, $d = d_1$ and the Helmholtz free energy must have the following particular form (da Costa Mattos, 1998, 2012): $\psi(\rho, \boldsymbol{\varepsilon}, \boldsymbol{\gamma}) = (1/\rho)\hat{\psi}(\boldsymbol{\varepsilon}, \boldsymbol{\gamma})$.

To set up a general constitutive theory it is then necessary to consider aspects of the second law of thermodynamics since dissipative behaviour must be taken into account. Using the previous definitions, the following expression can be obtained for the intrinsic dissipation d_1

$$d_1 = \left(\mathbf{T} - \rho \left[\frac{\partial \psi}{\partial \boldsymbol{\varepsilon}} + 2a \left(\frac{\partial \psi}{\partial \boldsymbol{\varepsilon}} \boldsymbol{\varepsilon} + \frac{\partial \psi}{\partial \boldsymbol{\gamma}} \boldsymbol{\gamma} \right) \right] \right) : \mathbf{D} - \rho \frac{\partial \psi}{\partial \boldsymbol{\gamma}} : \dot{\boldsymbol{\gamma}} \quad (5)$$

The relation of the internal variable $\boldsymbol{\gamma}$ with dissipative mechanisms is introduced through the intrinsic dissipation, which is assumed to have the following form

$$d_1 = \mathbf{h}_D(\mathbf{D}, \dot{\boldsymbol{\gamma}}) : \mathbf{D} + \mathbf{h}_\gamma(\mathbf{D}, \dot{\boldsymbol{\gamma}}) : \dot{\boldsymbol{\gamma}} \quad \text{with } \mathbf{h}_D \text{ and } \mathbf{h}_\gamma \text{ being symmetric tensor functions of } \mathbf{D} \text{ and } \dot{\boldsymbol{\gamma}} \quad (6)$$

Definition (6) implies that $d_1 = 0$ when $\mathbf{D} = \mathbf{0}$ and $\dot{\boldsymbol{\gamma}} = \mathbf{0}$. Hence, in any rigid body motion, the rate of energy dissipation d_1 may be positive ($d_1 > 0$) only if $\dot{\boldsymbol{\gamma}} \neq \mathbf{0}$, otherwise $d_1 = 0$. As a consequence, in this kind of motion, the only possible dissipative mechanism would be the microscopic changes related to the internal variable $\boldsymbol{\gamma}$. The choice of the internal variables and of the functions ψ , \mathbf{h}_D and \mathbf{h}_γ characterize completely a given fluid in this theory. Taking into account the previous definitions it is possible to conclude (da Costa Mattos, 2012) that the following relations must hold independently of $(\mathbf{D}, \dot{\boldsymbol{\gamma}}, \dot{\theta})$

$$\rho \frac{\partial \psi}{\partial \boldsymbol{\gamma}} + \mathbf{h}_\gamma(\mathbf{D}, \dot{\boldsymbol{\gamma}}) = \mathbf{0}; \quad \mathbf{T} = -P\mathbf{1} + \rho \left[\frac{\partial \psi}{\partial \boldsymbol{\varepsilon}} + 2a \left(\frac{\partial \psi}{\partial \boldsymbol{\varepsilon}} \boldsymbol{\varepsilon} + \frac{\partial \psi}{\partial \boldsymbol{\gamma}} \boldsymbol{\gamma} \right) \right] + \mathbf{h}_D(\mathbf{D}, \dot{\boldsymbol{\gamma}}) \quad (7)$$

Equations (7) form a set of objective constitutive equations (for incompressible fluids undergoing isothermal processes). These equations characterize the here called *generalized viscoelastic fluids*. The first equation in (7) relates the objective time derivative of $\boldsymbol{\gamma}$ with the other independent variables. Similar expressions appear in works concerned with solid mechanics and are generally called complementary equations. Nevertheless, these complementary equations are directly postulated and they are not obtained, like in the present work, as consequences of hypothesis concerning the free energy and the rate of energy dissipation. The stress tensor \mathbf{T} and the others variables are related through the second equation in (7). P is the hydrostatic pressure which is a multiplier of the incompressibility constraint $\text{tr}(\mathbf{D}) = 0$. P is an unknown quantity to be determined from the problem formed by the balance equations, the constitutive equations and a suitable set of boundary and initial conditions.

4. OLDROYD TYPE FLUIDS

The only possible choice of the functions ψ , \mathbf{h}_D and \mathbf{h}_γ in order to obtain the so-called Oldroyd type fluids within this thermodynamic framework is

$$\psi(\rho, \boldsymbol{\varepsilon}, \boldsymbol{\gamma}) = \frac{E}{2\rho}(\boldsymbol{\varepsilon} - \boldsymbol{\gamma}) : (\boldsymbol{\varepsilon} - \boldsymbol{\gamma}) ; \mathbf{h}_{\boldsymbol{\gamma}}(D, \dot{\boldsymbol{\gamma}}) = \eta_1 \dot{\boldsymbol{\gamma}} ; \mathbf{h}_D(D, \dot{\boldsymbol{\gamma}}) = \eta_2 \mathbf{D} - 2a\left(\frac{\partial\psi}{\partial\boldsymbol{\varepsilon}}\boldsymbol{\varepsilon} + \frac{\partial\psi}{\partial\boldsymbol{\gamma}}\boldsymbol{\gamma}\right) \quad (8)$$

With E , η_1 , η_2 being positive material constants. In this case, $\boldsymbol{\gamma}$ can be interpreted as an irreversible parcel of $\boldsymbol{\varepsilon}$ due to the dissipative mechanism of viscosity. Considering the following notation: $\boldsymbol{\sigma} = (\mathbf{T} + P\mathbf{1})$, it is possible to obtain from (7) the following constitutive equations

$$\boldsymbol{\sigma} = \rho \frac{\partial\psi}{\partial\boldsymbol{\varepsilon}} + \eta_2 \mathbf{D} = \frac{E[(\boldsymbol{\varepsilon} - \boldsymbol{\gamma}) + \frac{2\eta_2 \mathbf{D}}{\boldsymbol{\sigma}_1}]}{\boldsymbol{\sigma}_1} ; \rho \frac{\partial\psi}{\partial\boldsymbol{\gamma}} + \mathbf{h}_{\boldsymbol{\gamma}}(D, \dot{\boldsymbol{\gamma}}) = \mathbf{0} \Rightarrow -\frac{E[(\boldsymbol{\varepsilon} - \boldsymbol{\gamma})]}{\boldsymbol{\sigma}_1} + 2\eta_1 \dot{\boldsymbol{\gamma}} = \mathbf{0} \Rightarrow \boldsymbol{\sigma}_1 = 2\eta_1 \dot{\boldsymbol{\gamma}} \quad (9)$$

Combining the previous equation it results that

$$2\eta_1 \mathbf{D} = \lambda \dot{\boldsymbol{\sigma}}_1 + \boldsymbol{\sigma}_1 \quad (10)$$

where $\lambda = \frac{2\eta_1}{E}$. Such kind of models can be called of *Oldroyd type*. The tensor $\boldsymbol{\sigma}_2$ corresponds to a Newtonian contribution or to a fast relaxation mode; models with $\eta_2 = 0$ are generally called of *Maxwell type*. The parameter $a = 1, 0$ and -1 corresponds, respectively, to the lower-convected, Jaumann and upper-convected *Maxwell* models.

5. THE SECOND LAW OF THERMODYNAMICS

In this section it is discussed whether the *Oldroyd type* fluids verify automatically the SLT constraint $d_1 \geq 0$. It is possible to show that the intrinsic dissipation has the following form for this class of fluids

$$d_1 = \mathbf{h}_D : \mathbf{D} + \mathbf{h}_{\boldsymbol{\gamma}} : \dot{\boldsymbol{\gamma}} = \eta_2 \mathbf{D} : \mathbf{D} - \left(2a\left(\frac{\partial\psi}{\partial\boldsymbol{\varepsilon}}(\boldsymbol{\varepsilon} - \boldsymbol{\gamma})\right)\right) : \mathbf{D} + \eta_1 \dot{\boldsymbol{\gamma}} : \dot{\boldsymbol{\gamma}} \quad (11)$$

The terms $\eta_2 \mathbf{D} : \mathbf{D}$ and $\eta_1 \dot{\boldsymbol{\gamma}} : \dot{\boldsymbol{\gamma}}$ are always non negative, but the remaining term can in (11) may be positive or negative, depending on the process. Therefore, it is not possible to assure that the second law constraint $d_1 \geq 0$ is automatically verified.

$$d_1 \geq 0 \Leftrightarrow (\eta_2 \mathbf{D} : \mathbf{D} + \eta_1 \dot{\boldsymbol{\gamma}} : \dot{\boldsymbol{\gamma}}) \geq \left(2a\left(\frac{\partial\psi}{\partial\boldsymbol{\varepsilon}}(\boldsymbol{\varepsilon} - \boldsymbol{\gamma})\right)\right) : \mathbf{D} = \frac{2aE}{\rho}(\boldsymbol{\varepsilon} - \boldsymbol{\gamma})^2 : \mathbf{D} \quad (12)$$

It is important to remark that the term $\left(2a\left(\frac{\partial\psi}{\partial\boldsymbol{\varepsilon}}(\boldsymbol{\varepsilon} - \boldsymbol{\gamma})\right)\right) : \mathbf{D}$ vanishes in very particular flows. For instance, in a shear flow, defined by

$$v_1 = \dot{\vartheta} x_1 ; v_2 = v_3 = 0 \quad (13)$$

with v_1, v_2, v_3 being the Cartesian coordinates of the velocity \mathbf{v} and $\dot{\vartheta}$ the shear strain, the following relation holds $\left(\frac{\partial\psi}{\partial\boldsymbol{\varepsilon}}\boldsymbol{\varepsilon}\right) : \mathbf{D} = \left(\frac{\partial\psi}{\partial\boldsymbol{\gamma}}\boldsymbol{\gamma}\right) : \mathbf{D} = 0$, provided the components of the variables $\boldsymbol{\varepsilon}$, $\boldsymbol{\gamma}$, $\frac{\partial\psi}{\partial\boldsymbol{\varepsilon}}$, $\frac{\partial\psi}{\partial\boldsymbol{\gamma}}$ and \mathbf{D} have the following particular form in a Cartesian system of coordinates, what is usually observed in viscoelastic fluids (see da Costa Mattos, 1998, for instance).

$$\begin{aligned} \varepsilon_{22} &= -\varepsilon_{11} ; \frac{\partial\psi}{\partial\varepsilon_{22}} = -\frac{\partial\psi}{\partial\varepsilon_{11}} ; \gamma_{22} = -\gamma_{11} ; \frac{\partial\psi}{\partial\gamma_{22}} = -\frac{\partial\psi}{\partial\gamma_{11}} \\ \varepsilon_{13} &= \varepsilon_{23} = \varepsilon_{33} = \frac{\partial\psi}{\partial\varepsilon_{13}} = \frac{\partial\psi}{\partial\varepsilon_{23}} = \frac{\partial\psi}{\partial\varepsilon_{33}} = 0 \\ \gamma_{13} &= \gamma_{23} = \gamma_{33} = \frac{\partial\psi}{\partial\gamma_{13}} = \frac{\partial\psi}{\partial\gamma_{23}} = \frac{\partial\psi}{\partial\gamma_{33}} = 0 \end{aligned} \quad (14)$$

Therefore, any rheometric flow of like (14) will always be thermodynamically admissible for an *Oldroyd type* fluid, what cannot be necessarily affirmed for an arbitrary flow.

6 – CONCLUDING REMARKS

The systematic procedure to obtain constitutive relations adopted in this paper is a useful tool in the modeling of non-Newtonian fluids, mainly when dissipative mechanisms related to changes on the material structure or substructure must also be taken into account. In the theory, the choice of a particular objective time derivative in rate type constitutive equations can be interpreted as a constitutive assumption. For each objective time derivative it is associated a particular measure of deformation in order to assure that a local version of the second law of thermodynamics is automatically satisfied. In the case of the so-called *Oldroyd type* fluids, obtained considering the Gordon-Schowalter objective derivative, it is shown that it is not possible to assure that the SLT constraint is automatically verified for an arbitrary flow. Therefore, care must be taken in the use of this family of constitutive equations in very complex flows, what absolutely does not mean that these constitutive equations are not interesting from the engineering point of view.

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