

Article

Implicit Type Constitutive Relations for Elastic Solids and Their Use in the Development of Mathematical Models for Viscoelastic Fluids

Vít Průša ¹  and K. R. Rajagopal ^{2,*} 

¹ Faculty of Mathematics and Physics, Charles University, Sokolovská 83, 186 75 Praha 8, Czech Republic; prusv@karlin.mff.cuni.cz

² Department of Mechanical Engineering, Texas A&M University, College Station, TX 77843-3123, USA

* Correspondence: krajagopal@tamu.edu

Abstract: Viscoelastic fluids are non-Newtonian fluids that exhibit both “viscous” and “elastic” characteristics in virtue of the mechanisms used to store energy and produce entropy. Usually, the energy storage properties of such fluids are modeled using the same concepts as in the classical theory of nonlinear solids. Recently, new models for elastic solids have been successfully developed by appealing to implicit constitutive relations, and these new models offer a different perspective to the old topic of the elastic response of materials. In particular, a sub-class of implicit constitutive relations, namely relations wherein the left Cauchy–Green tensor is expressed as a function of stress, is of interest. We show how to use this new perspective in the development of mathematical models for viscoelastic fluids, and we provide a discussion of the thermodynamic underpinnings of such models. We focus on the use of Gibbs free energy instead of Helmholtz free energy, and using the standard Giesekus/Oldroyd-B models, we show how the alternative approach works in the case of well-known models. The proposed approach is straightforward to generalize to more complex settings wherein the classical approach might be impractical or even inapplicable.

Keywords: viscoelastic fluids; constitutive relations; thermodynamics; Hencky strain; Gibbs free energy; Helmholtz free energy

MSC: 76A05



Citation: Průša, V.; Rajagopal, K.R. Implicit Type Constitutive Relations for Elastic Solids and Their Use in the Development of Mathematical Models for Viscoelastic Fluids. *Fluids* **2021**, *6*, 131. <https://doi.org/10.3390/fluids6030131>

Academic Editor: Laura A. Miller

Received: 22 February 2021

Accepted: 18 March 2021

Published: 22 March 2021

Publisher’s Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

Contents

1 Introduction	2
2 Materials Specified via Implicit Constitutive Relations	3
2.1 Fluids	3
2.2 Solids	4
3 Viscoelastic Rate-Type Fluids	5
4 Gibbs Free Energy-Based Approach to Viscoelastic Rate-Type Fluids	5
4.1 Entropy Evolution Equation—General Case	6
4.2 Entropy Evolution Equation—Incompressible Fluids	9
4.2.1 Manipulations with Entropy Production—Direct Use of Gibbs Free Energy	9
4.2.2 Manipulations with Entropy Production—Indirect Use of Gibbs Free Energy	10
4.2.3 Constitutive Relations	11
4.2.4 Temperature Evolution Equation	11

5 Example—Giesekus/Oldroyd-B Viscoelastic Rate-Type Fluid in the Approach Based on the Gibbs Free Energy **11**

5.1 Giesekus/Oldroyd-B Model via Helmholtz Free Energy—Classical Approach 12

5.2 Approach Based on Gibbs Free Energy 14

 5.2.1 Oldroyd-B Model 15

 5.2.2 Giesekus Model 19

5.3 Approach Based on Gibbs Free Energy and Entropy Production Maximization 20

 5.3.1 Oldroyd-B Model 21

 5.3.2 Giesekus Model 23

6 Conclusions **24**

A Daleckii–Krein Formula and Its Consequences **25**

References **26**

1. Introduction

Most of the classical treatises on continuum mechanics when discussing constitutive theory start with the assumption that stress is given as a function or a functional of the deformation; see for example Truesdell and Noll [1], Müller [2], Coleman and Noll [3]. However, it has been argued that this approach is too restrictive; see especially Rajagopal [4,5].

For example, it turns out that it is worthwhile to generalize the classical incompressible Navier–Stokes fluid model $\mathbb{T} = -p\mathbb{I} + 2\nu\mathbb{D}$ as

$$\mathbb{T} = -p\mathbb{I} + \mathbb{T}_\delta, \tag{1a}$$

$$\mathbb{D} = \mathfrak{f}(\mathbb{T}_\delta), \tag{1b}$$

where we use the standard notation \mathbb{T} for the Cauchy stress tensor, p for the mechanical pressure, \mathbb{D} for the symmetric part of the velocity gradient and \mathfrak{f} for a tensorial function. This approach to the generalization of the classical incompressible Navier–Stokes fluid model is in contrast with the classical approach based on the formula

$$\mathbb{T} = -p\mathbb{I} + \mathfrak{g}(\mathbb{D}), \tag{2}$$

where \mathfrak{g} is a tensorial function (here, one can think of classical models for shear-thinning/thickening fluids). Note that even if the relation $\mathbb{D} = \mathfrak{f}(\mathbb{T}_\delta)$ is invertible and thus can be rewritten in the classical form $\mathbb{T}_\delta = \mathfrak{g}(\mathbb{D})$, it might be still convenient to use the representation (1b) because the representation (1b) might be much simpler than the classical one (meaning that the formula $\mathbb{D} = \mathfrak{f}(\mathbb{T}_\delta)$ might have a simple analytical form, while the inverse formula $\mathbb{T}_\delta = \mathfrak{g}(\mathbb{D})$ could be complicated or impossible to write down using elementary functions). If (1b) is not invertible, then the two approaches are clearly strikingly different. Furthermore, one can even go one step further and replace (1b) by a more general implicit relation $\mathfrak{k}(\mathbb{T}_\delta, \mathbb{D}) = \mathbb{O}$, where \mathfrak{k} is a tensorial function and \mathbb{O} denotes the zero tensor.

Similarly, in the context of mathematical models for elastic solids, one can generalize the standard isotropic Cauchy elastic materials, wherein one assumes that $\mathbb{T} = \mathfrak{h}(\mathbb{B})$, where \mathfrak{h} is a tensorial function and \mathbb{B} denotes the left Cauchy–Green tensor. The generalization of this formula is

$$\mathbb{B} = \mathfrak{i}(\mathbb{T}) \tag{3}$$

or even $\mathfrak{j}(\mathbb{T}, \mathbb{B}) = \mathbb{O}$, where \mathfrak{i} and \mathfrak{j} are again tensorial functions. Clearly, everything that has been said in the context of models for fluids holds as well for models for elastic solids.

Since viscoelastic fluids are non-Newtonian fluids that exhibit both “viscous” and “elastic” characteristics, it is desirable to determine how to exploit the concept of implicit type constitutive relations in the context of mathematical models for these fluids. In particular, the phenomenological theory of viscoelastic rate-type fluids is based on the assumption that the elastic response of the fluid is modeled using the same concepts as in the classical

theory of nonlinear solids (in particular, the assumed formulae for Helmholtz free energy borrow heavily from the classical theory of nonlinear solids). However, if one wants to model the elastic response in the spirit of recent advances in theory of nonlinear elastic solids that exploit constitutive relations of type (3), the phenomenological derivation of mathematical models for viscoelastic rate-type fluids must be modified accordingly. Such modifications are discussed—with a special emphasis on the corresponding thermodynamic basis—in the current contribution.

The paper is organized as follows. First, we briefly review the concept of constitutive relations for fluids and solids developed using the approaches (1) or (3) and its generalizations to the fully implicit setting $\mathfrak{k}(\mathbb{T}_\delta, \mathbb{D}) = \mathbb{O}$ or $\mathfrak{j}(\mathbb{T}, \mathbb{B}) = \mathbb{O}$; see Section 2. Then, we proceed with a brief review of the classical derivation of viscoelastic rate-type models based on the concept of the natural configuration and the characterization of the elastic response with Helmholtz free energy; see Section 3.

In Section 4 we identify the Hencky strain tensor as the convenient tensorial quantity and Gibbs free energy as the convenient thermodynamic potential that allows one to work with the (elastic) constitutive response of type (3), and we outline a general procedure that allows one to develop novel viscoelastic rate-type models based on these concepts. In Section 5, we study the classical Giesekus/Oldroyd-B model both in the classical framework and the newly proposed framework. For this model, it is possible to write down explicitly all formulae in both approaches, which allows us to clearly document the general theory. Finally, we also discuss the applicability of the maximization of the entropy production hypothesis—see Rajagopal and Srinivasa [6]—in the theory of constitutive relations.

2. Materials Specified via Implicit Constitutive Relations

Before we proceed with viscoelastic fluids, below, we briefly summarize the state-of-the-art regarding the novel approach to the mathematical modeling of viscous fluids and elastic solids.

2.1. Fluids

A simple generalization of the Navier–Stokes fluid as indicated in (1) is the constitutive relation in the form

$$\mathbb{D} = \left(\alpha \left(1 + \beta |\mathbb{T}_\delta|^2 \right)^n + \delta \right) \mathbb{T}_\delta, \tag{4}$$

introduced in Le Roux and Rajagopal [7] (for a thorough discussion of a simpler model with $\delta = 0$, see Málek et al. [8]). With properly tuned parameter values, this constitutive relation leads to the characteristic S-shaped curve in the strain rate–stress diagram; thus, the model (4) can serve as simple phenomenological model for flows of substances that exhibit such behavior—see for example Boltenhagen et al. [9], Grob et al. [10] and Mari et al. [11] to name a few (further references and a thorough discussion can be found in Perláčová and Průša [12] and Janečka and Průša [13]). Note that if the constitutive relation leads to the S-shaped curve in the strain rate–stress diagram, then (4) is not invertible, and it can not be rewritten in the classical form $\mathbb{T}_\delta = \mathfrak{g}(\mathbb{D})$. Other models that are easy to describe using (1) are the models for incompressible fluids with pressure-dependent viscosity (see Rajagopal [5]), or for that matter models wherein the viscosity depends on the stress such as the models used for the flow of ice; see Blatter [14] and Pettit and Waddington [15] and also Málek and Průša [16] for further references regarding this class of models.

Other models that are easy to describe using (1) or the implicit relation $\mathfrak{k}(\mathbb{T}_\delta, \mathbb{D}) = \mathbb{O}$ are the models with activation criterion introduced by Bingham [17] and Herschel and Bulkeley [18]. These models are usually written down in the form of a dichotomy relation (see for example Duvaut and Lions [19]), as follows:

$$|\mathbb{T}_\delta| \leq \tau_* \Leftrightarrow \mathbb{D} = \mathbb{O} \quad \text{and} \quad |\mathbb{T}_\delta| > \tau_* \Leftrightarrow \mathbb{T}_\delta = \frac{\tau_* \mathbb{D}}{|\mathbb{D}|} + 2\nu(|\mathbb{D}|)\mathbb{D}, \tag{5}$$

where ν is a positive function and τ_* is the yield stress. If we use the alternative framework based on the constitutive relation $\mathfrak{k}(\mathbb{T}_\delta, \mathbb{D}) = \mathbb{O}$, then these models can be rewritten as

$$\mathbb{D} = \frac{1}{2\nu(|\mathbb{D}|)} \frac{(|\mathbb{T}_\delta| - \tau_*)^+}{|\mathbb{T}_\delta|} \mathbb{T}_\delta, \tag{6}$$

where $x^+ = \max\{x, 0\}$ and ν is a non-negative function. This observation can be exploited in the discussion of the physical and mathematical properties of the models; see for example Rajagopal and Srinivasa [20] and Bulíček et al. [21]. For further discussion of generalized models of type $\mathfrak{k}(\mathbb{T}_\delta, \mathbb{D}) = \mathbb{O}$, we also refer the reader to Perláčová and Průša [12]. Note that, in all cases discussed above, the constitutive relation between the stress and the symmetric part of the velocity gradient has been related to entropy production mechanisms (see for example Janečka and Průša [13] for a through discussion).

Readers interested in analytical and semianalytical solutions to boundary value problems for fluids described by constitutive relations of the type $\mathbb{T}_\delta = \mathfrak{g}(\mathbb{D})$ or $\mathfrak{k}(\mathbb{T}_\delta, \mathbb{D}) = \mathbb{O}$ are referred to Málek et al. [8], Le Roux and Rajagopal [7], Srinivasan and Karra [22], Narayan and Rajagopal [23], Fusi and Farina [24], Fusi [25], Housiadas et al. [26], Gomez-Constante and Rajagopal [27] and Fetecau and Bridges [28], to name a few. The numerical solution of the corresponding governing equations is investigated in Janečka et al. [29], while a rigorous numerical analysis for various models that fall into this class is discussed in Dienes et al. [30], Stebel [31], Hirn et al. [32], Süli and Tscherpel [33], Farrell et al. [34], Farrell and Gazca-Orozco [35]. A rigorous mathematical theory for some of the aforementioned models is developed in Bulíček et al. [21,36] and Maringová and Žabenský [37]; see also Blechta et al. [38] for a recent review.

The further generalization of the algebraic relation $\mathfrak{k}(\mathbb{T}_\delta, \mathbb{D}) = \mathbb{O}$ is the implicit relation of the type

$$\mathfrak{H}_{s=0}^{+\infty}(\mathbb{T}(t-s), \mathbb{C}_t(t-s)) = \mathbb{O}, \tag{7}$$

that relates the histories of the stress tensor and the relative right Cauchy–Green tensor; for details, see Průša and Rajagopal [39]. Special instances of (7) are rate-type and differential-type models for viscoelastic fluids and integral models for viscoelastic fluids, while in all these cases, one can consider models with pressure/stress dependent material parameters; see for example Karra et al. [40], Kannan and Rajagopal [41], Housiadas [42,43] and Arcangioli et al. [44] and references therein.

2.2. Solids

Regarding the similar development in the case of constitutive relations for solids, where in the classical approach is based on constitutive relations in the form $\mathbb{T} = \mathfrak{h}(\mathbb{B})$, the non-standard relations of the type $\mathbb{B} = \mathfrak{i}(\mathbb{T})$ or even $\mathfrak{j}(\mathbb{T}, \mathbb{B}) = \mathbb{O}$ have been considered without a proper thermodynamic basis or clearly articulated rationale for the choice of such constitutive relations in some early works such as Morgan [45], Fitzgerald [46] or Blume [47] and Xiao and Chen [48], Xiao et al. [49]. A systematic study of constitutive relations of the type $\mathbb{B} = \mathfrak{i}(\mathbb{T})$ with a coherent explanation based on issues of causality for the choice of such constitutive relations was initiated by Rajagopal [4], and the number of works focused on this concept has been growing ever since. Regarding elastic (non-dissipative) solids, we refer the reader to a recent review by Bustamante and Rajagopal [50] and references therein, while some aspects of the description of non-elastic response such as plasticity are discussed in Rajagopal and Srinivasa [51,52] and Cichra and Průša [53] (a discussion and references on works dealing with the invertibility of the constitutive relations can be found in Sfyris and Bustamante [54]; one of the earliest studies of the same topic is that by Truesdell and Moon [55]). As an example of the benefits of the proposed change of perspective, we can mention especially the study by Muliana et al. [56], who address the classical topic of the mathematical modeling of the mechanical response of rubber.

From the current perspective, the important aspects of the novel approach to the constitutive theory of solids are the following. First, if one wants to work with constitutive relations of

the type $\mathbb{B} = \mathfrak{i}(\mathbb{T})$, then it is convenient to work with the Hencky strain tensor $\mathbb{H} =_{\text{def}} \frac{1}{2} \ln \mathbb{B}$ instead of the left Cauchy–Green tensor \mathbb{B} . (This observation is made in various works in elasticity and inelasticity, see especially Xiao and Chen [48], Xiao et al. [49,57]. Note also that in the context of viscoelastic fluids the preferred quantity to work with is—from the numerical perspective—the log-conformation tensor, see Fattal and Kupferman [58,59], Hulsen et al. [60], that is the logarithm of the conformation tensor. Apparently, this is not a coincidence.) Second, the thermodynamic potential of choice is Gibbs free energy, see Rajagopal and Srinivasa [61] and Srinivasa [62]. In particular, in the case of elastic solids the formula for the Hencky strain $\mathbb{H} = \mathfrak{i}(\mathbb{T})$ arises via the differentiation of a potential, and the potential can be identified as Gibbs free energy, see especially Gokulnath et al. [63] and Průša et al. [64].

3. Viscoelastic Rate-Type Fluids

Since viscoelastic fluids are fluids that exhibit both “viscous” and “elastic” characteristics (see for example Snoeijer et al. [65] for a recent discussion concerning the same), one can ask whether the novel approach to the modeling of the response of elastic solids can be also applied in this context. The point is the following: traditionally, the elastic properties of viscoelastic fluids are from the phenomenological perspective modeled using the same concepts as in the classical theory of nonlinear solids, wherein the Cauchy stress tensor is assumed to be a function of the left Cauchy–Green tensor. The question is what happens if an opposite perspective to the classical one is taken, whereby the left Cauchy–Green stress tensor is assumed to be a function of the stress tensor, or one even assumes that these quantities are related by a implicit algebraic relation.

We focus on this question, and we show how to use the new perspective in the development of mathematical models for viscoelastic fluids and discuss in detail the thermodynamic underpinnings of such models. In particular, we focus on the use of Gibbs free energy instead of Helmholtz free energy. Using the standard Giesekus/Oldroyd-B models, we show how the alternative approach works in the case of well-known models, and we argue that the proposed approach is straightforward to generalize to more complex settings.

Our basic framework is based on the concept of evolving natural configuration (see Eckart [66] and Rajagopal and Srinivasa [67]) that has been successfully applied in various settings; see for example Málek et al. [68], Narayan et al. [69], Málek et al. [70], Málek et al. [71], Tůma et al. [72], Málek et al. [73], Řehoř et al. [74] or Sumith et al. [75] to name a few (for other approaches to the mathematical modeling of viscoelastic rate-type fluids, see Leonov [76,77], Wapperom and Hulsen [78] or the GENERIC framework; see Grmela and Öttinger [79], Öttinger and Grmela [80] and Pavelka et al. [81]).

4. Gibbs Free Energy-Based Approach to Viscoelastic Rate-Type Fluids

The concept of an evolving natural configuration is based on the assumption that the overall response of a viscoelastic fluid is composed of a dissipative (viscous) response and a non-dissipative (elastic) response, while the latter can be described using the concepts known from the theory of nonlinear elasticity. However, the left Cauchy–Green tensor for the total response \mathbb{B} must be replaced by the left Cauchy–Green tensor $\mathbb{B}_{\kappa_p(t)}$ associated to the elastic part of the fluid response. The subscript $\kappa_p(t)$ denotes the instantaneously relaxed or “natural” configuration; see Rajagopal and Srinivasa [67] for a detailed rationale (another detailed explanation of the concept of natural configuration is given in a recent review article by Málek and Průša [16]—however, note that in the current contribution we do not explicitly use the decomposition of the total deformation gradient $\mathbb{F} = \mathbb{F}_{\kappa_p(t)} \mathbb{G}$ to the dissipative and non-dissipative part. In particular, we do not explicitly work with the tensor \mathbb{G} , which makes our approach different from the treatment in Rajagopal and Srinivasa [67] and Málek et al. [82]). If we want to exploit the novel approach to the modeling of elastic response, we have to mimic (3), but the full Cauchy–Green tensor \mathbb{B} must be replaced by the partial Cauchy–Green tensor $\mathbb{B}_{\kappa_p(t)}$.

Furthermore, as we have already indicated, the partial left Cauchy–Green tensor $\mathbb{B}_{\kappa_{p(t)}}$ should be replaced by the corresponding Hencky strain tensor, and the thermodynamic potential of choice should be Gibbs free energy. The details are worked out below. Note that unlike in other works focused on the use of Gibbs potential (see especially Narayan et al. [83]), we are not assuming that the processes of interest are isothermal. The theory outlined below is applicable in a non-isothermal setting.

The main idea behind the presented approach is that the behavior of the material in the processes of interest is determined by two factors, namely its ability to store energy and produce entropy. The energy storage mechanisms are in the present case specified by the choice of Gibbs free energy g , while the entropy production mechanisms are specified by the choice of the formula for the entropy production ζ . Since the entropy η is obtained via the differentiation of g , the evolution equation for η then necessarily links the assumed form of Gibbs free energy g and the assumed entropy production ζ . This link can be exploited in the identification of the constitutive relations. Consequently, the first step of the proposed approach is to derive an evolution equation for the entropy.

4.1. Entropy Evolution Equation—General Case

The fundamental equation in continuum mechanics is the evolution equation for the specific internal energy e , which for the internal energy per unit mass, $[e] = \text{J/kg}$, is

$$\rho \frac{de}{dt} = \mathbb{T} \bullet \mathbb{D} - \text{div } \mathbf{j}_q, \tag{8}$$

For more information, see standard texts on continuum thermodynamics such as Truesdell and Noll [1] or Müller [2] (the notation in (8) is the standard one, where the symbol \mathbb{T} stands for the Cauchy stress tensor, \mathbb{D} denotes the symmetric part of the velocity gradient, ρ denotes the density, and \mathbf{j}_q denotes the heat flux). The evolution equation (8) is the starting point for the derivation of the desired entropy evolution equation.

In the present approach to the theory of viscoelastic rate-type fluids, we assume that the specific internal energy is a function of the specific entropy η , density ρ and the Hencky strain tensor associated to the elastic response,

$$\mathbb{H}_{\kappa_{p(t)}} =_{\text{def}} \frac{1}{2} \ln \mathbb{B}_{\kappa_{p(t)}}. \tag{9}$$

Furthermore, it is convenient to split the Hencky strain tensor to the traceless (deviatoric) part $\mathbb{H}_{\kappa_{p(t)}, \delta}$ and the spherical part $h_{\kappa_{p(t)}}$,

$$\mathbb{H}_{\kappa_{p(t)}} = h_{\kappa_{p(t)}} \mathbb{I} + \mathbb{H}_{\kappa_{p(t)}, \delta}, \tag{10}$$

where we introduce the notation $\mathbb{A}_\delta =_{\text{def}} \mathbb{A} - \frac{1}{3}(\text{Tr } \mathbb{A})\mathbb{I}$ for the traceless (deviatoric) part of the corresponding tensor. We note that the deviatoric part of the Hencky strain tensor $\mathbb{H}_{\kappa_{p(t)}, \delta}$ is equal to the rescaled Hencky strain tensor:

$$\overline{\mathbb{H}_{\kappa_{p(t)}}} =_{\text{def}} \frac{1}{2} \ln \overline{\mathbb{B}_{\kappa_{p(t)}}} \tag{11}$$

associated to the rescaled left Cauchy–Green tensor $\overline{\mathbb{B}_{\kappa_{p(t)}}$ that is defined as

$$\overline{\mathbb{B}_{\kappa_{p(t)}}} =_{\text{def}} \frac{\mathbb{B}_{\kappa_{p(t)}}}{J_{\kappa_{p(t)}}^{\frac{2}{3}}}, \tag{12}$$

where $J_{\kappa_{p(t)}} =_{\text{def}} \det \mathbb{F}_{\kappa_{p(t)}}$. The equality $\mathbb{H}_{\kappa_{p(t)}, \delta} = \overline{\mathbb{H}_{\kappa_{p(t)}}$ is discussed for example in Průša et al. [64]; see also Xiao et al. [49]. The usage of the rescaled left Cauchy–Green tensor $\overline{\mathbb{B}_{\kappa_{p(t)}}$ is a common practice in the theory of slightly compressible solids; see for example Horgan

and Saccomandi [84]. The rationale is that $\det \overline{\mathbb{B}_{\kappa_{p(t)}}} = 1$, and thus the deformation is conveniently split into volume-preserving and volume-changing parts. We also note that if the elastic part of the response is volume-preserving, then $\det \mathbb{B}_{\kappa_{p(t)}} = 1$ and consequently $\text{Tr} \mathbb{H}_{\kappa_{p(t)}} = 0$, which in this special case implies that $\mathbb{H}_{\kappa_{p(t)}} = \mathbb{H}_{\kappa_{p(t)}, \delta} = \overline{\mathbb{H}_{\kappa_{p(t)}}$.

Regarding the specific internal energy e , we therefore assume that

$$e = e(\eta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta}), \tag{13}$$

which allows us to seamlessly deal with incompressible materials as well (if the material is homogeneous and incompressible, we can simply remove ρ from (13) since the density is in this case constant. Similarly, if the response of the elastic component is modeled as a response of an incompressible material, then $h_{\kappa_{p(t)}}$ is a constant and we can simply remove the $h_{\kappa_{p(t)}}$ form (13). However, we need to introduce the corresponding Lagrange multipliers that enforce the incompressibility constraint). Introducing the thermodynamic temperature θ in the standard manner as

$$\theta =_{\text{def}} \frac{\partial e}{\partial \eta}(\eta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta}), \tag{14}$$

we can rewrite (8) in terms of Helmholtz free energy ψ ,

$$\psi(\theta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta}) =_{\text{def}} \left(e(\eta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta}) - \theta \eta \right) \Big|_{\eta = \eta(\theta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta})} \tag{15}$$

as

$$\begin{aligned} \rho \theta \frac{d\eta}{dt} &= \mathbb{T} \bullet \mathbb{D} - \rho \frac{\partial \psi}{\partial \rho}(\theta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta}) \frac{d\rho}{dt} - \rho \frac{\partial \psi}{\partial h_{\kappa_{p(t)}}}(\theta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta}) \frac{dh_{\kappa_{p(t)}}}{dt} \\ &\quad - \rho \left(\frac{\partial \psi}{\partial \mathbb{H}_{\kappa_{p(t)}, \delta}}(\theta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta}) \right) \bullet \frac{d\mathbb{H}_{\kappa_{p(t)}, \delta}}{dt} - \text{div } j_q. \end{aligned} \tag{16}$$

In the next to the last term, we can consider only the traceless part of $\frac{\partial \psi}{\partial \mathbb{H}_{\kappa_{p(t)}, \delta}}$ since it enters the dot product with the traceless tensor $\frac{d\mathbb{H}_{\kappa_{p(t)}, \delta}}{dt}$. Finally, we can split the Cauchy stress tensor into the mean normal stress $m =_{\text{def}} \frac{1}{3} \text{Tr } \mathbb{T}$ and the deviatoric part \mathbb{T}_δ ,

$$\mathbb{T} = m \mathbb{I} + \mathbb{T}_\delta, \tag{17}$$

and we get

$$\begin{aligned} \rho \theta \frac{d\eta}{dt} &= m \text{div } \mathbf{v} + \mathbb{T}_\delta \bullet \mathbb{D}_\delta + \rho^2 \frac{\partial \psi}{\partial \rho}(\theta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta}) \text{div } \mathbf{v} \\ &\quad - \rho \frac{\partial \psi}{\partial h_{\kappa_{p(t)}}}(\theta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta}) \frac{dh_{\kappa_{p(t)}}}{dt} - \rho \left(\frac{\partial \psi}{\partial \mathbb{H}_{\kappa_{p(t)}, \delta}}(\theta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta}) \right) \bullet \frac{d\mathbb{H}_{\kappa_{p(t)}, \delta}}{dt} - \text{div } j_q. \end{aligned} \tag{18}$$

We have also used the balance of mass $\frac{d\rho}{dt} + \rho \text{div } \mathbf{v} = 0$. We recall that the specific internal energy and the specific Helmholtz free energy are related by the Legendre transformation, and that the following formula holds:

$$\eta = - \frac{\partial \psi}{\partial \theta}(\theta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta}). \tag{19}$$

The manipulation outlined above therefore gives us the desired evolution Equation (16) for the entropy η . This equation is the starting point for the specification of the constitutive relations. In the standard approach, in principle, it is desired to provide a

constitutive relation for the Cauchy stress tensor \mathbb{T} and formulae for the time derivatives of $h_{\kappa_{p(t)}}$ and $\mathbb{H}_{\kappa_{p(t)}, \delta}$ such that the right-hand side of (16) is nonnegative, which in turn guarantees that the second law of thermodynamics holds in the given material and during the given class of processes (see for example Dostálík et al. [85] for applications of this procedure to several well-known viscoelastic rate-type models). However, we might not want to use (16) directly; we want to rewrite it in terms of Gibbs free energy g .

In order to use Gibbs free energy, we need to introduce the thermodynamic pressure p_{th} , and the stress $\mathbb{T}_{\kappa_{p(t)}}$, which we again split into the spherical part $p_{\kappa_{p(t)}}$ and the traceless part $\mathbb{T}_{\kappa_{p(t)}, \delta}$,

$$\mathbb{T}_{\kappa_{p(t)}} =_{\text{def}} p_{\kappa_{p(t)}} \mathbb{I} + \mathbb{T}_{\kappa_{p(t)}, \delta}. \tag{20}$$

Using the definitions

$$p_{th} =_{\text{def}} \rho^2 \frac{\partial \psi}{\partial \rho}(\theta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta}), \tag{21a}$$

$$\frac{p_{\kappa_{p(t)}}}{\rho} =_{\text{def}} \frac{\partial \psi}{\partial h_{\kappa_{p(t)}}}(\theta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta}) \tag{21b}$$

$$\frac{\mathbb{T}_{\kappa_{p(t)}, \delta}}{\rho} =_{\text{def}} \left(\frac{\partial \psi}{\partial \mathbb{H}_{\kappa_{p(t)}, \delta}}(\theta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta}) \right)_{\delta}, \tag{21c}$$

we can further rewrite (16) in such a way that it is ready for the use of specific Gibbs free energy g (definition (21a) is the standard definition of the thermodynamic pressure as it is used in the classical thermodynamics of compressible fluids). If we further introduce the notation

$$s_{\kappa_{p(t)}} =_{\text{def}} \frac{p_{\kappa_{p(t)}}}{\rho} \tag{22a}$$

$$\mathbb{S}_{\kappa_{p(t)}, \delta} =_{\text{def}} \frac{\mathbb{T}_{\kappa_{p(t)}, \delta}}{\rho}, \tag{22b}$$

the specific Gibbs free energy is then defined as

$$g(\theta, p_{th}, s_{\kappa_{p(t)}}, \mathbb{S}_{\kappa_{p(t)}, \delta}) =_{\text{def}} \left(\psi(\theta, \rho, h_{\kappa_{p(t)}}, \mathbb{H}_{\kappa_{p(t)}, \delta}) + \frac{p_{th}}{\rho} - s_{\kappa_{p(t)}} h_{\kappa_{p(t)}} - \mathbb{S}_{\kappa_{p(t)}, \delta} \bullet \mathbb{H}_{\kappa_{p(t)}, \delta} \right) \Big|_{\rho=\rho(\theta, p_{th}, s_{\kappa_{p(t)}}, \mathbb{S}_{\kappa_{p(t)}, \delta})}, \dots \tag{23}$$

We note that the transition to Gibbs free energy is in fact done by another Legendre transformation, and that the use of the Legendre transformation implies that

$$\eta = - \frac{\partial g}{\partial \theta}(\theta, p_{th}, s_{\kappa_{p(t)}}, \mathbb{S}_{\kappa_{p(t)}, \delta}), \tag{24a}$$

$$\frac{1}{\rho} = \frac{\partial g}{\partial p_{th}}(\theta, p_{th}, s_{\kappa_{p(t)}}, \mathbb{S}_{\kappa_{p(t)}, \delta}), \tag{24b}$$

$$h_{\kappa_{p(t)}} = - \frac{\partial g}{\partial s_{\kappa_{p(t)}}}(\theta, p_{th}, s_{\kappa_{p(t)}}, \mathbb{S}_{\kappa_{p(t)}, \delta}). \tag{24c}$$

$$\mathbb{H}_{\kappa_{p(t)}, \delta} = - \left(\frac{\partial g}{\partial \mathbb{S}_{\kappa_{p(t)}, \delta}}(\theta, p_{th}, s_{\kappa_{p(t)}}, \mathbb{S}_{\kappa_{p(t)}, \delta}) \right)_{\delta}. \tag{24d}$$

If we use definitions (21), we see that (18) can be rewritten as

$$\rho \theta \frac{d\eta}{dt} = - \frac{m + p_{th}}{\rho} \frac{d\rho}{dt} + \mathbb{T}_{\delta} \bullet \mathbb{D}_{\delta} - p_{\kappa_{p(t)}} \frac{dh_{\kappa_{p(t)}}}{dt} - \mathbb{T}_{\kappa_{p(t)}, \delta} \bullet \frac{d\mathbb{H}_{\kappa_{p(t)}, \delta}}{dt} - \text{div } \mathbf{j}_q, \tag{25}$$

which upon using formulae (24) yields

$$\begin{aligned} \rho\theta \frac{d\eta}{dt} = & -\rho(m + p_{th}) \frac{d}{dt} \left(\frac{\partial g}{\partial p_{th}} \left(\theta, p_{th}, s_{\kappa_{p(t)}}, \mathbb{S}_{\kappa_{p(t)}, \delta} \right) \right) + \mathbb{T}_\delta \bullet \mathbb{D}_\delta + p_{\kappa_{p(t)}} \frac{d}{dt} \left(\frac{\partial g}{\partial s_{\kappa_{p(t)}}} \left(\theta, p_{th}, s_{\kappa_{p(t)}}, \mathbb{S}_{\kappa_{p(t)}, \delta} \right) \right) \\ & + \mathbb{T}_{\kappa_{p(t)}, \delta} \bullet \frac{d}{dt} \left(\left(\frac{\partial g}{\partial \mathbb{S}_{\kappa_{p(t)}, \delta}} \left(\theta, p_{th}, s_{\kappa_{p(t)}}, \mathbb{S}_{\kappa_{p(t)}, \delta} \right) \right) \right) - \text{div } \mathbf{j}_q. \end{aligned} \quad (26)$$

This is a general evolution equation for the specific entropy η .

4.2. Entropy Evolution Equation—Incompressible Fluids

Let us now manipulate (25) into a form that is convenient for the ongoing investigation of the constitutive relations. For the sake of simplicity, we now restrict ourselves to incompressible fluids, which means that the density is a constant and it no longer enters the formula for Gibbs free energy. Further, we will not split the elastic response into the volume-preserving and volume-changing part; that is, we use the full Hencky strain tensor $\mathbb{H}_{\kappa_{p(t)}}$ instead of the pair $\mathbb{H}_{\kappa_{p(t)}, \delta}$ and $h_{\kappa_{p(t)}}$, and similarly for the reduced stress $\mathbb{S}_{\kappa_{p(t)}}$. In this case, Gibbs free energy is therefore given as

$$g = g\left(\theta, \mathbb{S}_{\kappa_{p(t)}}\right), \quad (27)$$

and the evolution equation for the entropy (26) reduces to

$$\rho\theta \frac{d\eta}{dt} = \mathbb{T}_\delta \bullet \mathbb{D}_\delta + \mathbb{T}_{\kappa_{p(t)}} \bullet \frac{d}{dt} \left[\frac{\partial g}{\partial \mathbb{S}_{\kappa_{p(t)}}} \left(\theta, \mathbb{S}_{\kappa_{p(t)}} \right) \right] - \text{div } \mathbf{j}_q, \quad (28)$$

and relations (24) read

$$\eta = -\frac{\partial g}{\partial \theta} \left(\theta, \mathbb{S}_{\kappa_{p(t)}} \right), \quad (29a)$$

$$\mathbb{H}_{\kappa_{p(t)}} = -\frac{\partial g}{\partial \mathbb{S}_{\kappa_{p(t)}}} \left(\theta, \mathbb{S}_{\kappa_{p(t)}} \right). \quad (29b)$$

We note that if we deal with an incompressible substance, then the Cauchy stress tensor is split into the spherical part $-p\mathbb{I}$ and the deviatoric part \mathbb{T}_δ ,

$$\mathbb{T} = -p\mathbb{I} + \mathbb{T}_\delta, \quad (30)$$

and that the pressure p —which can be now understood as the Lagrange multiplier enforcing the incompressibility constraint—constitutes a new unknown field to be solved. Namely, it is not given by an equation of state as a function of density, temperature and other variables. Furthermore, the incompressibility constraint $\text{div } \mathbf{v} = 0$ implies that $\mathbb{D}_\delta = \mathbb{D}$.

Finally, we note that if we deal with an isotropic elastic response, which is the case in the rest of the paper, then (29b) in virtue of the standard representation theorem for isotropic functions (see for example Truesdell and Noll [1]) implies that the tensors $\mathbb{H}_{\kappa_{p(t)}}$ and $\mathbb{S}_{\kappa_{p(t)}}$ commute; that is, $\mathbb{H}_{\kappa_{p(t)}} \mathbb{S}_{\kappa_{p(t)}} = \mathbb{S}_{\kappa_{p(t)}} \mathbb{H}_{\kappa_{p(t)}}$.

4.2.1. Manipulations with Entropy Production—Direct Use of Gibbs Free Energy

If we further assume that Gibbs free energy has the property that the mixed derivative vanishes—that is, if

$$\frac{\partial^2 g}{\partial \theta \partial \mathbb{S}_{\kappa_{p(t)}}} = 0, \quad (31)$$

then (26) reduces to

$$\rho\theta \frac{d\eta}{dt} = \mathbb{T}_\delta \bullet \mathbb{D}_\delta + \rho \mathbb{S}_{\kappa_{p(t)}} \bullet \frac{\partial^2 g}{\partial \mathbb{S}_{\kappa_{p(t)}}^2} (\theta, \mathbb{S}_{\kappa_{p(t)}}) \frac{d\mathbb{S}_{\kappa_{p(t)}}}{dt} - \text{div } \mathbf{j}_q. \tag{32}$$

The notation might be slightly ambiguous here. Using the index notation, we have

$$\mathbb{S}_{\kappa_{p(t)}} \bullet \frac{\partial^2 g}{\partial \mathbb{S}_{\kappa_{p(t)}}^2} (\theta, \mathbb{S}_{\kappa_{p(t)}}) \frac{d\mathbb{S}_{\kappa_{p(t)}}}{dt} = \mathbb{S}_{\kappa_{p(t)}ij} \frac{\partial^2 g}{\partial \mathbb{S}_{\kappa_{p(t)}ij} \partial \mathbb{S}_{\kappa_{p(t)}mn}} \frac{d\mathbb{S}_{\kappa_{p(t)}mn}}{dt},$$

where we have used the summation convention.

The reduced stress tensor $\mathbb{S}_{\kappa_{p(t)}}$ is a second-order tensor in the current configuration that transforms normals to infinitesimal surfaces to traction vectors; thus, we see that it is a tensor of type $\binom{2}{0}$. If we need to calculate the time derivative of such a tensor, then the natural concept is the upper convected derivative,

$$\overset{\nabla}{\mathbb{S}}_{\kappa_{p(t)}} =_{\text{def}} \frac{d\mathbb{S}_{\kappa_{p(t)}}}{dt} - \mathbb{L}\mathbb{S}_{\kappa_{p(t)}} - \mathbb{S}_{\kappa_{p(t)}}\mathbb{L}^\top. \tag{33}$$

See for example Stumpf and Hoppe [86] for geometrical underpinnings of the concept of the upper convected derivative and its link to the concept of the Lie derivative. Using the definition of the upper convected derivative, we see that (32) can be rewritten as

$$\rho\theta \frac{d\eta}{dt} = \left\{ \mathbb{T}_\delta + 2\rho \left[\mathbb{S}_{\kappa_{p(t)}} \frac{\partial^2 g}{\partial \mathbb{S}_{\kappa_{p(t)}}^2} (\theta, \mathbb{S}_{\kappa_{p(t)}}) \mathbb{S}_{\kappa_{p(t)}} \right]_\delta \right\} \bullet \mathbb{D}_\delta + \rho \mathbb{S}_{\kappa_{p(t)}} \bullet \frac{\partial^2 g}{\partial \mathbb{S}_{\kappa_{p(t)}}^2} (\theta, \mathbb{S}_{\kappa_{p(t)}}) \overset{\nabla}{\mathbb{S}}_{\kappa_{p(t)}} - \text{div } \mathbf{j}_q. \tag{34}$$

In order to clarify the slightly ambiguous notation, we can resort to the index notation. We set

$$\left[\mathbb{S}_{\kappa_{p(t)}} \frac{\partial^2 g}{\partial \mathbb{S}_{\kappa_{p(t)}}^2} \mathbb{S}_{\kappa_{p(t)}} \right]_{mp} = \mathbb{S}_{\kappa_{p(t)}ij} \frac{\partial^2 g}{\partial \mathbb{S}_{\kappa_{p(t)}ij} \partial \mathbb{S}_{\kappa_{p(t)}mn}} \mathbb{S}_{\kappa_{p(t)}np'}$$

where we use the summation convention. Since $\mathbb{S}_{\kappa_{p(t)}}$ is a symmetric tensor that commutes with $\frac{\partial g}{\partial \mathbb{S}_{\kappa_{p(t)}}$, we see that $\mathbb{S}_{\kappa_{p(t)}} \frac{\partial^2 g}{\partial \mathbb{S}_{\kappa_{p(t)}}^2} \mathbb{S}_{\kappa_{p(t)}}$ is also a symmetric tensor.

We are exploiting the fact that $\mathbb{S}_{\kappa_{p(t)}}$ commutes with $\frac{\partial g}{\partial \mathbb{S}_{\kappa_{p(t)}}$ and the cyclic property of the trace. The fact that $\mathbb{S}_{\kappa_{p(t)}}$ commutes with $\frac{\partial g}{\partial \mathbb{S}_{\kappa_{p(t)}}$ follows again from the representation theorems for isotropic functions. Finally, the standard manipulation of the heat flux term yields

$$\rho \frac{d\eta}{dt} + \text{div} \left(\frac{\mathbf{j}_q}{\theta} \right) = \frac{1}{\theta} \left\{ \left\{ \mathbb{T}_\delta + 2\rho \left[\mathbb{S}_{\kappa_{p(t)}} \frac{\partial^2 g}{\partial \mathbb{S}_{\kappa_{p(t)}}^2} (\theta, \mathbb{S}_{\kappa_{p(t)}}) \mathbb{S}_{\kappa_{p(t)}} \right]_\delta \right\} \bullet \mathbb{D}_\delta + \rho \mathbb{S}_{\kappa_{p(t)}} \bullet \frac{\partial^2 g}{\partial \mathbb{S}_{\kappa_{p(t)}}^2} (\theta, \mathbb{S}_{\kappa_{p(t)}}) \overset{\nabla}{\mathbb{S}}_{\kappa_{p(t)}} \right\} - \frac{\mathbf{j}_q \bullet \nabla \theta}{\theta^2}, \tag{35}$$

which is the formula used by Rajagopal and Srinivasa [61].

4.2.2. Manipulations with Entropy Production—Indirect Use of Gibbs Free Energy

Note however that (25) can be also manipulated differently, and in this case no further structural assumptions regarding the specific Gibbs free energy are necessary. In the incompressible setting, (25) reads

$$\rho\theta \frac{d\eta}{dt} = \mathbb{T}_\delta \bullet \mathbb{D}_\delta - \mathbb{T}_{\kappa_{p(t)}} \bullet \frac{d\mathbb{H}_{\kappa_{p(t)}}}{dt} - \text{div } \mathbf{j}_q. \tag{36}$$

Unlike in the previous manipulation, we keep the Hencky strain tensor $\mathbb{H}_{\kappa_{p(t)}}$ in the equation, but we replace its material time derivative by the upper convected derivative,

$$\overset{\nabla}{\mathbb{H}}_{\kappa_{p(t)}} = \frac{d\mathbb{H}_{\kappa_{p(t)}}}{dt} - \mathbb{L}\mathbb{H}_{\kappa_{p(t)}} - \mathbb{H}_{\kappa_{p(t)}}\mathbb{L}^\top. \tag{37}$$

The rationale for this manipulation is the same as in the case of the stress $\mathbb{S}_{\kappa_{p(t)}}$. Next, using the standard manipulation of the heat flux j_q , we see that (25) can be finally rewritten as

$$\rho \frac{d\eta}{dt} + \text{div} \left(\frac{j_q}{\theta} \right) = \frac{1}{\theta} \left\{ \left\{ \mathbb{T}_\delta - 2\rho \left[\mathbb{S}_{\kappa_{p(t)}} \mathbb{H}_{\kappa_{p(t)}} \right]_\delta \right\} \bullet \mathbb{D}_\delta + \rho \mathbb{S}_{\kappa_{p(t)}} \bullet \overset{\nabla}{\mathbb{H}}_{\kappa_{p(t)}} \right\} - \frac{j_q \bullet \nabla \theta}{\theta^2}. \tag{38}$$

4.2.3. Constitutive Relations

Either (38) or (35) can serve as a starting point for the specification of constitutive relations. Once we have postulated a formula for Gibbs free energy, which is tantamount to the specification of the energy storage mechanism in the material of interest, we can proceed with the specification of the entropy production mechanisms. This boils down to the specification of the formula for the right-hand side of the generic entropy evolution equation that has the structure

$$\rho \frac{d\eta}{dt} + \text{div } j_\eta = \zeta, \tag{39}$$

where j_η denotes the entropy flux and ζ denotes the entropy production. Once the required entropy production—the right-hand side or entropy evolution equation—is specified, we compare it with the entropy production implied by the choice of Gibbs free energy—i.e., with the right-hand-side of (38) or (35)—which allows us to fix the constitutive equations for \mathbb{T} and \mathbb{T}_{th} . Examples of this procedure are given below in Section 5.2.

A more sophisticated version of this approach is based on the assumption of the maximization of the entropy production; see Rajagopal and Srinivasa [6]. This approach is worked out in Section 5.3.

4.2.4. Temperature Evolution Equation

Finally, let us remark that once the specific Gibbs free energy is given, one can then exploit the relation between the entropy and the specific Gibbs free energy (see (24a)) and convert (35) or (38), respectively, to an evolution equation for the temperature:

$$\frac{d\eta}{dt} = -\frac{d}{dt} \left(\frac{\partial g}{\partial \theta} (\theta, \mathbb{S}_{\kappa_{p(t)}}) \right) = -\frac{\partial^2 g}{\partial \theta^2} (\theta, \mathbb{S}_{\kappa_{p(t)}}) \frac{d\theta}{dt} - \frac{\partial^2 g}{\partial \theta \partial \mathbb{S}_{\kappa_{p(t)}}} (\theta, \mathbb{S}_{\kappa_{p(t)}}) \bullet \frac{d\mathbb{T}_{\kappa_{p(t)}}}{dt}. \tag{40}$$

Note that if we use the assumption (31) that the mixed derivative of the specific Gibbs free energy vanishes, then the formula (40) simplifies to $\frac{d\eta}{dt} = -\frac{\partial^2 g}{\partial \theta^2} (\theta, \mathbb{S}_{\kappa_{p(t)}}) \frac{d\theta}{dt}$. If it is not the case, we need to consider the term $\frac{\partial^2 g}{\partial \theta \partial \mathbb{S}_{\kappa_{p(t)}}} (\theta, \mathbb{S}_{\kappa_{p(t)}}) \bullet \frac{d\mathbb{T}_{\kappa_{p(t)}}}{dt}$ as well. The explicit formula for this term, however, depends on the rate-type constitutive relation for the stress tensor $\mathbb{T}_{\kappa_{p(t)}}$.

5. Example—Giesekus/Oldroyd-B Viscoelastic Rate-Type Fluid in the Approach Based on the Gibbs Free Energy

We now use the approach based on assuming that Gibbs free energy and appropriate entropy producing mechanisms develop the popular incompressible rate-type viscoelastic fluid model due to Giesekus; see Giesekus [87]. Note that this model reduces, for a special choice of parameters, to the Oldroyd-B model; see Oldroyd [88]. In this case we have the luxury of writing down simple explicit formulas both for Helmholtz free energy and Gibbs free energy as well as for the entropy production. This makes the model ideal for clarifying key ideas.

5.1. Giesekus/Oldroyd-B Model via Helmholtz Free Energy—Classical Approach

Let us first summarize basic facts regarding the classical incompressible viscoelastic rate-type model derived by Giesekus for heat-conducting fluids with constant specific heat at a constant volume $c_{V,ref}$ (note that the original derivation, see Giesekus [87], is done in the purely mechanical context; no temperature evolution equation is given. The corresponding temperature evolution equation is derived for example in Wapperom and Hulsen [78]). The governing equations for the mechanical quantities are in this case

$$\operatorname{div} \mathbf{v} = 0, \tag{41a}$$

$$\rho \frac{d\mathbf{v}}{dt} = \operatorname{div} \mathbb{T} + \rho \mathbf{b}, \tag{41b}$$

$$\nu_1 \overline{\mathbb{B}_{\kappa_{p(t)}}^\nabla} = -\mu(\theta) \left[\alpha \mathbb{B}_{\kappa_{p(t)}}^2 + (1 - 2\alpha) \mathbb{B}_{\kappa_{p(t)}} - (1 - \alpha) \mathbb{I} \right], \tag{41c}$$

and the temperature evolution equation takes the form

$$\begin{aligned} \rho c_{V,ref} \frac{d\theta}{dt} = & \operatorname{div}(\kappa \nabla \theta) + 2\nu \mathbb{D}_\delta \bullet \mathbb{D}_\delta + \frac{\mu(\theta)^2}{2\nu_1} \operatorname{Tr} \left[\alpha \mathbb{B}_{\kappa_{p(t)}}^2 + (1 - 3\alpha) \mathbb{B}_{\kappa_{p(t)}} + (1 - \alpha) \mathbb{B}_{\kappa_{p(t)}}^{-1} + (3\alpha - 2) \mathbb{I} \right] \\ & + \theta \frac{d\mu}{d\theta} \mathbb{D}_\delta \bullet \left(\mathbb{B}_{\kappa_{p(t)}} \right)_\delta - \theta \frac{\mu(\theta)}{2\nu_1} \frac{d\mu}{d\theta} \operatorname{Tr} \left[\mathbb{B}_{\kappa_{p(t)}} + \mathbb{B}_{\kappa_{p(t)}}^{-1} - 2\mathbb{I} \right]. \end{aligned} \tag{41d}$$

The Cauchy stress tensor \mathbb{T} is given by the formulae

$$\mathbb{T} = m\mathbb{I} + \mathbb{T}_\delta, \quad \mathbb{T}_\delta = 2\nu \mathbb{D}_\delta + \mu(\theta) \left(\mathbb{B}_{\kappa_{p(t)}} \right)_\delta, \tag{41e}$$

where $\alpha \in [0, 1]$ is a model parameter, ν, ν_1 are positive material constants or functions that can eventually depend on temperature, and μ is a material constant or a function proportional to the thermodynamic temperature; μ plays the role of the shear modulus for the elastic response, while the combination $\frac{\nu_1}{\mu(\theta)}$ plays the role of relaxation time; see (41c) (a more complex temperature dependence of μ is not allowed. If we were dealing a more complex function than the constant/linear function of temperature, then we would get a model with specific heat at constant volume that depends on $\mathbb{B}_{\kappa_{p(t)}}$; see for example comments in Hron et al. [89]. While such models might be useful, we do not consider them in the present contribution). As we shall see later, the material constant/function μ enters, unlike ν and ν_1 , the formula for the specific Helmholtz free energy ψ , whose derivatives with respect to θ and $\mathbb{B}_{\kappa_{p(t)}}$ determine the properties of the material. Therefore, we shall explicitly write $\mu(\theta)$ in order to emphasize the temperature dependence of μ . On the other hand, for ν and ν_1 , the temperature dependence is not—from the theoretical point of view—as important as in the case of μ ; thus, we simply write ν and ν_1 instead of $\nu(\theta)$ and $\nu_1(\theta)$. Finally, the symbol \mathbf{b} denotes the body force.

A few remarks concerning the system (41) are in order. First, we note that the right-hand side of (41c) can be rewritten as

$$-\mu(\theta) \left[\alpha \mathbb{B}_{\kappa_{p(t)}}^2 + (1 - 2\alpha) \mathbb{B}_{\kappa_{p(t)}} - (1 - \alpha) \mathbb{I} \right] = -\mu(\theta) \left[\left(\mathbb{B}_{\kappa_{p(t)}} - \mathbb{I} \right) + \alpha \left(\mathbb{B}_{\kappa_{p(t)}} - \mathbb{I} \right)^2 \right]. \tag{42}$$

In this form, it is straightforward to see that if $\mathbb{B}_{\kappa_{p(t)}} = \mathbb{I}$, then the right-hand side of (41c) vanishes (this is useful information if one is interested in the steady states predicted by (41) in the case of zero external forcing). Second, the last term in the temperature evolution equation (41d) can be rewritten as

$$\begin{aligned} \frac{\mu(\theta)^2}{2\nu_1} \operatorname{Tr} \left[\alpha \mathbb{B}_{\kappa_{p(t)}}^2 + (1 - 3\alpha) \mathbb{B}_{\kappa_{p(t)}} + (1 - \alpha) \mathbb{B}_{\kappa_{p(t)}}^{-1} + (3\alpha - 2) \mathbb{I} \right] \\ = \frac{\mu(\theta)^2}{2\nu_1} \operatorname{Tr} \left[\mathbb{B}_{\kappa_{p(t)}}^{-1} \left((1 - \alpha) \mathbb{I} + \alpha \mathbb{B}_{\kappa_{p(t)}} \right) \left(\mathbb{B}_{\kappa_{p(t)}} - \mathbb{I} \right)^2 \right]. \end{aligned} \tag{43}$$

This manipulation shows that the corresponding term is nonnegative for $\alpha \in [0, 1]$, and that it vanishes for $\mathbb{B}_{\kappa_{p(t)}} = \mathbb{I}$, a prospective steady state in the system without external forcing. Third, we recall that if we set $\alpha = 0$, then we obtain the standard Oldroyd-B model; see Oldroyd [88].

The specific Helmholtz free energy ψ is in the case of model (41) known to be given by the formula

$$\psi(\theta, \mathbb{B}_{\kappa_{p(t)}}) =_{\text{def}} \psi_{\text{thermal}}(\theta) + \frac{\mu(\theta)}{2\rho} \left(\text{Tr} \mathbb{B}_{\kappa_{p(t)}} - 3 - \ln \det \mathbb{B}_{\kappa_{p(t)}} \right), \tag{44a}$$

$$\psi_{\text{thermal}}(\theta) =_{\text{def}} -c_{V,\text{ref}} \theta \left(\ln \frac{\theta}{\theta_{\text{ref}}} - 1 \right). \tag{44b}$$

and the entropy production reads

$$\dot{\zeta} = \frac{1}{\theta} \left\{ 2\nu \mathbb{D}_\delta \bullet \mathbb{D}_\delta + \frac{\mu(\theta)^2}{2\nu_1} \text{Tr} \left[\alpha \mathbb{B}_{\kappa_{p(t)}}^2 + (1 - 3\alpha) \mathbb{B}_{\kappa_{p(t)}} + (1 - \alpha) \mathbb{B}_{\kappa_{p(t)}}^{-1} + (3\alpha - 2) \mathbb{I} \right] \right\} + \kappa \frac{\nabla \theta \bullet \nabla \theta}{\theta^2}. \tag{45}$$

Note that (43) shows that the entropy production is nonnegative. We recall that the Hencky strain tensor associated to the elastic response $\mathbb{H}_{\kappa_{p(t)}}$ is given by (9), hence we have

$$\mathbb{B}_{\kappa_{p(t)}} = e^{2\mathbb{H}_{\kappa_{p(t)}}}, \tag{46}$$

and the formula for Helmholtz free energy ψ in terms of $\mathbb{H}_{\kappa_{p(t)}}$ reads

$$\psi(\theta, \mathbb{H}_{\kappa_{p(t)}}) =_{\text{def}} \psi_{\text{thermal}}(\theta) + \frac{\mu(\theta)}{2\rho} \left(\text{Tr} e^{2\mathbb{H}_{\kappa_{p(t)}}} - 3 - \ln \det e^{2\mathbb{H}_{\kappa_{p(t)}}} \right). \tag{47}$$

Using the fact that the Hencky strain tensor is represented by a symmetric matrix and the standard identity $\det e^{\mathbb{A}} = e^{\text{Tr} \mathbb{A}}$, we see that (47) can be rewritten as

$$\psi(\theta, \mathbb{H}_{\kappa_{p(t)}}) =_{\text{def}} \psi_{\text{thermal}}(\theta) + \frac{\mu(\theta)}{2\rho} \left(\text{Tr} e^{2\mathbb{H}_{\kappa_{p(t)}}} - 3 - 2 \text{Tr} \mathbb{H}_{\kappa_{p(t)}} \right). \tag{48}$$

The stress $\mathbb{T}_{\kappa_{p(t)}}$ is then given via the definition $\frac{\mathbb{T}_{\kappa_{p(t)}}}{\rho} = \frac{\partial \psi}{\partial \mathbb{H}_{\kappa_{p(t)}}}(\theta, \mathbb{H}_{\kappa_{p(t)}})$, see (21), which in our case yields

$$\mathbb{T}_{\kappa_{p(t)}} = \mu(\theta) \left(e^{2\mathbb{H}_{\kappa_{p(t)}}} - \mathbb{I} \right), \tag{49}$$

where we have used Lemma A1. Equation (49) can be solved explicitly for the Hencky strain $\mathbb{H}_{\kappa_{p(t)}}$,

$$\mathbb{H}_{\kappa_{p(t)}} = \frac{1}{2} \ln \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right). \tag{50}$$

Note that if we rewrite (49) in terms of $\mathbb{B}_{\kappa_{p(t)}}$, then we get

$$\mathbb{T}_{\kappa_{p(t)}} = \mu(\theta) \left(\mathbb{B}_{\kappa_{p(t)}} - \mathbb{I} \right). \tag{51}$$

Having obtained Formula (50), we can use the definition of the specific Gibbs free energy $g = \psi - \frac{\mathbb{T}_{\kappa_{p(t)}}}{\rho} \bullet \mathbb{H}_{\kappa_{p(t)}}$, and we can write down an explicit formula for Gibbs free energy corresponding to Helmholtz free energy as introduced in (44),

$$g(\theta, \mathbb{T}_{\kappa_{p(t)}}) = g_{\text{thermal}}(\theta) + \frac{\mu(\theta)}{2\rho} \left(\text{Tr} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} \right) - \text{Tr} \ln \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right) \right) - \frac{1}{2\rho} \mathbb{T}_{\kappa_{p(t)}} \bullet \ln \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right), \tag{52}$$

which is easy to convert to the final form

$$g(\theta, \mathbb{S}_{\kappa_{p(t)}}) = g_{\text{thermal}}(\theta) + \frac{\mu(\theta)}{2\rho} \text{Tr} \left(\frac{\rho}{\mu(\theta)} \mathbb{S}_{\kappa_{p(t)}} - \left[\frac{\rho}{\mu(\theta)} \mathbb{S}_{\kappa_{p(t)}} + \mathbb{I} \right] \ln \left[\frac{\rho}{\mu(\theta)} \mathbb{S}_{\kappa_{p(t)}} + \mathbb{I} \right] \right). \tag{53}$$

Since the Legendre transformation from Helmholtz free energy to Gibbs free energy is done with respect to the deformation and the temperature variable is kept intact, we see that the function $g_{\text{thermal}}(\theta)$ is identical to the function $\psi_{\text{thermal}}(\theta)$.

We note that if we use spectral representation of $\mathbb{S}_{\kappa_{p(t)}}$, then (52) can be rewritten as

$$g(\theta, \mathbb{S}_{\kappa_{p(t)}}) = g_{\text{thermal}}(\theta) + \frac{\mu(\theta)}{2\rho} \sum_{i=1}^3 \left(\frac{\rho}{\mu(\theta)} \lambda_i - \left[\frac{\rho}{\mu(\theta)} \lambda_i + 1 \right] \ln \left[\frac{\rho}{\mu(\theta)} \lambda_i + 1 \right] \right), \quad (54)$$

where $\{\lambda_i\}_{i=1}^3$ are eigenvalues of $\mathbb{S}_{\kappa_{p(t)}}$. The function $f(x) = x - (x + 1) \ln(x + 1)$ that appears on the right-hand side of (54) is a nonpositive concave function that vanishes if and only if $x = 0$. Furthermore, by appealing to (53), we see that Gibbs free energy becomes infinite once the quantity $\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}$ vanishes. This means that the quantity $\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}$ remains a positive definite matrix (this is equivalent to the positive definiteness of $\mathbb{B}_{\kappa_{p(t)}}$ in the classical approach). Finally, we observe that Gibbs free energy vanishes for $\mathbb{T}_{\kappa_{p(t)}} = \mathbb{O}$, which corresponds to the fact that the Helmholtz free energy vanishes for $\mathbb{B}_{\kappa_{p(t)}} = \mathbb{I}$.

5.2. Approach Based on Gibbs Free Energy

Let us now assume that Gibbs free energy g is given by (53), and let us rederive the governing equations (41) within the approach based on Gibbs free energy (instead of Helmholtz free energy, we start with Gibbs free energy, and we observe what steps need to be taken if we want to rederive the Giesekus model). Using the standard matrix calculus, we can observe that

$$\frac{\partial g}{\partial \mathbb{S}_{\kappa_{p(t)}}}(\theta, \mathbb{S}_{\kappa_{p(t)}}) = -\frac{1}{2} \ln \left(\frac{\rho}{\mu(\theta)} \mathbb{S}_{\kappa_{p(t)}} + \mathbb{I} \right), \quad (55)$$

which is the expected result, since the use of Legendre transformation implies that $\mathbb{H}_{\kappa_{p(t)}} = -\frac{\partial g}{\partial \mathbb{S}_{\kappa_{p(t)}}}(\theta, \mathbb{S}_{\kappa_{p(t)}})$ (compare (55) and (50)).

Now, we need to exploit the evolution equation for the entropy. In particular, we need to propose an evolution equation for $\mathbb{T}_{\kappa_{p(t)}}$ and a constitutive relation for \mathbb{T} in such a way that the entropy production is nonnegative (the thermal part—the heat flux—can be manipulated in the standard manner).

$$\rho \frac{d\eta}{dt} + \text{div} \left(\frac{j_q}{\theta} \right) = \frac{1}{\theta} \left\{ \mathbb{T}_\delta \bullet \mathbb{D}_\delta + \mathbb{T}_{\kappa_{p(t)}} \bullet \frac{d\mathbb{H}_{\kappa_{p(t)}}}{dt} \right\} - \frac{j_q \bullet \nabla \theta}{\theta^2}. \quad (56)$$

The term $\mathbb{T}_{\kappa_{p(t)}} \bullet \frac{d\mathbb{H}_{\kappa_{p(t)}}}{dt}$ can be expressed as

$$\begin{aligned} \mathbb{T}_{\kappa_{p(t)}} \bullet \frac{d\mathbb{H}_{\kappa_{p(t)}}}{dt} &= \mathbb{T}_{\kappa_{p(t)}} \bullet e^{-2\mathbb{H}_{\kappa_{p(t)}}} e^{2\mathbb{H}_{\kappa_{p(t)}}} \frac{d\mathbb{H}_{\kappa_{p(t)}}}{dt} = \frac{1}{2} \mathbb{T}_{\kappa_{p(t)}} \bullet e^{-2\mathbb{H}_{\kappa_{p(t)}}} \frac{d}{dt} e^{2\mathbb{H}_{\kappa_{p(t)}}} \\ &= \frac{1}{2} \mathbb{T}_{\kappa_{p(t)}} \bullet e^{-2\mathbb{H}_{\kappa_{p(t)}}} \bullet \left(e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}} + \mathbb{I} e^{2\mathbb{H}_{\kappa_{p(t)}}} + e^{2\mathbb{H}_{\kappa_{p(t)}}} \mathbb{I}^\top \right) = \mathbb{T}_{\kappa_{p(t)}} \bullet \mathbb{D} + \frac{1}{2} \mathbb{T}_{\kappa_{p(t)}} \bullet e^{-2\mathbb{H}_{\kappa_{p(t)}}} \bullet e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}}, \end{aligned} \quad (57)$$

while the motivation for this manipulation is to get the term $\mathbb{T}_{\kappa_{p(t)}} \bullet \mathbb{D}$, which can be conveniently manipulated in (56). In (57), we use the cyclic property of the trace and the fact that $\mathbb{T}_{\kappa_{p(t)}}$ and $\mathbb{H}_{\kappa_{p(t)}}$ commute, which implies that $\mathbb{T}_{\kappa_{p(t)}}$ and $e^{-2\mathbb{H}_{\kappa_{p(t)}}$ commute as well. If we substitute (57) into (56), we get

$$\rho \frac{d\eta}{dt} + \text{div} \left(\frac{j_q}{\theta} \right) = \frac{1}{\theta} \left\{ \left[\mathbb{T}_\delta - \mathbb{T}_{\kappa_{p(t)}, \delta} \right] \bullet \mathbb{D}_\delta - \frac{1}{2} \mathbb{T}_{\kappa_{p(t)}} \bullet e^{-2\mathbb{H}_{\kappa_{p(t)}}} \bullet e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}} \right\} - \frac{j_q \bullet \nabla \theta}{\theta^2}. \quad (58)$$

The primitive quantity in the approach based on Gibbs free energy is the reduced stress tensor $\mathbb{S}_{\kappa_{p(t)}} = \frac{\mathbb{T}_{\kappa_{p(t)}}}{\rho}$, and we need to find an evolution equation for this quantity (or an evolution equation for $\mathbb{T}_{\kappa_{p(t)}}$, since these quantities differ only by the multiplication by the density ρ , which is in our case a constant). We recall that the evolution equation for $\mathbb{T}_{\kappa_{p(t)}}$ and the constitutive relation for \mathbb{T} must be chosen in such a way that the corresponding terms on the right-hand side of (56) are nonnegative.

5.2.1. Oldroyd-B Model

Before we discuss the Giesekus model, we focus on the case $\alpha = 0$ which leads to the Oldroyd-B model. Regarding the entropy production terms in (58), the following simple choice

$$\mathbb{T}_{\kappa_{p(t)}} =_{\text{def}} -\nu_1 e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}}, \tag{59a}$$

$$\mathbb{T}_\delta =_{\text{def}} \mathbb{T}_{\kappa_{p(t)},\delta} + 2\nu_1 \mathbb{D}_\delta, \tag{59b}$$

is of particular interest. If we use (59) in (58), then we get

$$\rho \frac{d\eta}{dt} + \text{div} \left(\frac{j_q}{\theta} \right) = \frac{1}{\theta} \left\{ 2\nu_1 \mathbb{D}_\delta \bullet \mathbb{D}_\delta + \frac{\nu_1}{2} e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}} e^{-2\mathbb{H}_{\kappa_{p(t)}}} \bullet e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}} \right\} - \frac{j_q \bullet \nabla \theta}{\theta^2}, \tag{60}$$

Thus, the entropy production in nonnegative mechanical processes is required. Indeed, the term in the entropy production can be rewritten as

$$\begin{aligned} e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}} e^{-2\mathbb{H}_{\kappa_{p(t)}}} \bullet e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}} &= \text{Tr} \left[e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}} e^{-2\mathbb{H}_{\kappa_{p(t)}}} e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}} \right] \\ &= \text{Tr} \left[e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}} e^{-\mathbb{H}_{\kappa_{p(t)}}} e^{-\mathbb{H}_{\kappa_{p(t)}}} e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}} \right] = e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}} e^{-\mathbb{H}_{\kappa_{p(t)}}} \bullet e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}} e^{-\mathbb{H}_{\kappa_{p(t)}}}, \end{aligned} \tag{61}$$

or as

$$\frac{\nu_1}{2} e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}} e^{-2\mathbb{H}_{\kappa_{p(t)}}} \bullet e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}} = \frac{1}{2\nu_1} \mathbb{T}_{\kappa_{p(t)}} e^{-2\mathbb{H}_{\kappa_{p(t)}}} \bullet \mathbb{T}_{\kappa_{p(t)}}, \tag{62}$$

which makes its nonnegativity self-evident (we have exploited the symmetry of all involved matrices). The term can be also understood as a weighted scalar product of matrices $e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}}$ with the weight $e^{-2\mathbb{H}_{\kappa_{p(t)}}} = \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1}$, where we have used the fact that

$$\mathbb{H}_{\kappa_{p(t)}} = -\frac{\partial g}{\partial \mathbb{S}_{\kappa_{p(t)}}}(\theta, \mathbb{S}_{\kappa_{p(t)}})$$

and the particular formula for Gibbs free energy; see (55).

Note that the last observation implies that if we use constitutive assumptions (59) and (55), then the entropy evolution Equation (60) rewritten in terms of $\mathbb{T}_{\kappa_{p(t)}}$ reads

$$\rho \frac{d\eta}{dt} + \text{div} \left(\frac{j_q}{\theta} \right) = \frac{1}{\theta} \left\{ 2\nu_1 \mathbb{D}_\delta \bullet \mathbb{D}_\delta + \frac{1}{2\nu_1} \mathbb{T}_{\kappa_{p(t)}} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} \bullet \mathbb{T}_{\kappa_{p(t)}} \right\} - \frac{j_q \bullet \nabla \theta}{\theta^2}. \tag{63}$$

In this case, the nonnegativity of the entropy production in mechanical processes still holds—Equation (63) is the same as Equation (60) but rewritten in terms of different variables. However, the nonnegativity of the entropy production is less straightforward to recognize than in the formulation based on $\mathbb{H}_{\kappa_{p(t)}}$.

If we further assume that the heat flux is given by the standard Fourier law,

$$j_q = -\kappa \nabla \theta, \tag{64}$$

where κ is a nonnegative constant, then the entropy production both in thermal and mechanical processes is nonnegative and the second law of thermodynamics holds.

Once we have identified the energy storage mechanisms—the specific Gibbs free energy (53)—and the entropy production mechanisms—the terms on the right-hand side of the entropy evolution Equation (60)—the complete system of governing equations is obtained as follows. First, we use (59a) and (49), and we can conclude that the evolution equation for $\mathbb{H}_{\kappa_{p(t)}}$ reads

$$-\nu_1 \overline{e^{2\mathbb{H}_{\kappa_{p(t)}}}} = \mu(\theta) \left(e^{2\mathbb{H}_{\kappa_{p(t)}}} - \mathbb{I} \right), \tag{65}$$

which can also be rewritten in terms of $\mathbb{T}_{\kappa_{p(t)}}$ as

$$\nu_1 \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} \right) + \mathbb{T}_{\kappa_{p(t)}} = 2\nu_1 \mathbb{D}. \tag{66}$$

In the latter case, it suffices to take the upper convected derivative of (49) and use (59a). We recall that the definition of the upper convected derivative implies that $\overset{\nabla}{\mathbb{I}} = -2\mathbb{D}$. Finally, if we wanted to, we could go back to the left Cauchy–Green tensor $\mathbb{B}_{\kappa_{p(t)}}$ (see (46)), which would allow us to rewrite (65) as

$$\nu_1 \overline{\mathbb{B}_{\kappa_{p(t)}}} = -\mu(\theta) \left(\mathbb{B}_{\kappa_{p(t)}} - \mathbb{I} \right). \tag{67}$$

This is the evolution equation (41c) for $\alpha = 0$.

If we use (66), then the system of governing equations for the mechanical quantities reads

$$\operatorname{div} \mathbf{v} = 0, \tag{68a}$$

$$\rho \frac{d\mathbf{v}}{dt} = \operatorname{div} \mathbb{T} + \rho \mathbf{b}, \tag{68b}$$

$$\nu_1 \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} \right) + \mathbb{T}_{\kappa_{p(t)}} = 2\nu_1 \mathbb{D}, \tag{68c}$$

where the Cauchy stress tensor \mathbb{T} is given by the formula

$$\mathbb{T} = m\mathbb{I} + 2\nu\mathbb{D}_\delta + \mathbb{T}_{\kappa_{p(t)}, \delta}. \tag{68d}$$

Since (68c) can be rewritten as (67), and since (49) holds—that is, $\mathbb{T}_{\kappa_{p(t)}} = \mu(\theta) \left(e^{2\mathbb{H}_{\kappa_{p(t)}}} - \mathbb{I} \right)$ holds—we see that (68) is equivalent to the mechanical part of (41) (a minor redefinition of the mean normal stress m is necessary. However, this is an inconsequential modification of the governing equations).

Regarding the temperature evolution equation (41d), we have to start with the entropy evolution equation either in the form (60) or in the form (63). In particular, we need to show that the entropy production identified in (60) or equivalently in (63) is tantamount to the entropy production (45) that leads to the Oldroyd-B model (we recall that Oldroyd-B model corresponds to $\alpha = 0$). The critical term can be manipulated as follows:

$$\begin{aligned} \frac{\nu_1}{2} \overline{e^{2\mathbb{H}_{\kappa_{p(t)}}}} e^{-2\mathbb{H}_{\kappa_{p(t)}}} \bullet \overline{e^{2\mathbb{H}_{\kappa_{p(t)}}}} &= \frac{\nu_1}{2} \overline{\mathbb{B}_{\kappa_{p(t)}}} \mathbb{B}_{\kappa_{p(t)}}^{-1} \bullet \overline{\mathbb{B}_{\kappa_{p(t)}}} = \frac{\nu_1}{2} \frac{\mu(\theta)}{\nu_1} \left(\mathbb{B}_{\kappa_{p(t)}} - \mathbb{I} \right) \mathbb{B}_{\kappa_{p(t)}}^{-1} \bullet \frac{\mu(\theta)}{\nu_1} \left(\mathbb{B}_{\kappa_{p(t)}} - \mathbb{I} \right) \\ &= \frac{\mu(\theta)^2}{2\nu_1} \operatorname{Tr} \left[\mathbb{B}_{\kappa_{p(t)}}^{-1} \left(\mathbb{B}_{\kappa_{p(t)}} - \mathbb{I} \right)^2 \right]. \end{aligned} \tag{69}$$

Making use of identity (43), we see that the entropy production mechanisms postulated in (60) or (63) are identical to those postulated in the standard derivation of the Oldroyd-B model (45). Note that (69) explicitly shows that the entropy production for the Oldroyd-B fluid can be equivalently written down either in terms of an algebraic formula for $\mathbb{B}_{\kappa_{p(t)}}$ or in terms of algebraic formula to the rates $\frac{\nabla}{\mathbb{B}_{\kappa_{p(t)}}$.

Since the entropy production mechanisms ξ are now confirmed to be the same in both approaches, we see that the right-hand side of the entropy evolution equation

$$\rho \frac{d\eta}{dt} + \operatorname{div} \left(\frac{\mathbf{j}_q}{\theta} \right) = \xi \quad (70)$$

is the same both for the model derived via Gibbs free energy and Helmholtz free energy. Finally, it suffices to recall that the the entropy η is given in terms of derivatives of the potentials as $\eta(\theta, \mathbb{S}_{\kappa_{p(t)}}) = -\frac{\partial g}{\partial \theta}(\theta, \mathbb{S}_{\kappa_{p(t)}})$ and $\eta(\theta, \mathbb{B}_{\kappa_{p(t)}}) = -\frac{\partial \psi}{\partial \theta}(\theta, \mathbb{B}_{\kappa_{p(t)}})$; see (29a) and (19). Consequently, if we substitute η into (70), we get the same evolution equation for the temperature for both approaches (in the case of Gibbs free energy, the primitive variable will be the reduced stress tensor $\mathbb{S}_{\kappa_{p(t)}}$, while in the case of Helmholtz free energy, the primitive variable will be the left Cauchy–Green tensor associated to the elastic response $\mathbb{B}_{\kappa_{p(t)}}$). For the convenience of readers, both approaches are compared in Summaries 1 and 2.

Summary 1: Incompressible Oldroyd-B model via Gibbs free energy

Specific Gibbs free energy:

$$g(\theta, \mathbb{S}_{\kappa_{p(t)}}) =_{\text{def}} g_{\text{thermal}}(\theta) + \frac{\mu(\theta)}{2\rho} \text{Tr} \left(\frac{\rho}{\mu(\theta)} \mathbb{S}_{\kappa_{p(t)}} - \left[\frac{\rho}{\mu(\theta)} \mathbb{S}_{\kappa_{p(t)}} + \mathbb{I} \right] \ln \left[\frac{\rho}{\mu(\theta)} \mathbb{S}_{\kappa_{p(t)}} + \mathbb{I} \right] \right)$$

$$g_{\text{thermal}}(\theta) =_{\text{def}} -c_{V,\text{ref}} \theta \left(\ln \frac{\theta}{\theta_{\text{ref}}} - 1 \right)$$

Reduced stress (notation):

$$\mathbb{S}_{\kappa_{p(t)}} =_{\text{def}} \frac{\mathbb{T}_{\kappa_{p(t)}}}{\rho}$$

Entropy production:

$$\zeta = \frac{1}{\theta} \left\{ 2\nu \mathbb{D}_\delta \bullet \mathbb{D}_\delta + \frac{1}{2\nu_1} \mathbb{T}_{\kappa_{p(t)}} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} \bullet \mathbb{T}_{\kappa_{p(t)}} \right\} + \kappa \frac{\nabla \theta \bullet \nabla \theta}{\theta^2}$$

Material parameters: $c_{V,\text{ref}}$ specific heat at constant volume—positive constant; $\mu(\theta)$ shear modulus—nonnegative function, typically proportional to θ ; ν, ν_1 viscosity—nonnegative functions of the primitive variables, typically constants; κ thermal conductivity—nonnegative function of the primitive variables, typically constant

Evolution equations (mechanical variables $m, \mathbf{v}, \mathbb{T}_{\kappa_{p(t)}}$ and thermal variable θ):

$$\text{div } \mathbf{v} = 0$$

$$\rho \frac{d\mathbf{v}}{dt} = \text{div } \mathbb{T} + \rho \mathbf{b}$$

$$\nu_1 \overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} \right)} + \mathbb{T}_{\kappa_{p(t)}} = 2\nu_1 \mathbb{D}$$

$$-\rho \theta \frac{d}{dt} \left(\frac{\partial g}{\partial \theta}(\theta, \mathbb{S}_{\kappa_{p(t)}}) \right) = \text{div}(\kappa \theta) + 2\nu \mathbb{D}_\delta \bullet \mathbb{D}_\delta + \frac{1}{2\nu_1} \mathbb{T}_{\kappa_{p(t)}} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} \bullet \mathbb{T}_{\kappa_{p(t)}}$$

Cauchy stress tensor:

$$\mathbb{T} = m \mathbb{I} + 2\nu \mathbb{D}_\delta + \mathbb{T}_{\kappa_{p(t)}, \delta}$$

Thermodynamical relations:

$$\mathbb{H}_{\kappa_{p(t)}} = -\frac{\partial g}{\partial \mathbb{S}_{\kappa_{p(t)}}}(\theta, \mathbb{S}_{\kappa_{p(t)}})$$

$$\eta = -\frac{\partial g}{\partial \theta}(\theta, \mathbb{S}_{\kappa_{p(t)}})$$

Left Cauchy–Green tensor $\mathbb{B}_{\kappa_{p(t)}}$ and Hencky strain tensor $\mathbb{H}_{\kappa_{p(t)}}$ associated to the elastic response:

$$\mathbb{B}_{\kappa_{p(t)}} = e^{2\mathbb{H}_{\kappa_{p(t)}}$$

Summary 2: Incompressible Oldroyd-B model via Helmholtz free energy

Specific Helmholtz free energy:

$$\psi(\theta, \mathbb{B}_{\kappa_{p(t)}}) =_{\text{def}} \psi_{\text{thermal}}(\theta) + \frac{\mu(\theta)}{2\rho} \left(\text{Tr} \mathbb{B}_{\kappa_{p(t)}} - 3 - \ln \det \mathbb{B}_{\kappa_{p(t)}} \right)$$

$$\psi_{\text{thermal}}(\theta) =_{\text{def}} -c_{V,\text{ref}}\theta \left(\ln \frac{\theta}{\theta_{\text{ref}}} - 1 \right).$$

Entropy production:

$$\zeta = \frac{1}{\theta} \left\{ 2\nu \mathbb{D}_\delta \bullet \mathbb{D}_\delta + \frac{\mu(\theta)^2}{2\nu_1} \text{Tr} \left[\mathbb{B}_{\kappa_{p(t)}} + \mathbb{B}_{\kappa_{p(t)}}^{-1} - 2\mathbb{I} \right] \right\} + \kappa \frac{\nabla\theta \bullet \nabla\theta}{\theta^2}$$

Material parameters: $c_{V,\text{ref}}$ specific heat at constant volume—positive constant; $\mu(\theta)$ shear modulus—nonnegative function, typically proportional to θ ; ν, ν_1 viscosity—nonnegative functions of the primitive variables, typically constants; κ thermal conductivity—nonnegative function of the primitive variables, typically constant

Evolution equations (mechanical variables $m, \mathbf{v}, \mathbb{B}_{\kappa_{p(t)}}$ and thermal variable θ):

$$\text{div } \mathbf{v} = 0$$

$$\rho \frac{d\mathbf{v}}{dt} = \text{div } \mathbb{T} + \rho \mathbf{b}$$

$$\nu_1 \frac{\overset{\nabla}{\mathbb{B}}_{\kappa_{p(t)}}}{dt} = -\mu(\theta) \left[\mathbb{B}_{\kappa_{p(t)}} - \mathbb{I} \right]$$

$$-\rho\theta \frac{d}{dt} \left(\frac{\partial\psi}{\partial\theta}(\theta, \mathbb{B}_{\kappa_{p(t)}}) \right) = \text{div}(\kappa\theta) + 2\nu \mathbb{D}_\delta \bullet \mathbb{D}_\delta + \frac{\mu(\theta)^2}{2\nu_1} \text{Tr} \left[\mathbb{B}_{\kappa_{p(t)}} + \mathbb{B}_{\kappa_{p(t)}}^{-1} - 2\mathbb{I} \right]$$

Cauchy stress tensor:

$$\mathbb{T} = m\mathbb{I} + 2\nu \mathbb{D}_\delta + \mu(\theta) \left(\mathbb{B}_{\kappa_{p(t)}} \right)_\delta$$

Thermodynamical relations:

$$\eta = -\frac{\partial\psi}{\partial\theta}(\theta, \mathbb{B}_{\kappa_{p(t)}})$$

Left Cauchy–Green tensor $\mathbb{B}_{\kappa_{p(t)}}$ and Hencky strain tensor $\mathbb{H}_{\kappa_{p(t)}}$ associated to the elastic response:

$$\mathbb{B}_{\kappa_{p(t)}} = e^{2\mathbb{H}_{\kappa_{p(t)}}}$$

5.2.2. Giesekus Model

Regarding the Giesekus model, the derivation in the approach based on Gibbs free energy is a slight modification of the derivation of the Oldroyd-B model presented above. The Gibbs free energy remains the same, see (53), and the entropy evolution equation remains the same (58) as well, that is

$$\rho \frac{d\eta}{dt} + \text{div} \left(\frac{j_q}{\theta} \right) = \frac{1}{\theta} \left\{ \left[\mathbb{T}_\delta - \mathbb{T}_{\kappa_{p(t)}, \delta} \right] \bullet \mathbb{D}_\delta - \frac{1}{2} \mathbb{T}_{\kappa_{p(t)}} e^{-2\mathbb{H}_{\kappa_{p(t)}}} \bullet e^{\frac{\overset{\nabla}{\mathbb{H}}_{\kappa_{p(t)}}}{2}} \right\} - \frac{j_q \bullet \nabla\theta}{\theta^2}. \quad (71)$$

The entropy production mechanisms are however modified to

$$\rho \frac{d\eta}{dt} + \text{div} \left(\frac{j_q}{\theta} \right) = \frac{1}{\theta} \left\{ 2\nu \mathbb{D}_\delta \bullet \mathbb{D}_\delta + \frac{1}{2\nu_1} \mathbb{T}_{\kappa_{p(t)}} e^{-2\mathbb{H}_{\kappa_{p(t)}}} \bullet \left[\mathbb{T}_{\kappa_{p(t)}} \left(\mathbb{I} + \frac{\alpha}{\mu(\theta)} \mathbb{T}_{\kappa_{p(t)}} \right) \right] \right\} + \kappa \frac{\nabla\theta \bullet \nabla\theta}{\theta^2}, \quad (72)$$

which implies constitutive relations

$$\mathbf{j}_q \stackrel{\text{def}}{=} -\kappa \nabla \theta, \tag{73a}$$

$$\mathbb{T}_{\kappa_{p(t)}} \left(\mathbb{I} + \frac{\alpha}{\mu(\theta)} \mathbb{T}_{\kappa_{p(t)}} \right) \stackrel{\text{def}}{=} -\nu_1 e^{-\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}}, \tag{73b}$$

$$\mathbb{T}_\delta \stackrel{\text{def}}{=} \mathbb{T}_{\kappa_{p(t)}, \delta} + 2\nu \mathbb{D}_\delta. \tag{73c}$$

It is straightforward to check that the entropy production specified in (72) is nonnegative. Indeed, the critical term can be manipulated as follows:

$$\begin{aligned} \mathbb{T}_{\kappa_{p(t)}} e^{-2\mathbb{H}_{\kappa_{p(t)}}} \bullet \left[\mathbb{T}_{\kappa_{p(t)}} \left(\mathbb{I} + \frac{\alpha}{\mu(\theta)} \mathbb{T}_{\kappa_{p(t)}} \right) \right] &= \mathbb{T}_{\kappa_{p(t)}} e^{-2\mathbb{H}_{\kappa_{p(t)}}} \bullet \mathbb{T}_{\kappa_{p(t)}} \left[\alpha \left(\mathbb{I} + \frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} \right) + (1 - \alpha) \mathbb{I} \right] \\ &= (1 - \alpha) \mathbb{T}_{\kappa_{p(t)}} e^{-2\mathbb{H}_{\kappa_{p(t)}}} \bullet \mathbb{T}_{\kappa_{p(t)}} + \alpha \mathbb{T}_{\kappa_{p(t)}} e^{-2\mathbb{H}_{\kappa_{p(t)}}} \bullet \left(\mathbb{I} + \frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} \right) \mathbb{T}_{\kappa_{p(t)}} \\ &= (1 - \alpha) \mathbb{T}_{\kappa_{p(t)}} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} \bullet \mathbb{T}_{\kappa_{p(t)}} + \alpha \mathbb{T}_{\kappa_{p(t)}} \bullet \mathbb{T}_{\kappa_{p(t)}}, \end{aligned} \tag{74}$$

where we have used the relation (50) between the stress tensor $\mathbb{T}_{\kappa_{p(t)}}$ and the Hencky strain tensor $\mathbb{H}_{\kappa_{p(t)}}$. The first term on the right-hand side of (74) is up to a positive factor $(1 - \alpha)$ the same as the term in the entropy production for the Oldroyd-B model (see (63)), while the nonnegativity of the term $\alpha \mathbb{T}_{\kappa_{p(t)}} \bullet \mathbb{T}_{\kappa_{p(t)}}$ is evident (note that the entropy production is particularly simple if we set $\alpha = 1$). Using the same steps as in the previous section, it is then straightforward to check that the constitutive relations (73) lead, once rewritten in terms of $\mathbb{B}_{\kappa_{p(t)}}$, to the governing equations for the standard Giesekus viscoelastic rate-type fluid.

To conclude, we see that the Giesekus viscoelastic rate-type fluid is specified by Gibbs free energy in the form

$$g(\theta, \mathbb{S}_{\kappa_{p(t)}}) \stackrel{\text{def}}{=} g_{\text{thermal}}(\theta) + \frac{\mu(\theta)}{2\rho} \text{Tr} \left(\frac{\rho}{\mu(\theta)} \mathbb{S}_{\kappa_{p(t)}} - \left[\frac{\rho}{\mu(\theta)} \mathbb{S}_{\kappa_{p(t)}} + \mathbb{I} \right] \ln \left[\frac{\rho}{\mu(\theta)} \mathbb{S}_{\kappa_{p(t)}} + \mathbb{I} \right] \right) \tag{75}$$

$$g_{\text{thermal}}(\theta) \stackrel{\text{def}}{=} -c_{V,\text{ref}} \theta \left(\ln \frac{\theta}{\theta_{\text{ref}}} - 1 \right) \tag{76}$$

where $\mathbb{S}_{\kappa_{p(t)}} \stackrel{\text{def}}{=} \frac{\mathbb{T}_{\kappa_{p(t)}}}{\rho}$, while the entropy production takes the form

$$\zeta = \frac{1}{\theta} \left\{ 2\nu \mathbb{D}_\delta \bullet \mathbb{D}_\delta + \frac{1}{2\nu_1} \left[(1 - \alpha) \mathbb{T}_{\kappa_{p(t)}} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} \bullet \mathbb{T}_{\kappa_{p(t)}} + \alpha \mathbb{T}_{\kappa_{p(t)}} \bullet \mathbb{T}_{\kappa_{p(t)}} \right] \right\} + \kappa \frac{\nabla \theta \bullet \nabla \theta}{\theta^2}. \tag{77}$$

5.3. Approach Based on Gibbs Free Energy and Entropy Production Maximization

In brief, the hypothesis of entropy production maximization (see Rajagopal and Srinivasa [6]) states that once the entropy production ability of the material is specified in terms of entropy production ζ given as a function of thermodynamics affinities/fluxes, and once all other structural constraints are taken into account, then the constitutive relations for fluxes/affinities as functions of affinities/fluxes must be chosen in such a way that the entropy production is maximized by this choice of fluxes/affinities (see also Málek and Průša [16] for an explanatory presentation and calculated examples). In the remainder of this section, we show how the procedure works for the Giesekus/Oldroyd-B models in the approach based on Gibbs free energy.

Note however that the terminology affinity/flux, albeit the standard one, might be seen as inappropriate for several reasons; see Rajagopal and Srinivasa [90]. In the current contribution, we therefore take the liberty of using the term resultant instead of affinity

and the term determinant instead of flux; see Rajagopal and Srinivasa [90] for the rationale behding this terminology.

5.3.1. Oldroyd-B Model

The Gibbs free energy remains the same as in the preceding sections (see (53)), and the entropy evolution equation remains the same (58) as well; that is,

$$\rho \frac{d\eta}{dt} + \text{div} \left(\frac{j_q}{\theta} \right) = \frac{1}{\theta} \left\{ \left[\mathbb{T}_\delta - \mathbb{T}_{\kappa_{p(t)}, \delta} \right] \bullet \mathbb{D}_\delta - \frac{1}{2} \mathbb{T}_{\kappa_{p(t)}} e^{-2\mathbb{H}_{\kappa_{p(t)}}} \bullet e^{\frac{\nabla}{2\mathbb{H}_{\kappa_{p(t)}}}} \right\} - \frac{j_q \bullet \nabla \theta}{\theta^2}. \quad (78)$$

In virtue of thermodynamic relation $\mathbb{H}_{\kappa_{p(t)}} = -\frac{\partial g}{\partial \mathbb{S}_{\kappa_{p(t)}}}(\theta, \mathbb{S}_{\kappa_{p(t}})$, for the given Gibbs free energy (53), we get

$$\mathbb{T}_{\kappa_{p(t)}} = \mu(\theta) \left(e^{2\mathbb{H}_{\kappa_{p(t)}}} - \mathbb{I} \right), \quad (79)$$

See Section 5.2 for details. This relation allows us to rewrite (78) as

$$\rho \frac{d\eta}{dt} + \text{div} \left(\frac{j_q}{\theta} \right) = \frac{1}{\theta} \left\{ \left[\mathbb{T}_\delta - \mathbb{T}_{\kappa_{p(t)}, \delta} \right] \bullet \mathbb{D}_\delta - \frac{1}{2} \mathbb{T}_{\kappa_{p(t)}} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} \bullet \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{\nabla} \right\} - \frac{j_q \bullet \nabla \theta}{\theta^2}. \quad (80)$$

This is the entropy production as implied by the choice of Gibbs free energy (53) (note that (80) is, unlike (78), written down in terms of the stress tensor $\mathbb{T}_{\kappa_{p(t)}}$). The rate-type quantities can be identified as resultants,

$$\mathbb{A}_1 =_{\text{def}} \mathbb{D}, \quad (81a)$$

$$\mathbb{A}_2 =_{\text{def}} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{\nabla} \quad (81b)$$

while the corresponding quantities in the dot products in (80) can be identified as determinants,

$$\mathbb{J}_1 =_{\text{def}} \mathbb{T}_\delta - \mathbb{T}_{\kappa_{p(t)}, \delta}, \quad (82a)$$

$$\mathbb{J}_2 =_{\text{def}} -\frac{1}{2} \mathbb{T}_{\kappa_{p(t)}} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} \quad (82b)$$

We also have the classical determinant–resultant pair j_q and $\nabla \theta$. On the other hand, the assumption regarding the specification of the entropy production for the Oldroyd-B model is the following:

$$\zeta = \frac{1}{\theta} \left\{ 2\nu \mathbb{D}_\delta \bullet \mathbb{D}_\delta + \frac{\nu_1}{2} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{\nabla} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} \bullet \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{\nabla} \right\} + \kappa \frac{\nabla \theta \bullet \nabla \theta}{\theta^2}, \quad (83)$$

See also (63) and the subsequent discussion (we need to rewrite (77) in such a way that it contains the resultants identified in (81)). The constitutive relations we need to identify are the constitutive relations for the Cauchy stress tensor \mathbb{T} and the stress tensor $\mathbb{T}_{\kappa_{p(t)}}$ and the heat flux j_q .

The entropy production maximization procedure requires us to maximize the entropy production ζ given by (83) with respect to resultants (81) and $\nabla \theta$, subject to the constraint

$$\zeta = \frac{1}{\theta} \left\{ \left[\mathbb{T}_\delta - \mathbb{T}_{\kappa_{p(t)}, \delta} \right] \bullet \mathbb{D}_\delta - \frac{1}{2} \mathbb{T}_{\kappa_{p(t)}} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} \bullet \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{\nabla} \right\} - \frac{j_q \bullet \nabla \theta}{\theta^2}, \quad (84)$$

as implied by the right-hand side of (80). The constrained maximization problem therefore reads

$$\max_{\mathbb{A}_1, \mathbb{A}_2, \nabla\theta} \zeta \quad \text{subject to} \quad \zeta - \frac{1}{\theta} \{ \mathbb{A}_1 \bullet \mathbb{J}_1 + \mathbb{A}_2 \bullet \mathbb{J}_2 \} + \frac{j_q \bullet \nabla\theta}{\theta^2} = 0, \quad (85)$$

where we have used the determinant/resultant notation introduced in (81) and (82). The auxiliary functional for the constrained maximisation problem reads

$$\Phi =_{\text{def}} \zeta + \lambda \left(\zeta - \frac{1}{\theta} \{ \mathbb{A}_1 \bullet \mathbb{J}_1 + \mathbb{A}_2 \bullet \mathbb{J}_2 \} + \frac{j_q \bullet \nabla\theta}{\theta^2} \right), \quad (86)$$

where λ is the Lagrange multiplier. The necessary conditions for the extremum are $\frac{\partial\Phi}{\partial\mathbb{A}_1} = \mathbb{0}$, $\frac{\partial\Phi}{\partial\mathbb{A}_2} = \mathbb{0}$ and $\frac{\partial\Phi}{\partial\nabla\theta} = \mathbb{0}$, which translates to

$$\frac{1 + \lambda}{\lambda} \frac{\partial\zeta}{\partial\mathbb{A}_1} = \frac{\mathbb{J}_1}{\theta}, \quad (87a)$$

$$\frac{1 + \lambda}{\lambda} \frac{\partial\zeta}{\partial\mathbb{A}_2} = \frac{\mathbb{J}_2}{\theta}, \quad (87b)$$

$$\frac{1 + \lambda}{\lambda} \frac{\partial\zeta}{\partial\nabla\theta} = -\frac{j_q}{\theta^2}. \quad (87c)$$

Let us first solve (87) for the Lagrange multiplier λ . Once we identify the value of the Lagrange multiplier λ , it is straightforward to find the determinants j_q , \mathbb{J}_1 and \mathbb{J}_2 for which the entropy production attains its maximum. Taking the dot product of individual equations in (87) with the corresponding resultant and taking the sum of these equations yields

$$\frac{1 + \lambda}{\lambda} \left(\frac{\partial\zeta}{\partial\mathbb{A}_1} \bullet \mathbb{A}_1 + \frac{\partial\zeta}{\partial\mathbb{A}_2} \bullet \mathbb{A}_2 + \frac{\partial\zeta}{\partial\nabla\theta} \bullet \nabla\theta \right) = \frac{1}{\theta} \{ \mathbb{A}_1 \bullet \mathbb{J}_1 + \mathbb{A}_2 \bullet \mathbb{J}_2 \} - \frac{j_q \bullet \nabla\theta}{\theta^2}. \quad (88)$$

Making use of constraint (84), we see that the right-hand side of (88) is equal to ζ . Furthermore, the direct differentiation of ζ given by the formula (83) yields

$$\frac{\partial\zeta}{\partial\mathbb{A}_1} = \frac{1}{\theta} 4\nu\mathbb{D}, \quad (89a)$$

$$\frac{\partial\zeta}{\partial\mathbb{A}_2} = \frac{1}{\theta} \frac{\nu_1}{2} \left[\overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} + \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} \overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)} \right], \quad (89b)$$

$$\frac{\partial\zeta}{\partial\nabla\theta} = 2\kappa \frac{\nabla\theta}{\theta^2}, \quad (89c)$$

which implies that

$$\frac{\partial\zeta}{\partial\mathbb{A}_1} \bullet \mathbb{A}_1 + \frac{\partial\zeta}{\partial\mathbb{A}_2} \bullet \mathbb{A}_2 + \frac{\partial\zeta}{\partial\nabla\theta} \bullet \nabla\theta = 2\zeta \quad (90)$$

This is not surprising since the entropy production is “quadratic” in resultants. Making use of (90) in (88) allows us to easily identify the Lagrange multiplier λ . Indeed (88) implies that $2 \frac{1+\lambda}{\lambda} \zeta = \zeta$; thus,

$$\frac{1 + \lambda}{\lambda} = \frac{1}{2}. \quad (91)$$

Having obtained the formula for the Lagrange multiplier, we can go back to (87) and find formulae for the determinants. We see that Equation (87) immediately implies that

$$\mathbb{T}_\delta - \mathbb{T}_{\kappa_{p(t)}, \delta} = 2\nu \mathbb{D}_\delta, \tag{92a}$$

$$\mathbf{j}_q = -\kappa \nabla \theta, \tag{92b}$$

$$\frac{\nu_1}{4} \left[\overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} + \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} \overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)} \right] = -\frac{1}{2} \mathbb{T}_{\kappa_{p(t)}} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1}. \tag{92c}$$

If (92c) holds, then the matrices $\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1}$ and $\overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)}$ commute, which means that (92c) in fact simplifies to

$$\nu_1 \overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)} = -\mathbb{T}_{\kappa_{p(t)}}. \tag{93}$$

This is the evolution equation for $\mathbb{T}_{\kappa_{p(t)}}$ in the case of Oldroyd-B model, see (66) and the discussion following this equation.

In order to show that the matrices $\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1}$ and $\overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)}$ commute, we need to show that they share the same eigenvectors. Let us assume that \mathbf{v} is an arbitrary eigenvector of $\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}$ associated to the eigenvalue $\tilde{\lambda}$ (since the matrix is symmetric positive definite, we know that the normalized eigenvectors form an orthonormal basis, and that the eigenvalues are positive. This also guarantees that the matrix is invertible). Such an eigenvector is also an eigenvector of matrix $\mathbb{T}_{\kappa_{p(t)}}$ with eigenvalue $\mu(\theta)(\tilde{\lambda} - 1)$. Making use of (92c), we see that

$$\frac{\nu_1}{4} \left[\overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)} \frac{1}{\tilde{\lambda}} + \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} \overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)} \right] \mathbf{v} = -\frac{1}{2} \frac{\mu(\theta)(\tilde{\lambda} - 1)}{\tilde{\lambda}} \mathbf{v}, \tag{94}$$

hence

$$\overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)} \mathbf{v} = -\frac{4}{\nu_1} \left(\frac{1}{\tilde{\lambda}} \mathbb{I} + \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} \right)^{-1} \frac{1}{2} \frac{\mu(\theta)(\tilde{\lambda} - 1)}{\tilde{\lambda}} \mathbf{v}, \tag{95}$$

which shows that $\overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)} \mathbf{v} = -\frac{\mu(\theta)}{\nu_1} (\tilde{\lambda} - 1) \mathbf{v}$. This manipulations shows that an arbitrary eigenvector of $\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}$ is also an eigenvector of $\overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)}$, as we set out to prove.

5.3.2. Giesekus Model

The derivation of the Giesekus models proceeds along the same lines as the derivation of the Oldroyd-B model. The energy storage mechanism characterized by the postulated Gibbs free energy is the same both for the Oldroyd-B model and the Giesekus model; thus, the constraint (84) is the same. The only difference is in the postulated entropy production mechanisms. Instead of the entropy production ζ given by (83), we now set

$$\zeta = \frac{1}{\theta} \left\{ 2\nu \mathbb{D}_\delta \bullet \mathbb{D}_\delta + \frac{\nu_1}{2} \overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)^{-1} \left[\alpha \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right) + (1 - \alpha) \mathbb{I} \right]^{-1} \bullet \overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I} \right)} \right\} + \kappa \frac{\nabla \theta \bullet \nabla \theta}{\theta^2}. \tag{96}$$

Few observations regarding (96) should be presented. First, if we set $\alpha = 0$, then we recover the postulated entropy production ζ for the Oldroyd-B model; see (83). Second, since the matrix $\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}$ is positive definite, and since $\alpha \in [0, 1]$, we see that the postulated entropy production is nonnegative. In fact, the critical term in the entropy production has the structure of a weighted dot product of matrices $\overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}\right)}$ that represent the resultant \mathbb{A}_2 , see (81b).

The constrained maximization problem (85) is now solved by the same procedure as in the case of Oldroyd-B model (the identification of determinants and resultants remains the same). In particular, the “quadratic” structure of the entropy production allows one to easily identify the Lagrange multiplier λ as a solution to the equation $\frac{1+\lambda}{\lambda} = \frac{1}{2}$. The counterpart of (92c) is then

$$\frac{\nu_1}{4} \left[\overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}\right)} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}\right)^{-1} \left[\alpha \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}\right) + (1 - \alpha)\mathbb{I} \right]^{-1} + \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}\right)^{-1} \left[\alpha \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}\right) + (1 - \alpha)\mathbb{I} \right]^{-1} \overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}\right)} \right] = -\frac{1}{2} \mathbb{T}_{\kappa_{p(t)}} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}\right)^{-1}, \tag{97}$$

and regarding the matrices on the left-hand side, it can be shown again that they commute. Equation (97) therefore collapses to

$$\nu_1 \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}\right)^{-1} \left[\alpha \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}\right) + (1 - \alpha)\mathbb{I} \right]^{-1} \overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}\right)} = -\mathbb{T}_{\kappa_{p(t)}} \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}\right)^{-1}, \tag{98}$$

which can be rewritten as

$$\nu_1 \overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}\right)} = -\mathbb{T}_{\kappa_{p(t)}} \left[\alpha \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}\right) + (1 - \alpha)\mathbb{I} \right] \tag{99}$$

or as

$$\nu_1 \overline{\left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \mathbb{I}\right)} = -\mu(\theta) \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)} + \alpha \left(\frac{\mathbb{T}_{\kappa_{p(t)}}}{\mu(\theta)}\right)^2 \right). \tag{100}$$

A simple substitution of (51) then reveals that (100) is tantamount to the standard evolution equation for $\mathbb{B}_{\kappa_{p(t)}}$ used in the Giesekus model; see (41c) and (42).

6. Conclusions

We have shown how a novel approach to the constitutive relations for elastic solids can be embedded in the development of mathematical models for viscoelastic fluids, and we have discussed in detail the thermodynamic underpinning of such models. In particular, we have focused on the use of Gibbs free energy instead of Helmholtz free energy. Using the standard Giesekus/Oldroyd-B models as examples, we have shown how the alternative approach works in the case of well-known models and in the fully non-isothermal setting. The proposed approach is straightforward to generalize to more complex settings wherein the classical approach based on the Gibbs potential might be impractical or even inapplicable.

Author Contributions: Both authors have contributed equally to the conceptualization, methodology and writing of the manuscript. Both authors have read and agreed to the published version of the manuscript.

Funding: V.P. thanks the Czech Science Foundation for the support through the project 20-11027X. K.R.R. thanks the Office of Naval Research for its support.

Acknowledgments: V.P. acknowledges the membership of the Nečas Center for Mathematical Modeling (NCMM).

Conflicts of Interest: The authors declare no conflict of interest.

Appendix A. Daleckii–Krein Formula and Its Consequences

Since we frequently need to differentiate matrix valued functions, it is worth recalling that the derivatives of matrix valued functions are easy to obtain using the Daleckii–Krein formula; see Daletskii and Krein [91] (short proofs of the same are given in Bhatia [92] (Theorem 5.3.1) and Bhatia [93] (Theorem V.3.3)). Regarding the matrix valued functions and especially tools for computations thereof, we refer the reader to Higham [94].

Theorem A1 (Daleckii–Krein formula). *Let \mathbb{A} be real symmetric matrix in $\mathbb{R}^{k \times k}$ with spectral decomposition $\mathbb{A} = \sum_{i=1}^k \lambda_i \mathbb{P}_i$, where $\{\lambda_i\}_{i=1}^k$ are the eigenvalues of \mathbb{A} , and $\{\mathbb{P}_i\}_{i=1}^k$ denotes the projection operator to the i -th (normalized) eigenvector \mathbf{v}_i ; that is, $\mathbb{P}_i =_{\text{def}} \mathbf{v}_i \otimes \mathbf{v}_i$. Let f be a continuously differentiable real function defined on the open set containing the spectrum of \mathbb{A} . Then, the corresponding matrix valued function \mathfrak{f} ,*

$$\mathfrak{f}(\mathbb{A}) =_{\text{def}} \sum_{i=1}^k f(\lambda_i) \mathbb{P}_i, \tag{A1}$$

is differentiable, and the Gateaux derivative of \mathfrak{f} at point \mathbb{A} in the direction \mathbb{X} is given by the formula

$$D_{\mathbb{A}} \mathfrak{f}(\mathbb{A})[\mathbb{X}] = \sum_{i=1}^k f'(\lambda_i) \mathbb{P}_i \mathbb{X} \mathbb{P}_i + \sum_{i=1}^k \sum_{\substack{j=1 \\ j \neq i}}^k \frac{f(\lambda_i) - f(\lambda_j)}{\lambda_i - \lambda_j} \mathbb{P}_i \mathbb{X} \mathbb{P}_j. \tag{A2}$$

Furthermore, let $[\mathbb{A} \circ \mathbb{B}]_{ij} =_{\text{def}} A_{ij} B_{ij}$ denote the element-wise Schur/Hadamard product of matrices \mathbb{A} and \mathbb{B} (no summation convention). Using the Schur product, we can rewrite (A2) as

$$[D_{\mathbb{A}} \mathfrak{f}(\mathbb{A})[\mathbb{X}]]_{ij} = \frac{f(\lambda_i) - f(\lambda_j)}{\lambda_i - \lambda_j} X_{ij} \tag{A3}$$

provided that the Schur product is taken in a basis in which \mathbb{A} is diagonal, and for $i = j$, the quotient in (A3) is interpreted as $f'(\lambda_i)$.

Note that in the continuum mechanics literature, the same formulae have been rediscovered independently in special cases; see for example Jog [95] for the case of the matrix logarithm. Using the Daleckii–Krein formula, it is straightforward to show the following two lemmas, which we have used frequently in our analysis.

Lemma A1. *Let \mathbb{A} and \mathbb{B} be real symmetric matrices in $\mathbb{R}^{k \times k}$ that commute; that is, $\mathbb{A}\mathbb{B} = \mathbb{B}\mathbb{A}$. Let f be a continuously differentiable real function defined on the open set containing the spectrum of \mathbb{A} , and let \mathfrak{f} be the matrix valued function associated with f , see (A1). Then,*

$$\frac{\partial}{\partial \mathbb{A}} \text{Tr}[\mathbb{B}\mathfrak{f}(\mathbb{A})] = \mathbb{B}\mathfrak{f}'(\mathbb{A}), \tag{A4}$$

where \mathfrak{f}' is the matrix valued function associated to f' ; that is,

$$\mathfrak{f}'(\mathbb{A}) = \sum_{i=1}^k f'(\lambda_i) \mathbb{P}_i, \tag{A5}$$

where $\mathbb{A} = \sum_{i=1}^k \lambda_i \mathbb{P}_i$ denotes the spectral decomposition of \mathbb{A} .

Proof. Since the matrices \mathbb{A} and \mathbb{B} are symmetric matrices and they commute, they share the same set of eigenvectors $\{\mathbf{v}_i\}_{i=1}^3$, and can be simultaneously diagonalized (they are diagonal in the same basis). Their spectral decomposition is $\mathbb{A} = \sum_{i=1}^k \lambda_i \mathbb{P}_i$ and $\mathbb{B} = \sum_{i=1}^k \mu_i \mathbb{P}_i$, where $\{\lambda_i\}_{i=1}^k$ and $\{\mu_i\}_{i=1}^k$ denote the eigenvalues of \mathbb{A} and \mathbb{B} respectively. Using the basis wherein the matrices have the diagonal form, the left-hand side of (A4) reads

$$\begin{aligned} \left[\frac{\partial}{\partial \mathbb{A}} \text{Tr}[\mathbb{B}f(\mathbb{A})] \right]_{op} &= \frac{\partial}{\partial A_{op}} \left[\sum_{m,n=1}^k \mu_m \delta_{mn} [f(\mathbb{A})]_{mn} \right] = \sum_{m,n=1}^k \mu_m \delta_{mn} \frac{\partial [f(\mathbb{A})]_{mn}}{\partial A_{op}} \\ &= \sum_{m,n=1}^k \mu_m \delta_{mn} \frac{f(\lambda_m) - f(\lambda_n)}{\lambda_m - \lambda_n} \delta_{mo} \delta_{np} = \sum_{o=1}^k \mu_o f'(\lambda_o) \delta_{op} = \mathbb{B}f'(\mathbb{A}). \end{aligned} \tag{A6}$$

□

Lemma A2. Suppose that \mathbb{A} and \mathbb{B} are real symmetric matrices in $\mathbb{R}^{k \times k}$ that are differentiable functions with respect to $t \in \mathbb{R}$. Furthermore, suppose that these matrices commute for all $t \in \mathbb{R}$, that is $\mathbb{A}\mathbb{B} = \mathbb{B}\mathbb{A}$, and let f be a continuously differentiable real function defined on the open set containing the spectrum of \mathbb{A} at every $t \in \mathbb{R}$. Then,

$$\text{Tr} \left[\mathbb{B} \frac{df(\mathbb{A})}{dt} \right] = \text{Tr} \left[\mathbb{B}f'(\mathbb{A}) \frac{d\mathbb{A}}{dt} \right], \tag{A7}$$

where f' is the matrix valued function associated to f' , see (A5).

Proof. We use the chain rule, Dalecki–Krein formula and spectral decomposition $\mathbb{A} = \sum_{i=1}^k \lambda_i \mathbb{P}_i$ and $\mathbb{B} = \sum_{i=1}^k \mu_i \mathbb{P}_i$,

$$\begin{aligned} \text{Tr} \left[\mathbb{B} \frac{df(\mathbb{A})}{dt} \right] &= \text{Tr} \left[\mathbb{B} \frac{\partial f(\mathbb{A})}{\partial \mathbb{A}} \left[\frac{d\mathbb{A}}{dt} \right] \right] \\ &= \text{Tr} \left[\left(\sum_{l=1}^k \mu_l \mathbb{P}_l \right) \left(\sum_{i=1}^k f'(\lambda_i) \mathbb{P}_i \frac{d\mathbb{A}}{dt} \mathbb{P}_i + \sum_{i=1}^k \sum_{\substack{j=1 \\ j \neq i}}^k \frac{f(\lambda_i) - f(\lambda_j)}{\lambda_i - \lambda_j} \mathbb{P}_i \frac{d\mathbb{A}}{dt} \mathbb{P}_j \right) \right] \\ &= \text{Tr} \left[\sum_{i=1}^k \mu_i f'(\lambda_i) \mathbb{P}_i \frac{d\mathbb{A}}{dt} \mathbb{P}_i \right] = \text{Tr} \left[\mathbb{B}f'(\mathbb{A}) \frac{d\mathbb{A}}{dt} \right], \end{aligned} \tag{A8}$$

where we have used the fact that $\mathbb{P}_i \mathbb{P}_j = \delta_{ij} \mathbb{P}_j$ and the properties of trace (no summation). □

Note that the identity does not in general hold without the trace. For example, it is not in general true that $\frac{de^{\mathbb{A}}}{dt} = e^{\mathbb{A}} \frac{d\mathbb{A}}{dt}$.

References

1. Truesdell, C.; Noll, W. *The Non-Linear Field Theories of Mechanics*, 3rd ed.; Springer: Berlin, Germany, 2004.
2. Müller, I. *Thermodynamics; Interaction of Mechanics and Mathematics*; Pitman: London, UK, 1985.
3. Coleman, B.D.; Noll, W. An approximation theorem for functionals, with applications in continuum mechanics. *Arch. Ration. Mech. Anal.* **1960**, *6*, 355–370. [[CrossRef](#)]
4. Rajagopal, K.R. On implicit constitutive theories. *Appl. Math.* **2003**, *48*, 279–319. [[CrossRef](#)]
5. Rajagopal, K.R. On implicit constitutive theories for fluids. *J. Fluid Mech.* **2006**, *550*, 243–249. [[CrossRef](#)]
6. Rajagopal, K.R.; Srinivasa, A.R. On thermomechanical restrictions of continua. *Proc. R. Soc. Lond. Ser. A Math. Phys. Eng. Sci.* **2004**, *460*, 631–651. [[CrossRef](#)]
7. Le Roux, C.; Rajagopal, K.R. Shear flows of a new class of power-law fluids. *Appl. Math.* **2013**, *58*, 153–177. [[CrossRef](#)]
8. Málek, J.; Průša, V.; Rajagopal, K.R. Generalizations of the Navier–Stokes fluid from a new perspective. *Int. J. Eng. Sci.* **2010**, *48*, 1907–1924. [[CrossRef](#)]

9. Boltenhagen, P.; Hu, Y.; Matthys, E.F.; Pine, D.J. Observation of bulk phase separation and coexistence in a sheared micellar solution. *Phys. Rev. Lett.* **1997**, *79*, 2359–2362. [[CrossRef](#)]
10. Grob, M.; Heussinger, C.; Zippelius, A. Jamming of frictional particles: A nonequilibrium first-order phase transition. *Phys. Rev. E* **2014**, *89*, 050201. [[CrossRef](#)] [[PubMed](#)]
11. Mari, R.; Seto, R.; Morris, J.F.; Denn, M.M. Nonmonotonic flow curves of shear thickening suspensions. *Phys. Rev. E* **2015**, *91*, 052302. [[CrossRef](#)] [[PubMed](#)]
12. Perláčová, T.; Průša, V. Tensorial implicit constitutive relations in mechanics of incompressible non-Newtonian fluids. *J. Non-Newton. Fluid Mech.* **2015**, *216*, 13–21. [[CrossRef](#)]
13. Janečka, A.; Průša, V. Perspectives on using implicit type constitutive relations in the modelling of the behaviour of non-Newtonian fluids. *AIP Conf. Proc.* **2015**, *1662*, 20003. [[CrossRef](#)]
14. Blatter, H. Velocity and stress-fields in grounded glaciers—A simple algorithm for including deviatoric stress gradients. *J. Glaciol.* **1995**, *41*, 333–344. [[CrossRef](#)]
15. Pettit, E.C.; Waddington, E.D. Ice flow at low deviatoric stress. *J. Glaciol.* **2003**, *49*, 359–369. [[CrossRef](#)]
16. Málek, J.; Průša, V. Derivation of equations for continuum mechanics and thermodynamics of fluids. In *Handbook of Mathematical Analysis in Mechanics of Viscous Fluids*; Giga, Y., Novotný, A., Eds.; Springer: Berlin/Heidelberg, Germany, 2018; pp. 3–72. [[CrossRef](#)]
17. Bingham, C.E. *Fluidity and Plasticity*; McGraw–Hill: New York, NY, USA, 1922.
18. Herschel, W.H.; Bulkley, R. Konsistenzmessungen von Gummi-Benzollösungen. *Colloid Polym. Sci.* **1926**, *39*, 291–300. [[CrossRef](#)]
19. Duvaut, G.; Lions, J.L. *Inequalities in Mechanics and Physics*; Springer: Berlin, Germany, 1976.
20. Rajagopal, K.R.; Srinivasa, A.R. On the thermodynamics of fluids defined by implicit constitutive relations. *Z. Angew. Math. Phys.* **2008**, *59*, 715–729. [[CrossRef](#)]
21. Bulíček, M.; Gwiazda, P.; Málek, J.; Świerczewska-Gwiazda, A. On unsteady flows of implicitly constituted incompressible fluids. *SIAM J. Math. Anal.* **2012**, *44*, 2756–2801. [[CrossRef](#)]
22. Srinivasan, S.; Karra, S. Flow of “stress power-law” fluids between parallel rotating discs with distinct axes. *Int. J. Non-Linear Mech.* **2015**, *74*, 73–83. [[CrossRef](#)]
23. Narayan, S.P.A.; Rajagopal, K.R. Unsteady flows of a class of novel generalizations of the Navier–Stokes fluid. *Appl. Math. Comput.* **2013**, *219*, 9935–9946. [[CrossRef](#)]
24. Fusi, L.; Farina, A. Flow of a class of fluids defined via implicit constitutive equation down an inclined plane: Analysis of the quasi-steady regime. *Eur. J. Mech. B Fluids* **2017**, *61*, 200–208. [[CrossRef](#)]
25. Fusi, L. Channel flow of viscoplastic fluids with pressure-dependent rheological parameters. *Phys. Fluids* **2018**, *30*, 073102. [[CrossRef](#)]
26. Housiadas, K.D.; Georgiou, G.C.; Tanner, R.I. A note on the unbounded creeping flow past a sphere for Newtonian fluids with pressure-dependent viscosity. *Int. J. Eng. Sci.* **2015**, *86*, 1–9. [[CrossRef](#)]
27. Gomez-Constante, J.P.; Rajagopal, K.R. Flow of a new class of non-Newtonian fluids in tubes of non-circular cross-sections. *Proc. R. Soc. A Math. Phys. Eng. Sci.* **2019**, *377*, 20180069. [[CrossRef](#)] [[PubMed](#)]
28. Fetecau, C.; Bridges, C. Analytical solutions for some unsteady flows of fluids with linear dependence of viscosity on the pressure. *Inverse Probl. Sci. Eng.* **2020**, *378*–395. [[CrossRef](#)]
29. Janečka, A.; Málek, J.; Průša, V.; Tierra, G. Numerical scheme for simulation of transient flows of non-Newtonian fluids characterised by a non-monotone relation between the symmetric part of the velocity gradient and the Cauchy stress tensor. *Acta Mech.* **2019**, *230*, 729–747. [[CrossRef](#)]
30. Diening, L.; Kreuzer, C.; Süli, E. Finite element approximation of steady flows of incompressible fluids with implicit power-law-like rheology. *SIAM J. Numer. Anal.* **2013**, *51*, 984–1015. [[CrossRef](#)]
31. Stebel, J. Finite element approximation of Stokes-like systems with implicit constitutive relation. In Proceedings of the Conference ALGORITMY, Podbanské, Slovakia, 9–14 September 2012; pp. 291–300.
32. Hirn, A.; Lanzendörfer, M.; Stebel, J. Finite element approximation of flow of fluids with shear-rate- and pressure-dependent viscosity. *IMA J. Numer. Anal.* **2012**, *32*, 1604–1634. [[CrossRef](#)]
33. Süli, E.; Tscherpel, T. Fully discrete finite element approximation of unsteady flows of implicitly constituted incompressible fluids. *IMA J. Numer. Anal.* **2019**, *40*, 801–849. [[CrossRef](#)]
34. Farrell, P.E.; Gazca-Orozco, P.A.; Süli, E. Numerical analysis of unsteady implicitly constituted incompressible fluids: 3-field formulation. *SIAM J. Num. Anal.* **2020**, *58*, 757–787. [[CrossRef](#)]
35. Farrell, P.E.; Gazca-Orozco, P.A. An augmented lagrangian preconditioner for implicitly constituted non-Newtonian incompressible flow. *SIAM J. Sci. Comp.* **2020**, *42*, B1329–B1349. [[CrossRef](#)]
36. Bulíček, M.; Gwiazda, P.; Málek, J.; Świerczewska-Gwiazda, A. On steady flows of incompressible fluids with implicit power-law-like rheology. *Adv. Calc. Var.* **2009**, *2*, 109–136. [[CrossRef](#)]
37. Maringová, E.; Žabenský, J. On a Navier–Stokes–Fourier-like system capturing transitions between viscous and inviscid fluid regimes and between no-slip and perfect-slip boundary conditions. *Nonlinear Anal.-Real World Appl.* **2018**, *41*, 152–178. [[CrossRef](#)]
38. Blechta, J.; Málek, J.; Rajagopal, K.R. On the classification of incompressible fluids and a mathematical analysis of the equations that govern their motion. *SIAM J. Math. Anal.* **2020**, *52*, 1232–1289. [[CrossRef](#)]

39. Průša, V.; Rajagopal, K.R. On implicit constitutive relations for materials with fading memory. *J. Non-Newton. Fluid Mech.* **2012**, *181–182*, 22–29. [[CrossRef](#)]
40. Karra, S.; Průša, V.; Rajagopal, K.R. On Maxwell fluids with relaxation time and viscosity depending on the pressure. *Int. J. Non-Linear Mech.* **2011**, *46*, 819–827. [[CrossRef](#)]
41. Kannan, K.; Rajagopal, K.R. A model for the flow of rock glaciers. *Int. J. Non-Linear Mech.* **2013**, *48*, 59–64. [[CrossRef](#)]
42. Housiadas, K.D. Internal viscoelastic flows for fluids with exponential type pressure-dependent viscosity and relaxation time. *J. Rheol.* **2015**, *59*, 769–791. [[CrossRef](#)]
43. Housiadas, K.D. Viscoelastic fluids with pressure-dependent viscosity; exact analytical solutions and their singularities in Poiseuille flows. *Int. J. Eng. Sci.* **2020**, *147*, 103207. [[CrossRef](#)]
44. Arcangioli, M.; Farina, A.; Fusi, L.; Saccomandi, G. Constitutive equations with pressure-dependent rheological parameters for describing ice creep. *J. Glaciol.* **2019**, *65*, 557–564. [[CrossRef](#)]
45. Morgan, A.J.A. Some properties of media defined by constitutive equations in implicit form. *Int. J. Eng. Sci.* **1966**, *4*, 155–178. [[CrossRef](#)]
46. Fitzgerald, J.E. A tensorial Hencky measure of strain and strain rate for finite deformations. *J. Appl. Phys.* **1980**, *51*, 5111–5115. [[CrossRef](#)]
47. Blume, J.A. On the form of the inverted stress-strain law for isotropic hyperelastic solids. *Int. J. Non-Linear Mech.* **1992**, *27*, 413–421. [[CrossRef](#)]
48. Xiao, H.; Chen, L.S. Hencky's logarithmic strain and dual stress-strain and strain-stress relations in isotropic finite hyperelasticity. *Int. J. Solids Struct.* **2003**, *40*, 1455–1463. [[CrossRef](#)]
49. Xiao, H.; Bruhns, O.; Meyers, A. Explicit dual stress-strain and strain-stress relations of incompressible isotropic hyperelastic solids via deviatoric Hencky strain and Cauchy stress. *Acta Mech.* **2004**, *168*, 21–33. [[CrossRef](#)]
50. Bustamante, R.; Rajagopal, K.R. A review of implicit constitutive theories to describe the response of elastic bodies. In *Constitutive Modelling of Solid Continua*; Merodio, J., Ogden, R., Eds.; Springer: Cham, Switzerland, 2020; pp. 187–230. [[CrossRef](#)]
51. Rajagopal, K.R.; Srinivasa, A.R. An implicit three-dimensional model for describing the inelastic response of solids undergoing finite deformation. *Z. Angew. Math. Phys.* **2016**, *67*, 86. [[CrossRef](#)]
52. Rajagopal, K.R.; Srinivasa, A.R. Inelastic response of solids described by implicit constitutive relations with nonlinear small strain elastic response. *Int. J. Plast.* **2015**, *71*, 1–9. [[CrossRef](#)]
53. Cichra, D.; Průša, V. A thermodynamic basis for implicit rate-type constitutive relations describing the inelastic response of solids undergoing finite deformation. *Math. Mech. Solids* **2020**, *25*, 1081286520932205. [[CrossRef](#)]
54. Sfyris, D.; Bustamante, R. Use of some theorems related with the tensor equation $AX + XA = H$ for some classes of implicit constitutive relations. *Q. J. Mech. Appl. Math.* **2013**, *66*, 157–163. [[CrossRef](#)]
55. Truesdell, C.; Moon, H. Inequalities sufficient to ensure semi-invertibility of isotropic functions. *J. Elast.* **1975**, *5*, 183–189. [[CrossRef](#)]
56. Muliana, A.; Rajagopal, K.R.; Tscharnuter, D.; Schritterser, B.; Saccomandi, G. Determining material properties of natural rubber using fewer material moduli in virtue of a novel constitutive approach for elastic bodies. *Rubber Chem. Technol.* **2018**, *91*, 375–389. [[CrossRef](#)]
57. Xiao, H.; Bruhns, O.T.; Meyers, A. Elastoplasticity beyond small deformations. *Acta Mech.* **2006**, *182*, 31–111. [[CrossRef](#)]
58. Fattal, R.; Kupferman, R. Constitutive laws for the matrix-logarithm of the conformation tensor. *J. Non-Newton. Fluid Mech.* **2004**, *123*, 281–285. [[CrossRef](#)]
59. Fattal, R.; Kupferman, R. Time-dependent simulation of viscoelastic flows at high Weissenberg number using the log-conformation representation. *J. Non-Newton. Fluid Mech.* **2005**, *126*, 23–37. [[CrossRef](#)]
60. Hulsen, M.A.; Fattal, R.; Kupferman, R. Flow of viscoelastic fluids past a cylinder at high Weissenberg number: Stabilized simulations using matrix logarithms. *J. Non-Newton. Fluid Mech.* **2005**, *127*, 27–39. [[CrossRef](#)]
61. Rajagopal, K.R.; Srinivasa, A.R. A Gibbs-potential-based formulation for obtaining the response functions for a class of viscoelastic materials. *Proc. R. Soc. A Math. Phys. Eng. Sci.* **2011**, *467*, 39–58. [[CrossRef](#)]
62. Srinivasa, A.R. On a class of Gibbs potential-based nonlinear elastic models with small strains. *Acta Mech.* **2015**, *226*, 571–583. [[CrossRef](#)]
63. Gokulnath, C.; Saravanan, U.; Rajagopal, K.R. Representations for implicit constitutive relations describing non-dissipative response of isotropic materials. *Z. Angew. Math. Phys.* **2017**, *68*, 129. [[CrossRef](#)]
64. Průša, V.; Rajagopal, K.R.; Tůma, K. Gibbs free energy based representation formula within the context of implicit constitutive relations for elastic solids. *Int. J. Non-Linear Mech.* **2020**, *121*, 103433. [[CrossRef](#)]
65. Snoeijer, J.H.; Pandey, A.; Herrada, M.A.; Eggers, J. The relationship between viscoelasticity and elasticity. *Proc. R. Soc. A Math. Phys. Eng. Sci.* **2020**, *476*, 20200419. [[CrossRef](#)] [[PubMed](#)]
66. Eckart, C. The thermodynamics of irreversible processes. IV. The theory of elasticity and anelasticity. *Phys. Rev.* **1948**, *73*, 373–382. [[CrossRef](#)]
67. Rajagopal, K.R.; Srinivasa, A.R. A thermodynamic frame work for rate type fluid models. *J. Non-Newton. Fluid Mech.* **2000**, *88*, 207–227. [[CrossRef](#)]
68. Málek, J.; Rajagopal, K.R.; Tůma, K. A thermodynamically compatible model for describing the response of asphalt binders. *Int. J. Pavement Eng.* **2015**, *16*, 297–314. [[CrossRef](#)]

69. Narayan, S.P.A.; Little, D.N.; Rajagopal, K.R. Modelling the nonlinear viscoelastic response of asphalt binders. *Int. J. Pavement Eng.* **2016**, *17*, 123–132. [[CrossRef](#)]
70. Málek, J.; Rajagopal, K.R.; Tůma, K. A thermodynamically compatible model for describing asphalt binders: Solutions of problems. *Int. J. Pavement Eng.* **2016**, *17*, 550–564. [[CrossRef](#)]
71. Málek, J.; Rajagopal, K.R.; Tůma, K. Derivation of the variants of the Burgers model using a thermodynamic approach and appealing to the concept of evolving natural configurations. *Fluids* **2018**, *3*. [[CrossRef](#)]
72. Tůma, K.; Stein, J.; Průša, V.; Friedmann, E. Motion of the vitreous humour in a deforming eye–fluid–structure interaction between a nonlinear elastic solid and viscoelastic fluid. *Appl. Math. Comput.* **2018**, *335*, 50–64. [[CrossRef](#)]
73. Málek, J.; Průša, V.; Skřivan, T.; Süli, E. Thermodynamics of viscoelastic rate-type fluids with stress diffusion. *Phys. Fluids* **2018**, *30*, 023101. [[CrossRef](#)]
74. Řehoř, M.; Gansen, A.; Sill, C.; Polińska, P.; Westermann, S.; Dheur, J.; Baller, J.; Hale, J.S. A comparison of constitutive models for describing the flow of uncured styrene-butadiene rubber. *J. Non-Newton. Fluid Mech.* **2020**, *286*, 104398. [[CrossRef](#)]
75. Sumith, S.; Kannan, K.; Shankar, K. A constitutive model for bentonite–water mixture and the effect of wall slip boundary conditions on its mechanical response. *Int. J. Non-Linear Mech.* **2020**, *119*, 103318. [[CrossRef](#)]
76. Leonov, A.I. Nonequilibrium thermodynamics and rheology of viscoelastic polymer media. *Rheol. Acta* **1976**, *15*, 85–98. [[CrossRef](#)]
77. Leonov, A.I. On a class of constitutive equations for viscoelastic liquids. *J. Non-Newton. Fluid Mech.* **1987**, *25*, 1–59. [[CrossRef](#)]
78. Wapperom, P.; Hulsen, M.A. Thermodynamics of viscoelastic fluids: The temperature equation. *J. Rheol.* **1998**, *42*, 999–1019. [[CrossRef](#)]
79. Grmela, M.; Öttinger, H.C. Dynamics and thermodynamics of complex fluids. I. Development of a general formalism. *Phys. Rev. E* **1997**, *56*, 6620–6632. [[CrossRef](#)]
80. Öttinger, H.C.; Grmela, M. Dynamics and thermodynamics of complex fluids. II. Illustrations of a general formalism. *Phys. Rev. E* **1997**, *56*, 6633–6655. [[CrossRef](#)]
81. Pavelka, M.; Klika, V.; Grmela, M. *Multiscale Thermo-Dynamics*; de Gruyter: Berlin, Germany, 2018.
82. Málek, J.; Rajagopal, K.R.; Tůma, K. On a variant of the Maxwell and Oldroyd-B models within the context of a thermodynamic basis. *Int. J. Non-Linear Mech.* **2015**, *76*, 42–47. [[CrossRef](#)]
83. Narayan, S.P.A.; Little, D.N.; Rajagopal, K.R. Nonlinear viscoelastic model for describing the response of asphalt binders within the context of a Gibbs-potential-based thermodynamic framework. *J. Eng. Mech.* **2015**, *141*. [[CrossRef](#)]
84. Horgan, C.O.; Saccomandi, G. Constitutive models for compressible nonlinearly elastic materials with limiting chain extensibility. *J. Elast.* **2004**, *77*, 123–138. [[CrossRef](#)]
85. Dostálík, M.; Průša, V.; Skřivan, T. On diffusive variants of some classical viscoelastic rate-type models. *AIP Conf. Proc.* **2019**, *2107*, 20002. [[CrossRef](#)]
86. Stumpf, H.; Hoppe, U. The application of tensor algebra on manifolds to nonlinear continuum mechanics—Invited survey article. *Z. Angew. Math. Mech.* **1997**, *77*, 327–339. [[CrossRef](#)]
87. Giesekus, H. A simple constitutive equation for polymer fluids based on the concept of deformation-dependent tensorial mobility. *J. Non-Newton. Fluid Mech.* **1982**, *11*, 69–109. [[CrossRef](#)]
88. Oldroyd, J.G. On the formulation of rheological equations of state. *Proc. R. Soc. A Math. Phys. Eng. Sci.* **1950**, *200*, 523–541.
89. Hron, J.; Miloš, V.; Průša, V.; Souček, O.; Tůma, K. On thermodynamics of viscoelastic rate type fluids with temperature dependent material coefficients. *Int. J. Non-Linear Mech.* **2017**, *95*, 193–208. [[CrossRef](#)]
90. Rajagopal, K.; Srinivasa, A. Some remarks and clarifications concerning the restrictions placed on thermodynamic processes. *Int. J. Eng. Sci.* **2019**, *140*, 26–34. [[CrossRef](#)]
91. Daletskii, J.L.; Krein, S.G. Integration and differentiation of functions of Hermitian operators and applications to the theory of perturbations. In *Thirteen Papers on Functional Analysis and Partial Differential Equations*; American Mathematical Society: Providence, RI, USA, 1965; Volume 47, pp. 1–30.
92. Bhatia, R. *Positive Definite Matrices*; Princeton University Press: Princeton, NJ, USA, 2015.
93. Bhatia, R. *Matrix Analysis*; Springer: Berlin/Heidelberg, Germany, 2013; Volume 169.
94. Higham, N.J. *Functions of Matrices*; Society for Industrial and Applied Mathematics: Philadelphia, PA, USA, 2008. [[CrossRef](#)]
95. Jog, C.S. The explicit determination of the logarithm of a tensor and its derivatives. *J. Elast.* **2008**, *93*, 141–148. [[CrossRef](#)]