CONSTITUTIVE RELATIONS AND MAXIMUM RATE OF ENTROPY PRODUCTION IN NON-EQUILIBRIUM PROCESSES

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ABSTRACT

The critical step in the formulation of complete set of governing equations for motion of continuous medium is the mathematical description of the behaviour of the *specific* material of interest. The material specific behaviour is characterised via *constitutive relations* that provide a link between the response of the material and the primitive state variables. (For example, if the material of interest is a fluid, then one needs to find a relation between the Cauchy stress tensor \mathbb{T} and the heat flux vector j_q , and the state variables, which are in the case of homogeneous incompressible fluid the velocity \mathbf{v} and the temperature θ .) If only the macroscopic response of the material is of interest, then the constitutive relations should be preferably expressed in terms of macroscopic quantities only, and the involved quantities should have a good macroscopic physical interpretation.

Apparently, the specification of constitutive relations requires one to use various additional concepts that are usually based on utilisation of thermodynamic of continuous medium. Rajagopal and Srinivasa [1] have argued that the concept of "maximisation of the rate of entropy production" can be very fruitful in the specification of constitutive relations for the response of complex materials. In particular, Rajagopal and Srinivasa [1] claim that

a specific choice among a competing class of constitutive functions can be made so that the state variables evolve in a way that maximizes the rate of entropy production.

We shall clarify this concept by the analysis of a very simple fluid model – the *incompressible Navier–Stokes model*. Further, we also document as how to use this concept in the analysis of the response of a canonical model for complex fluids – the *incompressible Maxwell/Oldroyd-B model*. In the latter case the concept of "maximisation of the rate of entropy production" is used jointly with another fruitful concept, namely with the concept of "evolving natural configuration", see Rajagopal and Srinivasa [2, 3] and Rajagopal and Srinivasa [4].

The incompressible Navier–Stokes fluid and incompressible Maxwell/Oldroyd-B fluid models must be dealt with in any theory of constitutive relations, hence they provide a nice example that allows one to compare the current approach with other existing thermodynamic approaches to the theory of constitutive relations.



Figure 1: Asphalt concrete (cross-section through a sample $10 \text{ cm} \times 5 \text{ cm}$, greyscale image). The response of apparently complex materials can be in many processes described by rate type viscoelastic solid/fluid models.

Naturally, the concepts of "maximisation of the rate of entropy production" and "evolving natural configuration" have been used in much more demanding settings that go beyond Navier–Stokes fluid and standard viscoelastic rate type models. Concerning *viscoelastic materials*, the interested reader is referred to Karra [5] and Karra and Rajagopal [6] who discuss thermo-oxidative degradation of polyimides, series of papers by Barot et al. [7], Sodhi and Rao [8], Moon et al. [9] and Sodhi et al. [10] discussing shape memory polymers including light activated shape memory polymers, and numerous papers on mechanical response of asphalt and asphalt concrete, see Krishnan and Rajagopal [11, 12, 13], Narayan et al. [14, 15], Málek et al. [16, 17] and Nivedya et al. [18]. Note that especially asphalt concrete, see Figure 1, is, due to its internal structure, a material that poses a real challenge for mathematical modelling. Further, crystallization in polymers is dealt with in Rao and Rajagopal [19] and the follow-up works by Kannan et al. [20], Kannan and Rajagopal [21, 22] and Kannan et al. [23], coupling of viscoelastic response with chemical reactions is studied in the context of vulcanisation of rubber in works by Kannan and Rajagopal [24] and Kundurthi et al. [25], see also Hall and Rajagopal [26].

Trivial example - incompressible Navier-Stokes fluid

Let us start with an extremely oversimplified setting that allows one to exemplify the maximisation procedure. We consider homogeneous *incompressible* heat non-conducting fluid with the energetic equation of state in the form $e =_{def} \hat{e}(\eta)$, where *e* denotes the specific internal energy,

[e] = J/kg, and η is the specific entropy, $[\eta] = J/kg \cdot K$. The standard manipulation allows one to identify the entropy production ξ in the generic evolution equation for the entropy

$$\rho \frac{\mathrm{d}\eta}{\mathrm{d}t} + \mathrm{div}\left(\frac{\boldsymbol{j}_q}{\boldsymbol{\theta}}\right) = \boldsymbol{\xi}.$$
(1)

In the present case the entropy production $\xi =_{def} \frac{\zeta}{\theta}$ is given by

where \mathbb{D} denotes the symmetric part of the velocity gradient $\mathbb{D} =_{def} \frac{1}{2} (\nabla \boldsymbol{v} + \nabla \boldsymbol{v}^{\mathsf{T}})$. The aim is to identify the constitutive relation that is, in this case, a formula for the Cauchy stress tensor \mathbb{T} in terms of \mathbb{D} , $\mathbb{T} = \mathfrak{f}(\mathbb{D})$.

 $\zeta = \mathbb{T} : \mathbb{D},$

Let us now assume that the material produces the entropy in such a way that the entropy production is given by the formula $\zeta = \widehat{\zeta}$, where

$$\widehat{\boldsymbol{\zeta}} =_{\text{def}} 2\mu \mathbb{D} : \mathbb{D}. \tag{3}$$

(2)

(The second law is satisfied since the entropy production is clearly nonnegative.) Quantity $\hat{\zeta}$ is, besides the internal energy \hat{e} , yet another scalar quantity that characterises the behaviour of the material. The specification of two scalar quantities $\hat{\zeta}$ and \hat{e} provides a characterisation of the entropy production ability of the fluid and the energy storage ability of the fluid. The question is whether this characterisation of the fluid of interest is sufficient for the specification of the constitutive relation between tensorial quantities \mathbb{T} and \mathbb{D} .

It turns out that the answer is positive provided that we appeal to the concept of maximisation of the entropy production introduced by Rajagopal and Srinivasa [1]. The chosen entropy production $\hat{\zeta}$ must be compatible with the generic formula (2), hence we have a constraint

$$\hat{\zeta} - \mathbb{T} : \mathbb{D} = 0. \tag{4}$$

This is the only equation that can be used in the analysis of the relation between \mathbb{T} and \mathbb{D} . Unfortunately, there exist many functions \mathfrak{f} such that $\mathbb{T} =_{def} \mathfrak{f}(\mathbb{D})$ solves the scalar equation (4). (We at the moment ignore the additional restrictions that would follow form the requirement on the isotropy of the fluid.) Which one is the right one?

The concept of maximisation of the entropy production indicates that the right formula $\mathbb{T} = \mathfrak{g}(\mathbb{D})$ is the outcome of the following maximisation procedure. Take fixed \mathbb{T} , the *ansatz* for the entropy production $\widehat{\zeta}$, and *maximise* the entropy production $\widehat{\zeta}$ with respect to \mathbb{D} subject to constraint (4) and other imposed constraints. In our case, the other constraint is the incompressibility constraint,

$$\mathrm{Tr}\,\mathbb{D}=0.\tag{5}$$

Consequently, the auxiliary function for the constrained maximisation problem reads

$$\Phi =_{\text{def}} \widehat{\zeta} + \lambda_1 \left(\widehat{\zeta} - \mathbb{T} : \mathbb{D} \right) + \lambda_2 \operatorname{Tr} \mathbb{D}, \tag{6}$$

where λ_1 and λ_2 are Lagrange multipliers. The condition on the extremum reads

$$\frac{\partial \widehat{\zeta}}{\partial \mathbb{D}} + \lambda_1 \left(\frac{\partial \widehat{\zeta}}{\partial \mathbb{D}} - \mathbb{T} \right) + \lambda_2 \mathbb{I} = 0, \tag{7}$$

which implies that

$$\mathbb{T} = \frac{\lambda_2}{\lambda_1} \mathbb{I} + \frac{1 + \lambda_1}{\lambda_1} \frac{\partial \widehat{\xi}}{\partial \mathbb{D}}.$$
(8)

The value of the fraction $\frac{1+\lambda_1}{\lambda_1}$ can be recovered by taking the product of (7) with \mathbb{D} , which, in virtue of the incompressibility condition (5), yields $\frac{1+\lambda_1}{\lambda_1} = \frac{\mathbb{T}:\mathbb{D}}{\frac{\partial \mathbb{C}}{\partial \mathbb{T}}:\mathbb{D}} = \frac{1}{2}$. Consequently the complete formula for the Cauchy stress tensor reads

$$\mathbb{T} = -\pi \mathbb{I} + 2\mu \mathbb{D},\tag{9}$$

where $\pi =_{def} -\frac{\lambda_1}{\lambda_2}$ denotes the pressure. The pressure is, as expected in the case of incompressible Navier–Stokes fluid, an additional unknown field quantity that must be determined by the solution of the whole system of governing equations. The benefit of the maximisation approach is that it automatically and explicitly indicates that the pressure is linked to the incompressibility constraint by the means of the corresponding Lagrange multiplier. In this sense, one gets a *clear interpretation of the pressure in incompressible fluid*.

Finally, one might note that the maximisation procedure has automatically lead us to the constitutive relation that describes *isotropic* fluid. This piece of information concerning the isotropy has not been supplied externally.

A lesson that comes from the analysis above is that the material behaviour can be fully specified by two *scalar* functions, the internal energy \hat{e} and the entropy production $\hat{\zeta}$. The constitutive relation that has *tensorial* character is a consequence of the choice of the two scalar functions and the concept of maximisation of the entropy production.

Complex example – incompressible Maxwell/Oldroyd-B fluid

A more complex example of the maximisation procedure is provided by the analysis of Maxwell/Oldroyd-B incompressible viscoelastic type models. Here the ambition is to derive these models without any explicit reference to the underlying microstructure of the material. In particular, if one wants to develop models for materials such as the asphalt concrete/binder, see Figure 1, one can hardly use microscopic concepts such as *conformation tensor* that are closely related to the *polymeric fluids*.

The evolution of the microstructure is on the phenomenological level accounted for using the concept of *natural configuration*, see Rajagopal and Srinivasa [2, 3] and Rajagopal and Srinivasa [27, 28]. Precise description of the use of this concept is, in the context of viscoelastic fluids, developed in Rajagopal and Srinivasa [4], while Maxwell/Oldroyd-B models are discussed in Málek et al. [29]. In principle, the basic idea is to virtually decompose the total deformation to a *dissipative* (viscous) part and the *elastic* part, see Figure 2. This decomposition is motivated by the corresponding one-dimensional spring-dashpot analogue for Maxwell fluid, and it in fact embodies the notion of *visco-elastic* material.



Figure 2: Viscoelastic Maxwell/Oldroyd-B type fluid – kinematics.

If the total deformation is seen as a composition of the two deformations, see Figure 2, then the total deformation gradient $\mathbb F$ can be written as

$$\mathbf{F} = \mathbb{F}_{\mathbf{\kappa}_{p(t)}} \mathbb{G},\tag{10}$$

where $\mathbb{F}_{\kappa_{p(t)}}$ and \mathbb{G} are the deformation gradients of the partial deformations. Motivated by the standard relation $\frac{d\mathbb{F}}{dt} = \mathbb{LF}$ between the spatial velocity gradient $\mathbb{L} =_{def} \nabla v$ and the deformation gradient \mathbb{F} , one introduces new tensorial quantities $\mathbb{L}_{\kappa_{p(t)}}$ and $\mathbb{D}_{\kappa_{p(t)}}$ defined as

$$\mathbb{L}_{\kappa_{p(t)}} =_{\text{def}} \frac{\mathrm{d}\mathbb{G}}{\mathrm{d}t} \mathbb{G}^{-1}, \qquad \mathbb{D}_{\kappa_{p(t)}} =_{\text{def}} \frac{1}{2} \left(\mathbb{L}_{\kappa_{p(t)}} + \mathbb{L}_{\kappa_{p(t)}}^{\mathsf{T}} \right). \tag{11}$$

The material time derivative of the left Cauchy–Green tensor $\mathbb{B}_{\kappa_{p(t)}} =_{def} \mathbb{F}_{\kappa_{p(t)}} \mathbb{F}_{\kappa_{p(t)}}^{\mathsf{T}}$ associated to the instantaneous elastic (non-dissipative) response¹ then reads

$$\frac{\mathrm{d}\mathbb{B}_{\mathbf{\kappa}_{p(t)}}}{\mathrm{d}t} = \mathbb{L}\mathbb{B}_{\mathbf{\kappa}_{p(t)}} + \mathbb{B}_{\mathbf{\kappa}_{p(t)}}\mathbb{L}^{\mathsf{T}} - 2\mathbb{F}_{\mathbf{\kappa}_{p(t)}}\mathbb{D}_{\mathbf{\kappa}_{p(t)}}\mathbb{F}_{\mathbf{\kappa}_{p(t)}}^{\mathsf{T}}.$$
(12)

The last formula reduces, using the definition of the upper convected derivative $\overset{\bigtriangledown}{\mathbb{A}} =_{\text{def}} \frac{d\mathbb{A}}{dt} - \mathbb{L}\mathbb{A} - \mathbb{A}\mathbb{L}^{\top}$, to the formula

$$\overline{\mathbb{B}}_{\kappa_{p(t)}}^{\nabla} = -2\mathbb{F}_{\kappa_{p(t)}}\mathbb{D}_{\kappa_{p(t)}}\mathbb{F}_{\kappa_{p(t)}}^{\mathsf{T}}.$$
(13)

This formula enables one to take time derivatives of $\mathbb{B}_{\kappa_{p(t)}}$ which in turn enables one to derive a counterpart of (1), provided that one has an *ansatz* for the internal/free energy.

Since $\mathbb{B}_{\kappa_{p(t)}}$ represents the elastic (non-dissipative) part of the response, it should naturally enter the formula for the internal/free energy of the material. The reason is that the energy storage ability is in *finite elasticity theory* described in terms of the left Cauchy–Green tensor $\mathbb{B} =_{def} \mathbb{F}\mathbb{F}^{\top}$. But in our case only a part of the total deformation is non-dissipative/elastic. Consequently, only $\mathbb{B}_{\kappa_{p(t)}}$ instead of \mathbb{B} should take the role of an additional variable in the formula for the internal/free energy.

This analysis motivates the choice of the free energy in the form

$$\widehat{\Psi} =_{\text{def}} - \overline{c_{\text{V}}}^{\text{iNSE}} \Theta \left(\ln \left(\frac{\Theta}{\Theta_{\text{ref}}} \right) - 1 \right) + \frac{\mu}{2\rho} \left(\text{Tr} \,\mathbb{B}_{\kappa_{p(t)}} - 3 - \ln \det \mathbb{B}_{\kappa_{p(t)}} \right), \tag{14}$$

where θ denotes the temperature, θ_{ref} is a fixed reference temperature, ρ denotes the constant density of the fluid, and \overline{cv}^{iNSE} and μ are positive constants. The first term in (14) represents the standard thermal contribution, while the second term in (14) is motivated by formulae for free energy frequently used in the theory of finite elasticity. (See for example Saccomandi [34], Horgan and Saccomandi [35] or Horgan and Murphy [36] and references therein for a list of stored energies used in the theory of elasticity.) Having specified formula (14), we have *determined the energy storage mechanisms in the fluid*.

Straightforward differentiation then allows one to identify the entropy production, that is the counterpart of (2). In the present case one gets

$$\zeta = \left[\left(\mathbb{T} - \mu \left(\mathbb{B}_{\kappa_{p(t)}} \right)_{\delta} \right) : \mathbb{D} + \mu \left(\mathbb{C}_{\kappa_{p(t)}} - \mathbb{I} \right) : \mathbb{D}_{\kappa_{p(t)}} \right] - \frac{\mathbf{j}_{q} \bullet \nabla \theta}{\theta},$$
(15)

where $\mathbb{C}_{\kappa_{p(t)}} =_{\text{def}} \mathbb{F}_{\kappa_{p(t)}}^{\top} \mathbb{F}_{\kappa_{p(t)}}$. Finally, if we specify entropy production mechanism in the fluid via formula

$$\widehat{\boldsymbol{\zeta}} =_{\text{def}} 2\boldsymbol{\nu} \mathbb{D} : \mathbb{D} + 2\boldsymbol{\nu}_1 \operatorname{Tr} \left(\mathbb{F}_{\kappa_{p(t)}} \mathbb{D}_{\kappa_{p(t)}} \left(\mathbb{F}_{\kappa_{p(t)}} \mathbb{D}_{\kappa_{p(t)}} \right)^{\mathsf{T}} \right) + \kappa \frac{|\nabla \boldsymbol{\theta}|^2}{\boldsymbol{\theta}}, \tag{16}$$

where v, v_1 and κ are positive constants, then we get a counterpart of (3), and we are ready to employ the maximisation procedure outlined in the previous section.

This time, we maximise entropy production with respect to \mathbb{D} , $\mathbb{D}_{\kappa_{p(t)}}$ and $\nabla \theta$, and we again take into account the additional constraint of incompressibility. (See Málek et al. [29] for details.) The procedure finally leads to the constitutive relations

$$\mathbb{T} = -\pi \mathbb{I} + \mathbb{T}_{\delta}, \qquad \mathbb{T}_{\delta} = 2\nu \mathbb{D} + \mu \big(\mathbb{B}_{\kappa_{p(t)}} \big)_{\delta}, \qquad (17a)$$

¹The notion of instantaneous elastic response is related to the behaviour of the material in creep and stress relaxation tests. This requires one to investigate the response of *nonlinear* systems governed by differential equations to stimuli with jump *discontinuities*. Convenient mathematical tools applicable in this setting have been discussed in Průša and Rajagopal [30, 31], Řehoř et al. [32] and Průša et al. [33].

$$\boldsymbol{j}_q = -\boldsymbol{\kappa} \nabla \boldsymbol{\theta}. \tag{17b}$$

Moreover, it turns out that the right hand side of (13) can be expressed in terms of $\mathbb{B}_{\kappa_{p(t)}}$ only, namely

$$\mathbf{v}_1 \overline{\mathbb{B}_{\kappa_{p(t)}}}^{\vee} + \mu \left(\mathbb{B}_{\kappa_{p(t)}} - \mathbb{I} \right) = \mathbb{O}.$$
(17c)

The constitutive relations are then exploited in the balance equations which leads to the governing equations used in the seminal paper by Oldroyd [37]. Indeed, upon defining $\widetilde{\mathbb{S}} =_{\text{def}} \mu (\mathbb{B}_{\kappa_{p(t)}} - \mathbb{I}) + 2\nu \mathbb{D}$ and $p =_{\text{def}} \pi + \frac{\mu}{3} \text{Tr} \mathbb{B}_{\kappa_{p(t)}} - \mu$, we end up with constitutive relations

$$\mathbb{T} = -p\mathbb{I} + \widetilde{\mathbb{S}},\tag{18a}$$

$$\mathbf{v}_{1}\overline{\left(\frac{\widetilde{\mathbb{S}}}{\mu}\right)} + \widetilde{\mathbb{S}} = 2\left(\mathbf{v}_{1} + \mathbf{v}\right)\mathbb{D} + 2\mathbf{v}_{1}\overline{\left(\frac{\mathbf{v}\mathbb{D}}{\mu}\right)},\tag{18b}$$

which is exactly the form used by Oldroyd [37].

Having commented on the most important steps in the derivation of Maxwell/Oldroyd-B model, let us now summarise the benefits of the current approach:

- 1. Quantity $\mathbb{B}_{\kappa_{p(t)}}$ has a direct interpretation in terms of kinematics of the material of interest. In particular, there is no need to use the concept of conformation tensor or vaguely interpret the additional tensorial variable as a convenient internal parameter.
- 2. The evolution equation for $\mathbb{B}_{\kappa_{p(i)}}$ is a direct consequence of the assumed kinematics of evolving natural configuration. In particular, the upper convected derivative appears naturally in the governing equations.
- 3. The pressure is interpreted in terms of Lagrange multiplier enforcing the incompressibility constraint.

Note that by changing the *ansatz* for the specific Helmholtz free energy $\widehat{\Psi}$ and entropy production $\widehat{\zeta}$, one can clearly develop models that can describe various types of material response. Moreover, the underlying kinematics can be more involved than that shown in Figure 2, more complex spring-dashpot analogues can be used as well, see for example Karra and Rajagopal [38].

Conclusion

The concept of *maximisation of the rate of entropy production* and the concept of *evolving natural configuration* are useful concepts in the construction of mathematical models (constitutive relations) for the response of complex materials. Remarks on the links between the concept of *maximisation of the rate of entropy production* and other concepts in non-equilibrium thermodynamics such as *gradient dynamics* can be found for example in the recent work by Janečka and Pavelka [39].

The introduced concepts are of importance especially in the settings where the specification of constitutive relations requires one to use purely phenomenological arguments, which is the case in many engineering applications. Some of the successful applications in the context of viscoelastic materials have been discussed in this abstract. References to other applications, such as nonlocal field theories, see for example Heida and Málek [40] and Heida [41], Heida et al. [42], can be found in review works Málek and Rajagopal [43] and Málek and Průša [44].

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