



## Objective time derivatives in nonequilibrium thermodynamics

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**Abstract.** In this paper we outline a framework of a thermodynamic theory where objective time derivatives appear in a natural way. The entropy production of a single component fluid with a tensorial internal variable is calculated as an example. Dependence on material quantities leads to objective derivatives in the constitutive relations resulting in a new rheological model. The viscosity and the viscometric functions are calculated for simple shear.

**Key words:** nonequilibrium thermodynamics, rheology, objective time derivatives, simple shear, Second Law.

### 1. INTRODUCTION

Frame independence is one of the fundamental issues in continuum physics. We believe that a true theory of nature should be independent of an observer. The observer independence is usually introduced through invariance of special transformations. However, one can give a concept of spacetime that is inherently independent of observers [1]. Based on that observer independent formalism one can resolve several old and new observer related paradoxes both in special relativistic and nonrelativistic spacetimes [2–5].

Recently we investigated the objective time derivatives of continuum physics from this point of view. We compared the usual, transformation based definition of objectivity [6] to the observer independent approach of Matolcsi and formulated the usual three dimensional objective time derivatives as projections of four dimensional Lie derivatives along the flow of a continuum. The comparison revealed several important differences, e.g. the nonrelativistic four-velocity  $(1, \mathbf{v})$ , which is the time derivative of the four vector  $(t, \mathbf{r}(t))$ , is an objective, observer independent quantity contrary to its spacelike projection, the relative velocity  $\mathbf{v}$  [7].

Objective time derivatives are common in rheology, but they appear in an ad-hoc way, independently of a thermodynamic theory. One of the exceptions is the approach of GENERIC [8], where the objective time derivatives appear as a consequence of the special structure of the theory, through the Jacobi identity.

Another exception is the approach of Verhás [9], which introduces the entropy balance through a corotational time derivative of the entropy function.

In the following we avoid the formalism of the nonrelativistic spacetime model and investigate some consequences with the help of the usual relative and three dimensional notions. In this paper we will show how objective time derivatives can appear in nonequilibrium thermodynamics.

In the next section we present the necessary notions and especially the formulas of objective time derivatives derived in a frame independent approach [10]. In the third section we introduce material fields and assume that the material relations are defined through them. Then we derive the entropy production for fluids with a single tensorial internal variable (dynamic degree of freedom). We define the material form of thermodynamic fluxes and forces in the entropy production and give the linear conductivity relations among them. Then we get a differential equation, a new rheological model, that is a thermodynamic and frame independent modification of the traditional Jeffreys model. In the last section we solve the equation for stationary simple shear and calculate the nonlinear viscosity and the viscometric functions.

### 2. OBJECTIVE TIME DERIVATIVES

The most important consequences of the observer independent nonrelativistic spacetime formalism are the following:

1. Four vectors and tensors are unavoidable, because the spacetime is not a simple Descartes product of time and space.
2. Vectors and covectors should be strictly distinguished, because there is no frame independent identification for them, contrary to the relativistic theories.

Using four vectors and covectors is particularly easy and natural in nonrelativistic spacetime with relative, frame dependent notions. The four motion can be given as  $(t, \chi_t(\mathbf{R}))$ , where  $\mathbf{R}$  is the position in the material frame (reference configuration) and  $\chi_t(\mathbf{R})$  motion [11]. Therefore the four-deformation gradient is

$$\Upsilon_t(\mathbf{R}) = D_{\mathbf{R}}(t, \chi_t(\mathbf{R})) = \begin{pmatrix} 1 & \mathbf{0} \\ \mathbf{v}_t(\mathbf{R}) & \nabla_{\mathbf{R}}\chi_t(\mathbf{R}) \end{pmatrix},$$

where  $\mathbf{F}_t = \nabla_{\mathbf{R}}\chi_t$  is the usual three dimensional deformation gradient (direct motion gradient) [11]. It is a derivative of a vector field, therefore it is a mixed second order tensor field. That is  $\mathbf{F}_t \in \mathbf{E} \otimes \mathbf{E}^*$ , where  $\mathbf{E}$  is the three dimensional vector field of the space (relative to an inertial observer) and the star denotes its dual. Recognizing these familiar components in the relative four-deformation gradient tensor we can understand how the four-notions can be avoided nonrelativistically. In the notation we will avoid them completely.

The vector and tensor quantities do not reflect material relations, therefore we pull them back to the reference configuration and we call the corresponding vectors and tensors *material*.

- The material form of the scalar field  $f(\mathbf{r}, t) \in \mathbb{R}$  is

$$f_t(\mathbf{R}) = f(\chi_t(\mathbf{R}), t). \quad (1)$$

- The material form of the vector field  $\mathbf{a}(\mathbf{r}, t) \in \mathbf{E}$  is

$$\mathbf{a}_A = \mathbf{a}_t(\mathbf{R}) = (\mathbf{F}^{-1}\mathbf{a})(\chi_t(\mathbf{R}), t). \quad (2)$$

- The material form of the covector field  $\mathbf{b}(\mathbf{r}, t) \in \mathbf{E}^*$  is

$$\mathbf{b}_A = \mathbf{b}_t(\mathbf{R}) = (\mathbf{F}^*\mathbf{b})(\chi_t(\mathbf{R}), t). \quad (3)$$

- The material form of the tensor field  $\mathbf{A}(\mathbf{r}, t) \in \mathbf{E} \times \mathbf{E}$  is

$$\mathbf{A}_A = \mathbf{A}_t(\mathbf{R}) = (\mathbf{F}^{-1}\mathbf{A}(\mathbf{F}^{-1})^*)(\chi_t(\mathbf{R}), t). \quad (4)$$

- The material form of the cotensor field  $\mathbf{B}(\mathbf{r}, t) \in \mathbf{E}^* \times \mathbf{E}^*$  is

$$\mathbf{B}_A = \mathbf{B}_t(\mathbf{R}) = (\mathbf{F}^*\mathbf{B}\mathbf{F})(\chi_t(\mathbf{R}), t). \quad (5)$$

- The material form of the mixed tensor field  $\mathbf{C}(\mathbf{r}, t) \in \mathbf{E} \times \mathbf{E}^*$  is

$$\mathbf{C}_A = \mathbf{C}_t(\mathbf{R}) = (\mathbf{F}^{-1}\mathbf{C}\mathbf{F})(\chi_t(\mathbf{R}), t). \quad (6)$$

Here we denoted the material form shortly with the subscript  $A$ . The material time derivative of a physical

quantity is the time derivative of its material form. From a mathematical point of view material time derivatives are the Lie derivatives according to the flow of the continuum. Therefore

- The material time derivative of the scalar field  $f(\mathbf{r}, t) \in \mathbb{R}$  is its substantial derivative

$$\frac{d}{dt}f_t(\mathbf{R}) = \dot{f} = \partial_t f + \mathbf{v} \cdot \nabla f. \quad (7)$$

- The material time derivative of the vector field  $\mathbf{a}(\mathbf{r}, t) \in \mathbf{E}$  is its upper convected derivative

$$\mathbf{a}^\diamond = \frac{d}{dt}\mathbf{a}_t(\mathbf{R}) = \mathbf{F}^{-1}\dot{\mathbf{a}} - \mathbf{F}^{-1}\dot{\mathbf{F}}\mathbf{F}^{-1}\mathbf{a} = (\dot{\mathbf{a}} - \nabla\mathbf{v}\mathbf{a})_A. \quad (8)$$

- The material time derivative of the covector field  $\mathbf{b}(\mathbf{r}, t) \in \mathbf{E}^*$  is its lower convected derivative

$$\mathbf{b}^\diamond = \frac{d}{dt}\mathbf{b}_t(\mathbf{R}) = \mathbf{F}^*\dot{\mathbf{b}} + \dot{\mathbf{F}}^*\mathbf{b} = (\dot{\mathbf{b}} + (\nabla\mathbf{v})^*\mathbf{b})_A. \quad (9)$$

- The material time derivative of the tensor field  $\mathbf{A}(\mathbf{r}, t) \in \mathbf{E} \times \mathbf{E}$  is its upper convected derivative

$$\begin{aligned} \mathbf{A}^\diamond &= \frac{d}{dt}\mathbf{F}^{-1}\mathbf{A}(\mathbf{F}^{-1})^* \\ &= \mathbf{F}^{-1}\dot{\mathbf{A}}(\mathbf{F}^{-1})^* - \mathbf{F}^{-1}\dot{\mathbf{F}}\mathbf{F}^{-1}\mathbf{A}(\mathbf{F}^{-1})^* \\ &\quad - \mathbf{F}^{-1}\mathbf{A}(\mathbf{F}^{-1}\dot{\mathbf{F}}\mathbf{F}^{-1})^* \\ &= (\dot{\mathbf{A}} - (\nabla\mathbf{v})\mathbf{A} - \mathbf{A}(\nabla\mathbf{v})^*)_A. \end{aligned} \quad (10)$$

- The material derivative of the cotensor field  $\mathbf{B}(\mathbf{r}, t) \in \mathbf{E}^* \times \mathbf{E}^*$  is

$$\begin{aligned} \mathbf{B}^\diamond &= \frac{d}{dt}\mathbf{F}^*\mathbf{B}\mathbf{F} = \mathbf{F}^*\dot{\mathbf{A}}\mathbf{F} + \dot{\mathbf{F}}^*\mathbf{B}\mathbf{F} + \mathbf{F}^*\mathbf{A}\dot{\mathbf{F}} \\ &= (\dot{\mathbf{B}} + (\nabla\mathbf{v})^*\mathbf{B} + \mathbf{B}(\nabla\mathbf{v}))_A. \end{aligned} \quad (11)$$

- The material derivative of the mixed tensor field  $\mathbf{C}(\mathbf{r}, t) \in \mathbf{E} \times \mathbf{E}^*$  is

$$\begin{aligned} \mathbf{C}^\diamond &= \frac{d}{dt}\mathbf{F}^{-1}\mathbf{C}\mathbf{F} = \mathbf{F}^{-1}\dot{\mathbf{C}}\mathbf{F} - \mathbf{F}^{-1}\dot{\mathbf{F}}\mathbf{F}^{-1}\mathbf{C}\mathbf{F} + \mathbf{F}^{-1}\mathbf{C}\dot{\mathbf{F}} \\ &= (\dot{\mathbf{C}} - (\nabla\mathbf{v})\mathbf{C} + \mathbf{C}(\nabla\mathbf{v}))_A. \end{aligned} \quad (12)$$

Here we have used that  $(\mathbf{F}^{-1})^\cdot = -\mathbf{F}^{-1}\dot{\mathbf{F}}\mathbf{F}^{-1}$  and  $\nabla\mathbf{v} = \dot{\mathbf{F}}\mathbf{F}^{-1}$ , therefore  $\nabla\mathbf{v}^* = (\mathbf{F}^{-1})^*\dot{\mathbf{F}}^*$ .

These are the ordinary objective time derivatives from the traditional three dimensional point of view, too. However, from the perspective of the four dimensional formalism the material time derivative of a three vector is a three vector but a material time derivative of a three-covector is a four-covector [10]. Therefore spacelike material time derivatives are not objective in a four dimensional sense in general. However, if the velocity field is independent of time, then the material time derivative of a four-covector is spacelike, therefore for

*stationary flows* the above traditional objective time derivatives are objective from the point of view of observer independent formulation, too.

The material time derivatives of the motion related quantities are special. For example, the material time derivative of the deformation gradient is its substantial time derivative

$$\mathbf{F}^\diamond = \frac{d}{dt} \mathbf{F}^{-1} \mathbf{F} (\mathbf{F}^{-1})^* = \dot{\mathbf{F}}. \quad (13)$$

For further details about the material forms and their time derivatives in a true frame independent framework see [10]. In the following we will see how these formulas are related to the Second Law of thermodynamics.

### 3. BASIC BALANCES AND CONSTITUTIVE THEORY

In thermodynamic rheology finite strain mechanics with internal variables are considered. In this demonstrative example we restrict ourselves to a single internal variable where the basic state space is spanned by the specific internal energy  $e$ , the deformation gradient  $\mathbf{F}$ , and a tensorial internal variable of state  $\xi$ .

The balance of momentum reads (in the absence of body forces) [11]

$$\rho \dot{\mathbf{v}} - \nabla \cdot \mathbf{t} = \mathbf{0}, \quad (14)$$

where  $\mathbf{v} = \frac{\partial \chi}{\partial t} |_{\mathbf{R}}$  is the velocity field, the dot above the velocity denotes the substantial time derivative, and  $\mathbf{t}$  is the Cauchy stress. The density  $\rho$  is not independent of the deformation gradient, because the density in the reference configuration is  $\rho_0 = \rho \det \mathbf{F} = \text{const}$ .

We can get the balance of kinetic energy by multiplying (14) with the velocity and reordering the terms

$$\rho \mathbf{v} \cdot \dot{\mathbf{v}} - \mathbf{v} \cdot \nabla \cdot \mathbf{t} = \rho \frac{d}{dt} \left( \frac{\mathbf{v}^2}{2} \right) - \nabla \cdot (\mathbf{v} \cdot \mathbf{t}) + \mathbf{t} : \nabla \mathbf{v} = \mathbf{0}. \quad (15)$$

The balance of internal energy can be calculated as the difference of the conserved total energy and the kinetic energy and is given as (e.g. [9])

$$\rho \dot{e} + \nabla \cdot \mathbf{q} = \mathbf{t} : \nabla \mathbf{v}. \quad (16)$$

Here  $\mathbf{q}$  is the heat flux.

Constitutive functions characterize the material and only the material. The Second Law is the cornerstone of any material theory. The entropy is a scalar material function that depends on the basic fields of the corresponding continua. Our basic assumption regarding the constitutive theory is that *entropy depends on the material form of the fields*, particularly  $(e_A, \mathbf{F}_A, \xi_A)$ . Then the time derivative of the entropy is

$$\dot{s}(e_A, \mathbf{F}_A, \xi_A) = \frac{\partial s}{\partial e_A} e^\diamond + \frac{\partial s}{\partial \mathbf{F}_A} \mathbf{F}^\diamond + \frac{\partial s}{\partial \xi} \xi^\diamond.$$

The objective time derivatives appear. However, the objective time derivative of a scalar and of the deformation gradient is the substantial time derivative, as we have seen in (7) and (13); therefore, we may substitute the balance of the internal energy (16). Then we get the usual form of the entropy production  $\sigma_s$ , with the material time derivative of the internal variable as (see e.g. [9])

$$\begin{aligned} \sigma_s &= \frac{1}{T^2} \mathbf{q} \cdot \nabla T + \frac{1}{T} \left( \mathbf{t} + \rho T \mathbf{F} \frac{\partial s}{\partial \mathbf{F}_A} \right) : \nabla \circ \mathbf{v} - \rho \frac{\partial s}{\partial \xi} : \xi^\diamond \\ &\geq 0. \end{aligned} \quad (17)$$

Here the entropy flux  $\mathbf{j}_s$  is assumed to have its classical form

$$\mathbf{j}_s = \frac{\mathbf{q}}{T}. \quad (18)$$

The thermodynamic fluxes and forces can be determined from the entropy production. The fluxes are unknown functions of the basic fields and their derivatives [12]. For heat conduction and for mechanical interaction the fluxes are the heat flux  $\mathbf{q}$  and the viscous stress  $\mathbf{F}^v = (\mathbf{t} + \rho T \mathbf{F} \frac{\partial s}{\partial \mathbf{F}_A})$ . In the last term of (17) the material time derivative indicates an unknown differential equation for the internal variable, therefore in the last term the thermodynamic flux is  $\xi^\diamond$  [9].

The relation of the thermodynamic forces and fluxes is constitutive, therefore the forces and fluxes themselves are material quantities. However, we will assume that the material manifold is the current configuration. This is a reasonable and generally accepted assumption in case of fluids. Therefore, the distinction will be important only in case of the *derivatives* of the material quantities.

Moreover, in the framework of a non-relativistic spacetime model we should pay particular attention to distinguishing between vectors and covectors. In this regard there is an uncertainty in the middle, mechanical term. The stress is a tensor in (14), but the velocity gradient is a mixed tensor field. However, the product of a tensor and a mixed tensor is a tensor and we cannot form its trace without any further ado. The situation can be clarified inspecting the way how we have got the balance of kinetic energy (15). There the product of the balance of momentum and velocity – two contravariant vectors – appeared. From the point of view of frame independent formulation we have exploited the identification of vectors and covectors in the vector space of relative velocities that is endowed by a scalar product. With this observation we can understand that the velocity gradient in (15) and therefore in (17) represents a cotensor field.

The distinction is important, because the thermodynamic forces and fluxes are material forms and the entropy production can be written with these quantities as

$$\sigma_s = \frac{1}{T^2} \mathbf{q}_A \cdot (\nabla T)_A + \mathbf{t}_A^v : (\nabla \circ \mathbf{v})_A - \rho \frac{\partial s}{\partial \xi_A} : \xi^\diamond \geq 0. \quad (19)$$

Here the viscous stress is a tensor field, therefore its material form, the thermodynamic flux of the mechanical interaction is

$$\mathbf{t}_A^v = \left( \mathbf{t} + \rho T \mathbf{F} \frac{\partial s}{\partial \mathbf{F}_A} \right)_A = \mathbf{F}^{-1} \mathbf{t}^v \mathbf{F}^{-1*}.$$

The thermodynamic force of the mechanical interaction, the material form of the velocity gradient  $\gamma = \nabla \circ \mathbf{v}$  as a cotensor field is

$$\gamma_A = \mathbf{F}^* \gamma \mathbf{F}. \quad (20)$$

In the following we do not pay attention to thermal interaction and assume that  $\mathbf{q} = \mathbf{0}$ . The linear relationship between the material thermodynamic forces and fluxes gives

$$\mathbf{t}_A^v = \mathbf{L}_1 \gamma_A - \mathbf{L}_{12} \rho \xi_A, \quad (21)$$

$$\xi^{\diamond} = \mathbf{L}_{21} \gamma_A - \mathbf{L}_2 \rho \xi_A. \quad (22)$$

For the sake of convenience the conductivity tensors  $\mathbf{L}_1$ ,  $\mathbf{L}_{12}$ ,  $\mathbf{L}_{21}$  and  $\mathbf{L}_2$  are considered as constants. We do not assume any symmetry relations.

One cannot eliminate the internal variable from (21)–(22) without any further ado, because the objective time derivative does not commute with constant matrix multiplication. Moreover, one should be careful in case of isotropic materials, too.

Now we assume that the material is isotropic and the internal variable is a symmetric, traceless second order tensor. Therefore the coupling is reduced to the symmetric, traceless parts of the thermodynamic forces and currents, according to the representation theorems of isotropic tensors (Curie principle). Applying the previously mentioned identification of the material and current configuration (21)–(22) is reduced to the following form

$$(\mathbf{t}^v)^{S0} = l_1 \gamma^{S0} - l_{12} \rho \xi^{S0}, \quad (23)$$

$$(\xi^{\diamond})^{S0} = l_{21} \gamma^{S0} - l_2 \rho \xi^{S0}. \quad (24)$$

Here we have denoted the symmetric traceless parts of the corresponding tensors with the superscripts  $S0$ .

#### 4. STATIONARY SIMPLE SHEAR

Simple shear is one of the basic setups for testing the rheological models. It is characterized by the following velocity gradient

$$\gamma = \begin{pmatrix} 0 & \kappa & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad (25)$$

where  $\kappa$  is the constant shear rate. Let us introduce a second order symmetric traceless tensor  $\xi$  as internal

variable with the following components in the reference frame where the velocity gradient is (25)

$$\xi = \begin{pmatrix} \xi_1 & \xi_{12} & \xi_{13} \\ \xi_{12} & \xi_2 & \xi_{23} \\ \xi_{13} & \xi_{23} & -\xi_1 - \xi_2 \end{pmatrix}.$$

The material derivative of  $\xi$  is

$$\xi^{\diamond} = -\xi \gamma^* - \gamma \xi = -\kappa \begin{pmatrix} 2\xi_{12} & \xi_2 & \xi_{23} \\ \xi_2 & 0 & 0 \\ \xi_{23} & 0 & 0 \end{pmatrix}.$$

Substituting the above relations into (23)–(24) we get the following solution for the components of the viscous stress and the internal variable

$$t_{13} = t_{31} = t_{23} = t_{32} = 0, \quad (26)$$

$$t_{12} = t_{21} = \frac{\kappa}{2} \left( l_1 - \frac{3l_2 l_{12} l_{21}}{2\kappa^2 + 3l_2^2} \right), \quad (27)$$

$$t_1 = -\frac{2\kappa^2 l_{12} l_{21}}{2\kappa^2 + 3l_2^2} = -2t_2, \quad (28)$$

$$\xi_{13} = \xi_{31} = \xi_{23} = \xi_{32} = 0, \quad (29)$$

$$\xi_{12} = \xi_{21} = \frac{3\kappa l_2 l_{21}}{4\kappa^2 + 6l_2^2}, \quad (30)$$

$$\xi_1 = \frac{2\kappa^2 l_{21} l_{21}}{2\kappa^2 + 3l_2^2} = -2\xi_2. \quad (31)$$

From the stresses one can get the nonlinear viscosity and the corresponding viscometric functions as

$$\hat{\eta} = \frac{t_{12}}{\kappa} = \frac{1}{2} \left( l_1 - \frac{3l_2 l_{12} l_{21}}{2\kappa^2 + 3l_2^2} \right), \quad (32)$$

$$\Psi_1 = -\frac{t_1 - t_1}{\kappa^2} = \frac{3l_{12} l_{21}}{2\kappa^2 + 3l_2^2}, \quad (33)$$

$$\Psi_2 = -\frac{t_2 - t_3}{\kappa^2} = 0. \quad (34)$$

The usual rheological relaxation times and viscosity coefficients originating from the Jeffreys model can be introduced as [9]

$$\tau = \frac{1}{\rho l_2}, \quad \eta = \frac{l_1 l_2 - l_{12} l_{21}}{2l_2}, \quad \tau_d = \frac{l_1}{\rho(l_1 l_2 - l_{12} l_{21})}.$$

Due to the nonnegativity of the entropy production  $\tau$ ,  $\eta$ , and  $\tau_d$  are nonnegative. Therefore the viscosity and the viscometric functions can be given equivalently in a more familiar form as

$$\hat{\eta} = \eta \frac{3 - 2\rho^2 \tau \tau_d \kappa^2}{3 + 2\rho^2 \tau^2 \kappa^2}, \quad (35)$$

$$\Psi_1 = 6\eta \rho \frac{\tau - \tau_d}{3 + 2\rho^2 \tau^2 \kappa^2}, \quad (36)$$

$$\Psi_2 = 0. \quad (37)$$

The viscosity and the first viscometric functions are qualitatively similar to the corresponding functions of other simple rheological models, like the corotational Jeffreys. The remarkable property of our model is the second viscometric function. According to the experiments, the second viscometric function is small compared to the first one [13,14]. One would expect that the simple linear viscoelastic models would reproduce that behaviour. However, they will not. The corotational Jeffreys model gives  $\Psi_2 = -\Psi_1/2$ , the upper convected Maxwell model gives  $\Psi_2 = 0$ . However, the upper convected Maxwell model predicts a shear rate independent viscosity.

As we have emphasized in the introduction, the given three dimensional time derivatives are frame independent only for steady flows. A comparison to time dependent experiments requires further investigations.

## 5. CONCLUSIONS

We have given an outline of the thermodynamic framework where objective time derivatives appear in a natural way. Our basic assumption was that the entropy should be the function of the material form of the basic state variables. We tested our assumption with deriving the simplest possible linear viscoelastic rheological model.

Contrary to the common practice in rheology, where one can introduce different time derivatives into the differential equation of the model in an ad-hoc way, our approach results in a unique form. Due to a strict distinction of vectors and covectors we have got a new rheological model. The solution of the model equations for stationary simple shear resulted in realistic results for the viscosity and for the viscometric functions.

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## Materiaalsed ajatuletised pöördumatute protsesside termodünaamikas

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On visandatud raamistik termodünaamika teooriale, milles materiaalsed tuletised ajast tekivad loomulikul viisil. Näitena on arvatud tensoriaalse sisemuutujaga ühekomponentse vedeliku entroopia juurdekasv. Materjali omaduste vastastikune sõltuvus tekitab materjali olekut kirjeldavates seostes materiaalseid tuletisi, mis viivad uue reoloogilise mudelini. Puhta nihke olukorra puhul on arvatud viskoossus ja leitud viskomeetrilised funktsioonid.