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General Aspects of Yield Stress Fluids – Terminology and Definition of Viscosity

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Abstract:
This work contributes to general theoretical aspects of yield stress fluids with significance for practical phenomenological material modeling. It introduces a terminology so that the material class 'yield stress fluid' is defined and can be distinguished from the terms 'solid' and 'liquid'. This new material classification is based on two criteria, the equilibrium relation and the flow function. In line with this terminology, an experimental procedure for classifying the material behavior is presented. The second key aspect of this paper is a discussion on the proper definition of the term 'viscosity'. The benefit of the differential viscosity over the dynamic viscosity in case of non-Newtonian fluids in general is worked out. This is shown by the most elementary yield stress fluid, the friction element, because it is the basis of the yield stress concept. Its constitutive equations are given for positive as well as negative strain rates and are also able to represent the preyield behavior. The theory presented in this article is also applied to the Maxwell, Kelvin-Voigt, and Bingham element to demonstrate the working principle.

Key words:
yield stress fluid, differential viscosity, apparent viscosity, equilibrium stress, friction element, Bingham element

Supplementary Information attached and also available at http://www.complexfluids.ethz.ch/SI_AR_24_14578.pdf

1 INTRODUCTION

The material behavior of soft matter can often be described by material models belonging to the class of yield stress fluids. So, how can yield stress fluids be distinguished from classical materials – solids and liquids – in case of phenomenological modeling? This directly leads to the question of convenient classification criteria to separate solids, liquids and yield stress fluids from each other. The search for a definition of the terms solid and liquid has a long history [1]. For example Bingham [2] said "If a body is continuously deformed by a very small shearing stress, it is a liquid, whereas if the deformation stops increasing after a time, the substance is a solid". Noll defined solids and liquids relating to its symmetry properties [3–8]). But Greve [7] pointed out that there are materials which are neither solids nor fluids in the sense of Noll's definitions. There also exists a classification into solid and liquid-like behavior depending on the storage and loss modulus. Solid-like behavior occurs if $G' > G''$, otherwise liquid-like behavior [9–14]. A disadvantage of this definition becomes obvious for example in case of the Maxwell element with the elastic moduls G and the viscosity η, which has to be classified either as solid or liquid depending on the angular frequency ω. For $\omega < G/\eta$ the loss modulus is greater than the storage modulus so that the Maxell element has to be treated as fluid. On the other hand it has to be classified as liquid for $\omega > G/\eta$ since the storage modulus dominates over the loss modulus. Thus, the search of phenomenological definitions of 'solid' and 'liquid', which can be applied theoretically as well as practically, is still necessary in general and especially to define the class of yield stress fluids. Furthermore, this contribution investigates the way of defining the term 'viscosity' since the behavior of yield stress fluids is dramatically different to the one of Newtonian fluids due to yielding.

Classifying materials as well as defining 'viscosity' is essential also in practice e.g. for material modeling. To determine the class of a material behavior should be the first step before applying constitutive equations. Determining the differential viscosity enables to extract the pure viscous properties of a yield stress fluidic material specimen which is necessary to model it. That is why, this article places a great emphasise on basics of yield stress fluids in the sense of phenomenological modeling, material classification as well as the differential viscosity and is useful in terms of practical questions. It

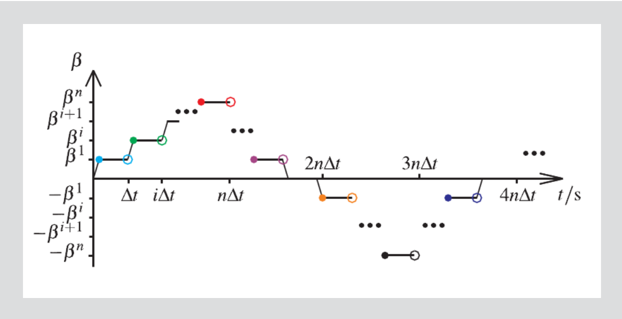

Figure 1: Determination of the equilibrium relation by the limit value of a relaxation process ($\beta = \gamma$) or creep process ($\beta = \tau$).

- introduces a terminology for yield stress fluids so that they can be distinguished from solids and liquids
- presents a measurement procedure to determine the class of a material behavior, either solid, liquid or yield stress fluid
- works out the deficit of defining the viscosity as dynamic viscosity in case of yield stress fluids and show the benefit of using the differential viscosity and
- defines the terms 'preyield' and 'postyield' for the friction and Bingham element.

The paper is outlined as follows: The first classification criterion, the equilibirum relation, is defined in Section 2. The flow function, the second classification criterion, as well as the differential viscosity are introduced by the steady state material response to a constant strain rate loading in Section 3. On this basis, the new material classification is proposed in Section 4 so that the terms 'solid', 'liquid', and 'yield stress fluid' can be distinguished. This theory is applied in Section 5 to standard and extended rheological elements to demonstrate its working principle. Therefore, the constitutive equations of the standard rheological elements are briefly summarised in Section 5.1. The advantage of the differential viscosity is demonstrated in Section 5.2. The new material classification is then applied to the standard rheological elements in Section 5.3 and extended rheological elements in Section 5.4.

In addition to these general aspects, the article uses modified constitutive equations for the friction element which ensure that the material model is valid for the preyield as well as for negative and positive strain rates. To define the terms 'preyield' and 'postyield' in case of the friction element, its constitutive equations are given in terms of the yield function, an associated flow rule, the Karush-Kuhn-Tucker and consistency conditions in Supplemental Information A. This allows for a more general representation in case of hardening. With the help of logical statements, given in Supplemental Information A.1 and A.2, the Karush-Kuhn-Tucker and consistency conditions can be reconstructed in a convenient way without discussing the optimisation with constraints in form of inequalities. The analytical expressions which are behind the figures of Section 4 to describe the behavior of solids, liquids and yield stress fluids are specified in Supplemental Information B.

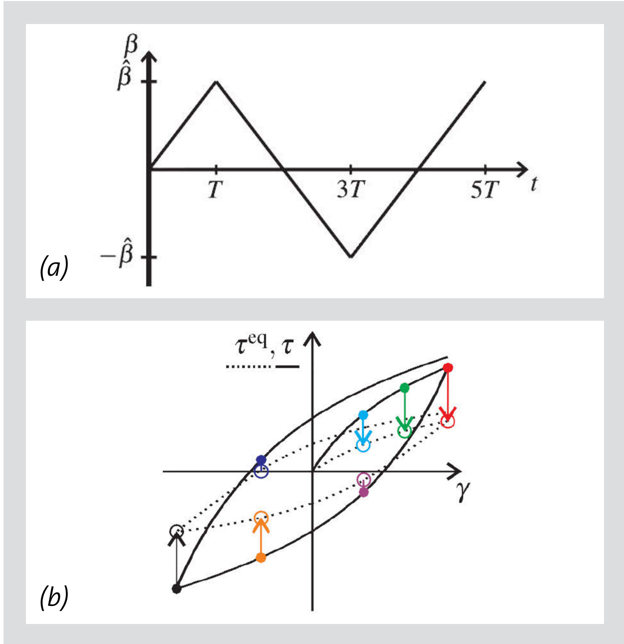

Figure 2: Total stress τ (b) of a viscoplastic material according to a cyclic loading (a) with the strain $\beta = \gamma$ as control quantity; equilibrium stress τ^{eq} (b) due to relaxation relating to Figure 1 with $\beta = \gamma$.

2 SPLIT OF THE TOTAL STRESS INTO AN EQUILIBRIUM AND OVERSTRESS

Haupt proposed a classification which divides the material behavior by examination of two classification criteria, the shape of the equilibrium relation $\tau^{eq} - \gamma^{eq}$ [6, 15, 16] and the rate dependency [6, 15, 17–20]. A state of equilibrium in this meaning is defined according to the basic assumption that a dynamic process always goes into a state of standstill if the external conditions are kept constant [6]. A stress which corresponds to a state of equilibrium in a material model is called equilibrium stress τ^{eq} [6, 15, 19]. The difference between the total stress and the equilibrium stress is defined as overstress τ^{ov} [6, 19, 20]

$$\tau := \tau^{eq} + \tau^{ov} \tag{1}$$

and vanishes by definition for the state of equilibrium. Each point ($\gamma^{eq.i}$, $\tau^{eq.i}$) on the equilibrium relation can be determined by the limit value of a relaxation process [6, 19, 21], see Figure 1 with $\beta = \gamma$, even if the equilibrium relation is zero. Here β is the control quantity of the process. For a better understanding, the total stress τ and the equilibrium stress τ^{eq} of a viscoplastic material [6] are plotted in Figure 2b for a cyclic loading [6] according to Figure 2a with $\beta = \gamma$ [6, 19, 21]. If relaxation experiments with sufficiently long holding times Δt at different strain levels γ^i are performed as it is shown in Figure 1, the total stress relaxes to the equilibrium stress as it is denoted by colored circles. In terms of theoretical and experimental observations

- relaxation ($\beta = \gamma =$ const) always implicates a state of equilibrium whereat (S1)

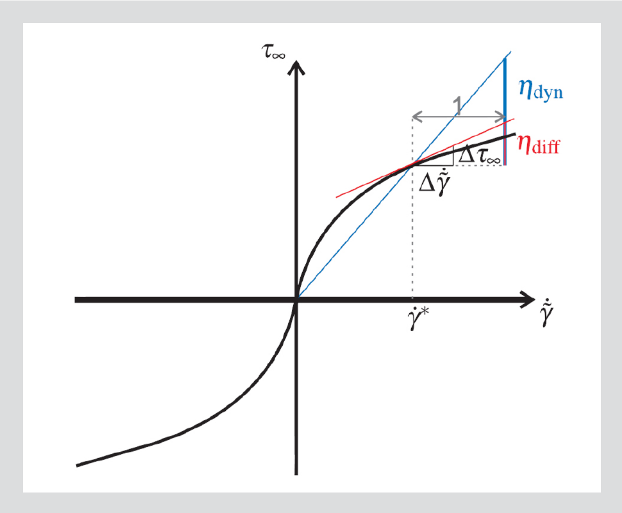

Figure 3: Difference between the dynamic and differential viscosity and equality between the differential viscosity and the coefficient of the linear term of the Taylor series which defines the slope of the flow function.

- a creep load ($\beta = \tau = \text{const}$) not always tends to an equilibrium [6, 22]. (S2)

This can be reproduced by the determination of the equilibirum of standard rheological elements in Section 5.3.

3 DIFFERENTIAL VISCOSITY DEFINED BY THE STEADY STATE MATERIAL RESPONSE TO A CONSTANT STRAIN RATE LOADING

To be able to define the differential viscosity, the steady state stress τ_∞ related to a constant strain rate $\dot{\tilde{\gamma}}$

$$\tau_\infty := \lim_{t\to\infty} \tau(t) \; for \; \{\dot{\gamma} = const\} =: \dot{\tilde{\gamma}} \tag{2}$$

has to be introduced first. This directly leads to the flow function $\tau_\infty(\dot{\tilde{\gamma}})$. The viscosity η is calculated from the flow function. It is a material parameter and cannot be measured but defined in a proper way. "It is, of course, entirely a matter of convention" [23]. Independent from the definition, the viscosity has to be an even function [24, 25]

$$\eta(-\dot{\tilde{\gamma}}) \overset{!}{=} \eta(\dot{\tilde{\gamma}}) \tag{3}$$

To ensure this, it is plotted for positive and negative strain rates in Figure 5. The apparent or dynamic viscosity is calculated by

$$\eta_{dyn} := \frac{\tau_\infty}{\dot{\tilde{\gamma}}} \tag{4}$$

A disadvantage of using the dynamic viscosity in case of yield stress fluids is that their zero viscosity

$$\eta_{o.dyn} = \lim_{\dot{\tilde{\gamma}}\to o} \frac{\tau_\infty}{\dot{\tilde{\gamma}}} = \infty \tag{5}$$

is infinite [26] if the stress does not tend to zero as fast as the strain rate. This can be explained by means of the friction and Bingham element in Section 5.2. One can get rid of this problem, if the viscosity is rather defined by the differential viscosity

$$\eta_{diff} := \frac{d\tau_\infty}{d\dot{\tilde{\gamma}}} \tag{6}$$

Another argument to prefer the differential against the dynamic viscosity in case of non-Newtonian fluids is its physical interpretation. It can be seen in Figure 3 that the increase of the stress $\Delta\tau_\infty$ due to the increase of the strain rate $\Delta\dot{\tilde{\gamma}}$ is directly related to the differential viscosity being the tangent to the flow function. Thus, it is nothing more than the linear term of the Taylor series

$$\tau_\infty(\dot{\gamma}^* + \Delta\dot{\tilde{\gamma}}) = \tau_\infty(\dot{\gamma}^*) + \underbrace{\left.\frac{d\tau_\infty}{d\dot{\tilde{\gamma}}}\right|_{\dot{\tilde{\gamma}}=\dot{\gamma}^*}}_{\eta_{diff}|_{\dot{\tilde{\gamma}}=\dot{\gamma}^*}} \Delta\dot{\tilde{\gamma}} + \ldots \tag{7}$$

and is given by $\Delta\tau_\infty = \eta_{diff}|_{\dot{\tilde{\gamma}}=\dot{\gamma}^*} \Delta\dot{\tilde{\gamma}}$. In contrast to this, the dynamic viscosity is just the secant and has no physical interpretation. This is explained in Section 5.2 on the basis of the Kelvin-Voigt, friction and Bingham element.

4 CLASSIFICATION OF MATERIAL BEHAVIOR INTO THREE TYPES: SOLIDS, LIQUIDS AND YIELD STRESS FLUIDS

In this work the definitions of solids, liquids and yield stress fluids are formulated related to the mechanical point of view of phenomenological modeling depending on two classification criteria, the equilibrium relation according to relaxation and the flow function.

Solid-like material or a solid is denoted as material, which has a non-zero equilibrium relation. The limit value τ_∞ of the stress response related to a loading with constant strain rate following Equation 2 does not exist so that the flow function is not defined (Definition D1).

The non-existent flow function is clear, since an increase of the strain by $\Delta\gamma$ is connected with an increase of the stress by $\Delta\tau$ in case of ideal solids without damage and fracture effects. That is why a constant strain rate results into a continuous increase of the stress so that no limit value τ_∞ can be reached. Furthermore, it does not matter if an equilibrium relation is determined

		Equilibrium relation according to relaxation	
		zero	non-zero
Flow function	existent	fluids	yield stress fluids
	non-existent	–	solids

Table 1: Three classes of material behavior depening on two classification criteria, the flow function and the equilibrium relation according to relaxation.

by creep or by relaxation. In both cases a non-zero equilibrium relation is found which is an observation of theoretical studies with a variety of material models but do not follow from a rigorous mathematical proof. For a detailed demonstration, an ideal viscoplastic material model is presented in Section 5.4.1.

Liquid-like material or a fluid is defined as material for which the limit value τ_∞ and thus the flow function are well defined. The equilibrium relation determined by relaxation is zero (Definition D2).

The determination of the equilibrium relation by creep is theoretically not possible for fluids because no state of standstill can be reached. A constant stress forces a fluid to flow indefinitely so that it cannot reach a time-constant strain. For practical reasons during a creep measurement the question: "Perhaps is there a time-constant strain if the time of observation Δt is increased?" would always remain as long as no time-constant strain is reached. To illustrate the working principle of the new material classification for fluids, the Maxwell element as an example of a viscoelastic fluid is discussed in Section 5.4.2.

For yield stress fluids [26–33] or materials with solid-liquid transition [28, 30, 34, 35], the equilibrium relation determined by relaxation is non-zero. The limit value τ_∞ and thus the flow function are well defined (Definition D3).

In contrast to solids, the investigation of an equilibrium point related to relaxation and creep in case of yield stress fluids may differ from each other but do not have to. Relaxation as well as creep identify the same equilibrium relation only in the preyield. In the postyield, yield stress fluids flow indefinitely related to creep. Thus, no state of standstill appears and no equilibrium relation can be identified. It is measured only by relaxation. In Section 5.4.3 the working principle of the new material classification is shown for a viscoplastic yield stress fluid.

For a better overview the three classes are ordered in Table 1. The remaining case of a zero equilibrium relation and a non-existent flow function is purely theoretical due to combinatorics. Because it has no practical relevance it is not being considered here in any way. The Definitions D1, D2, and D3 are practicable to classify one-dimensional operators so that one is able to choose a material model according to the classification. They are based on qualitative considerations related to the mechanical point of view of phenomenological modeling. However, these definitions leave the classification of generalised, three-dimensional material models open in some cases. Imagine for example an anisotropic material which behaves in one direction as fluid and in an other direction as solid. These material classification is applied to standard rheological elements, the Kelvin-Voigt, friction and Bingham element, for demonstration in Section 5.3.

5 DISCUSSION OF THE DIFFERENTIAL VISCOSITY AND DEMONSTRATION OF THE FUNCTIONALITY OF THE MATERIAL CLASSIFICATION BY STANDARD AND EXTENDED RHEOLOGICAL ELEMENTS

In the following the theoretical founded statements of this work are practically illustrated by their application to standard and extended rheological elements. First of all, the constitutive equations of the standard rheological elements are given in Section 5.1. Then, the benefit of defining a differential viscosity instead of a dynamic viscosity in case of yield stress fluids is shown in Section 5.2. The dynamic viscosity is only equal to the differential viscosity in the special case of Newtonian fluids (S3) and viscoelastic fluids [6] with linear viscous properties (S4). Finally, the functionality of the new material classification of Section 4 is described for standard rheological elments in Section 5.3 and is demonstrated for extended rheological elments in Section 5.4 by means of the investigation of the equilibrium relation (Section 2) and the flow function (Section 3).

5.1 CONSTITUTIVE EQUATIONS OF STANDARD RHEOLOGICAL ELEMENTS USED TO ILLUSTRATE THE THEORETICAL STATEMENTS OF THIS ARTICLE

The previously presented theory is illustrated in the next two sections with the help of standard rheological elements, the Kelvin-Voigt (Figure 4a), friction (Figure 4b), and Bingham element (Figure 4c). Thus, their constitutive equations are briefly summarised in this section. The one of the Kelvin-Voigt element yields [5, 6, 36]

$$\frac{\tau_{kv}}{G} = \gamma_{kv} + \frac{\eta}{G}\dot{\gamma}_{kv} \tag{8}$$

The constitutive equations of a single friction element have not often been discussed in literature. However, for this work it is inevitably necessary to investigate them, because the friction element is the basis of the yield stress concept [37–39]. Due to the lack of this investigation, the material equations of rheological elements which partially consist of a friction element are only valid for positive strain rates and are not able to ensure the equilibrium in the preyield. An example is given by the common definition of the stress of the

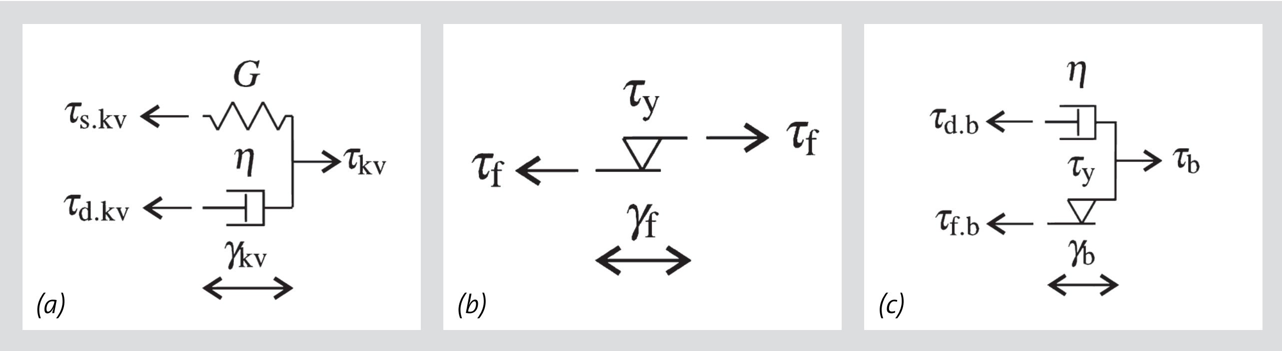

Figure 4: Defining stresses, strains and material parameters of the (a) Kelvin-Voigt, (b) friction, and (c) Bingham element used to illustrate the theoretical statements of this article.

Bingham element $\tau = \tau_y + \eta\dot{\gamma}$. Here, the stress of the friction element is given indirectly by $\tau = \tau_y$ which is only true for $\dot{\gamma} > 0$ but not for $\dot{\gamma} \leq 0$. To eliminate these disadvantages, the stress which acts on the friction element is given by

$$\tau_f = \tau_y \widetilde{sign}(\dot{\gamma}_f) \tag{9}$$

The function $\widetilde{\text{sign}}$ is related to the signum function of

$$\widetilde{sign}(\dot{\gamma}_f) = \begin{cases} -1 & : \dot{\gamma}_f < 0 \\ \xi & : \dot{\gamma}_f = 0 \;\; with \; -1 < \xi < 1 \\ 1 & : \dot{\gamma}_f > 0 \end{cases} \tag{10}$$

The definition $\widetilde{\text{sign}}(0) = \xi$ is chosen in contrast to the ordinary definition sign(0) = 0 of Equation 12 to take $-\tau_y < \tau_f < \tau_y$ into account if $\dot{\gamma}_f = 0$. In this sense the variable ξ is determined by the equilibrium so that also the material behavior in case of no plastic flow can be expressed. Thus, ξ lies in the interval $-1 < \xi < 1$. Because the inverse of $\widetilde{\text{sign}}$ is not a function, a closed relation $\dot{\gamma}_f(\tau_f)$ cannot be expressed. But it can be stated that

$$\dot{\gamma}_f = |\dot{\gamma}_f| sign(\tau_f) \; if \; |\tau_f| = \tau_y \tag{11}$$

with the ordinary defined signum function

$$sign(x) = \begin{cases} -1 & : x < 0 \\ 0 & : x = 0 \\ 1 & : x > 0 \end{cases} \tag{12}$$

$$sign(\tau_f) = sign(\dot{\gamma}_f) = \widetilde{sign}(\dot{\gamma}_f) \; if \; \{|\tau_f| = \tau_y, \dot{\gamma}_f \neq 0\} \tag{13}$$

The description of the constitutive equations in the sense of the classical theory of plasticity [6] with $F(\tau_f) = |\tau_f| - \tau_y$ (Equation A.1) is presented in Supplemental Information A, so that the terms 'preyield' and 'postyield' can be defined in case of the friction element by Equations A.11 which yield

$(F < 0) \vee [(F = 0) \wedge (\dot{F} < 0)]$: preyield
$(F = 0) \wedge [(\dot{F} = 0) \wedge (\zeta > 0)]$: postyield

The Bingham model is the most commonly used model in cement and concrete technology [40]. The constitutive equations of the Bingham element [32] follow in analogy to the one of the friction element. The stress equals

$$\tau_b = \tau_y \overline{sign}(\dot{\gamma}_b) + \eta\dot{\gamma}_b \tag{14}$$

Since Equation 14 gives $\dot{\gamma}_b = 0$ for $\tau_b = \tau_{f.b}$, no evolution of γ_b occurs if the absolute value of the applied stress $|\tau_b|$ is less than or equal the yield stress. That is why the friction element has to be described by $\widetilde{\text{sign}}$ instead of $\overline{\text{sign}}$ with

$$\overline{sign}(\dot{\gamma}_f) = \begin{cases} -1 & : \dot{\gamma}_f < 0 \\ \bar{\xi} & : \dot{\gamma}_f = 0 \;\; with \; -1 \leq \bar{\xi} \leq 1 \\ 1 & : \dot{\gamma}_f > 0 \end{cases} \tag{15}$$

According to the friction element also the variable $\bar{\xi}$ is determined by the equilibrium but lies in the interval $-1 \leq \bar{\xi} \leq 1$. The evolution of the strain rate depending on the stress is given by [41]

$$\dot{\gamma}_b = \begin{cases} 0 & : F(\tau_b) \leq 0 \\ \frac{1}{\eta}(|\tau_b| - \tau_y) sign(\tau_b) & : F(\tau_b) > 0 \end{cases} \tag{16}$$

The interpretation of the Bingham element as Perzyna type element [6, 41, 42] in analogy to the classical theory of viscoplasticity [6] leads to the description of

$$F(\tau_b) = |\tau_b| - \tau_y \begin{cases} \leq 0 & : \; preyield \\ > 0 & : \; postyield \end{cases} \tag{17}$$

in case of the Bingham element.

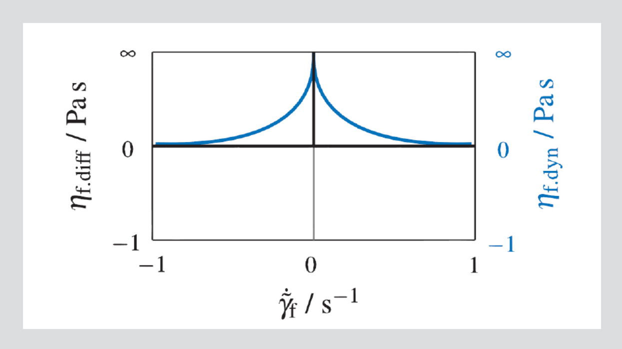

Figure 5: Differential and dynamic viscosity of the friction element.

5.2 ILLUSTRATING THE BENEFIT OF A DIFFERENTIAL VISCOSITY BY STANDARD RHEOLOGICAL ELMENTS

In this section the advantage of the differential viscosity (Section 3) in case of yield stress fluids is demonstrated. It is also shown that the differential viscosity gives the same results as the dynamic viscosity for Newtonian fluids and viscoelastic fluids [6] with linear viscous properties. Therefore, the flow function is evaluated so that the dynamic (Equation 4) and differential viscosity (Equation 6) can be determined. In case of the Kelvin-Voigt element, the flow function does not exist so that neither the dynamic nor the differential viscosity are defined. The flow function of the friction element exists and results into $\tau_{f.\infty} = \tau_y \widetilde{\text{sign}}(\dot{\tilde{\gamma}}_f)$. Thus, its differential viscosity is given by

$$\eta_{f.diff} = 2\tau_y \delta\left(\dot{\tilde{\gamma}}_f\right) \tag{18}$$

according to Equation 6 with $d\widetilde{\text{sign}}(\dot{\tilde{\gamma}}_f)/d\dot{\gamma}_f = 2\delta(\dot{\tilde{\gamma}}_f)$ and satisfies the condition of an even function (Equation 3). It is plotted in Figure 5. Here $\delta(\dot{\tilde{\gamma}}_f)$ represents the Dirac delta function. The viscosity equals zero for all non-zero strain rates. This result is mandatory because the viscosity is a material parameter which is connected to rate-dependent behavior [6,17,18], which is not the case for the friction element. In contrast to this, the dynamic viscosity of the friction element $\eta_{f.dyn}$ (Figure 5) attributes shear thinning behavior [9, 43 - 46] because it only tends to zero for $|\dot{\tilde{\gamma}}_f| \rightarrow \infty$. Here the deficit of the dynamic viscosity is obvious. Therefore, the viscosity is defined as differential viscosity for all non-Newtonian fluids. The same consequence occurs if one considers the Bingham element. The differential viscosity is given by

$$\eta_{b.diff} = 2\tau_y \delta\left(\dot{\tilde{\gamma}}_b\right) + \eta \tag{19}$$

Thus, its plot and the one of the dynamic viscosity is only shifted by η so that they are qualitatively identical to Figure 5. As for the friction element, the yield stress fluid character of the Bingham element can only be revealed if the differential viscosity is used.

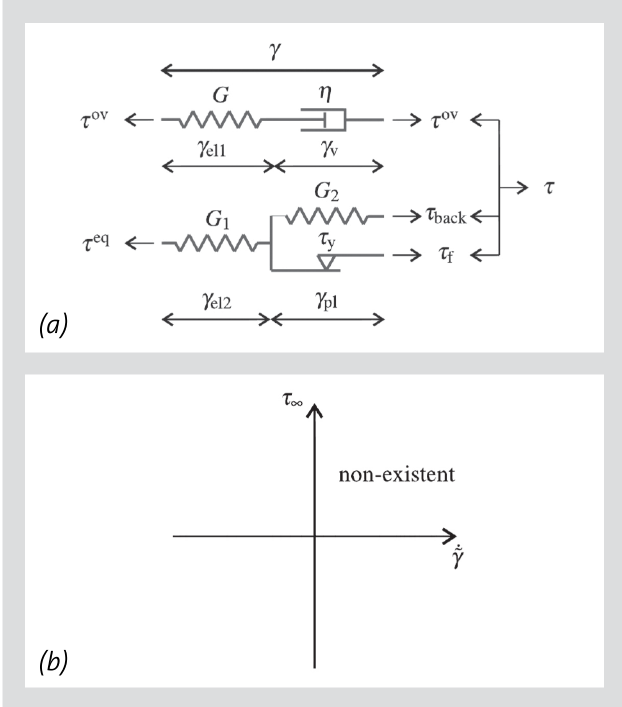

Figure 6: Example of an ideal solid (a) whose flow function is not defined (b).

5.3 APPLICATION OF THE MATERIAL CLASSIFICATION TO STANDARD RHEOLOGICAL ELEMENTS

In the following, the working principle of the material classification (Section 4) is explained. Therefore, the equilibrium relation (Section 2) and the flow function (Section 3) have to be determined. As required, the new material classification matches the literature in case of the Kelvin-Voigt element. Beside the non-existent flow function, relaxation and creep lead to non-zero equilibrium relations $\tau_{kv}{}^{eq} \equiv \tau_{s.kv}$ so that the Kelvin-Voigt element is considered as a solid. To determine the equilibrium relation of the friction element, first the relaxation behavior is investigated. Following Equation 10b, the stress is not uniquely defined for γ_f = const. If the loading of a friction element is changed from a stress controlled one to relaxation, τ_f equals the stress which was present directly before relaxation was applied so that this stress defines the equilibrium stress $\tau_f{}^{eq} \equiv \tau_f$. An equilibrium relation can be also identified for creep loading with $|\tau_f| < \tau_y$. In case of a creep load with $|\tau_f| = \tau_y$ the friction element flows and no equilibrium relation can be found. But if relaxation is applied, the stress $|\tau_f| = \tau_y$ is identified as equilibrium stress. This discussion explains the statements (S1) and (S2). Because both, an equilibrium relation as well as the flow function are well defined, the friction element is classified as yield stress fluid.

For the Bingham element the investigation of the equilibrium relation is qualitatively the same to that of the friction element. For creep loading with $|\tau_b| \leq \tau_y$ an equilibrium relation is found. The Bingham element flows in case of a creep load with $|\tau_f| > \tau_y$ so that no equilibrium relation can be identified. But if relaxation is applied, the stress in the Newtonian dashpot relaxes

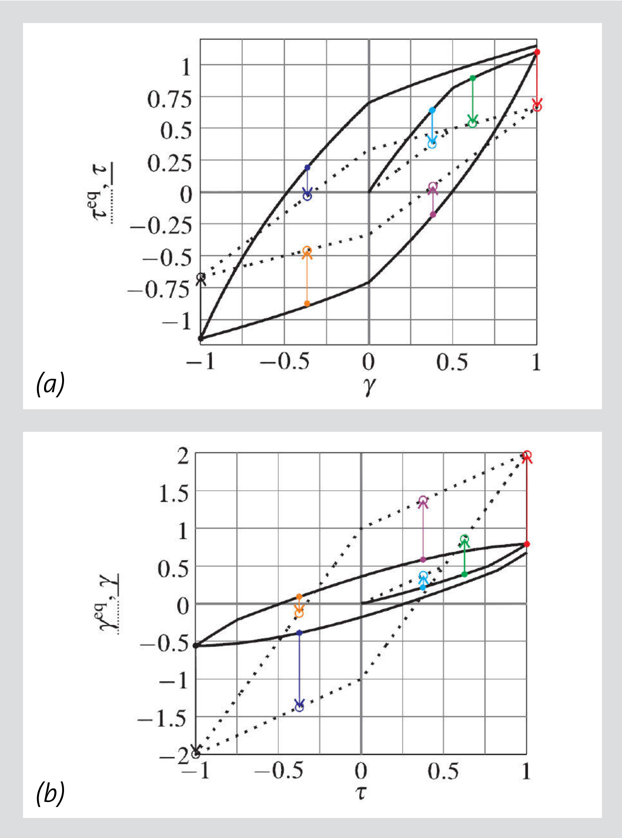

Figure 7: Investigation of the existence of an equilibrium relation for an ideal viscoplastic solid with G = 1 Pa, η = 0.5 Pas, G_1 = 1 Pa, G_2 = 0.5 Pa, τ_y = 0.5 Pa, T = 1 s, $\hat{\gamma}$ = 1 (a) and $\hat{\tau}$ = 1 Pa (b): Total stress $\tau(\gamma)$ and equilibrium stress $\tau^{eq}(\gamma)$ of the ideal viscoplastic solid of Figure 6a according to cyclic strain controlled loading $\gamma(t)$ (colored arrows indicate the relaxation of the overstress), (b) Total strain $\gamma(t)$ and equilibrium strain $\gamma^{eq}(t)$ of the ideal viscoplastic solid of Figure 6a according to cyclic stress controlled loading $\tau(t)$ (colored arrows indicate the redistribution of the total stress). $|\tau^{ov}|$ decreases, $|\tau^{eq}|$ increases as long as $\tau = \tau^{eq}$ and τ^{ov} = 0.

instantaneously and the stress in the friction element remains analog to the discussion above. Thus, the equilibrium is given by $\tau_b{}^{eq} \equiv \tau_{f.b}$. Due to the existing flow function, the new material classification classifies the Bingham element as yield stress fluid as normal.

5.4 APPLICATION OF THE MATERIAL CLASSIFICATION TO EXTENDED RHEOLOGICAL ELEMENTS

In this section, the response of one extended rheological element of each material class is calculated to validate the working principle of the new material classification in much more detail. Therefor, first the flow function is plotted. Second, the determination of the equilibrium relation is discussed due to relaxation as well as creep. The corresponding analytically calculated equations are given in Supplemental Information B.

5.4.1 Classification of a viscoplastic rheological element as solids

To illustrate a classification into the class of solids, an ideal viscoplastic material model with linear kinematic hardening due to the backstress τ_{back} [6, 42, 47, 48] according to Figure 6a is considered. The corresponding one-dimensional constitutive equations are briefly summarised in Supplemental Information B.1. Its non-defined flow function is signed in Figure 6b. The stress response $\tau(\gamma)$ as a result of a cyclic strain controlled loading (Figure 2a with $\beta = \gamma$) as well as the corresponding equilibrium relation $\tau^{eq}(\gamma)$ due to relaxation (Figure 1 with $\beta = \gamma$) are plotted in Figure 7a. The strain response $\gamma(\tau)$ related to a cyclic stress controlled loading (Figure 2a with $\beta = \tau$) as well as the resultant equilibrium relation $\gamma^{eq}(\tau)$ due to creep (Figure 1 with $\beta = \tau$) are given in Figure 7b. The corresponding analytical solutions are documented in Supplemental Information B.2 and B.3. In both cases, either relaxation or creep, an equilibrium relation can be observed which is illustrated by the arrows in Figure 7. Because of an existent flow function and a non-zero equilibrium relation according to relaxation the material behavior of the one-dimensional rheological element in Figure 6a is classified as solid-like.

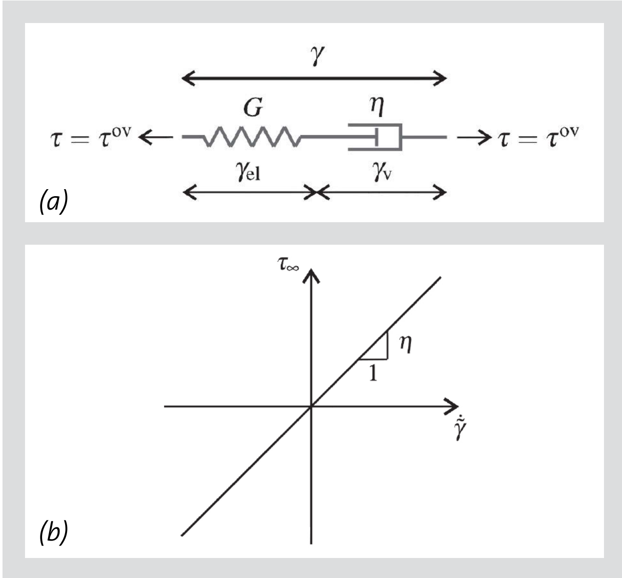

Figure 8: Example of a fluid (a) whose flow function is well defined (b).

5.4.2 Classification of the Maxwell element as fluid

The classification of the Maxwell element (Figure 8a) into the class of fluids is demonstrated in the following. Its well defined flow function is given in Figure 8b. For testing that the flow function is an odd function [49–52]

$$\tau_\infty\left(\dot{\gamma}\right) \stackrel{!}{=} -\tau_\infty\left(-\dot{\gamma}\right) \tag{20}$$

it is plotted for positive and negative strain rates. Figure 9a includes the stress response $\tau(\gamma)$ to a cyclic strain controlled loading related to Figure 2a with $\beta = \gamma$. Furthermore, the corresponding zero equilibrium relation $\tau^{eq}(\gamma)$ = 0 due to relaxation according to Figure 1 with $\beta = \gamma$ is plotted. The strain response $\gamma(\tau)$ to a cyclic stress

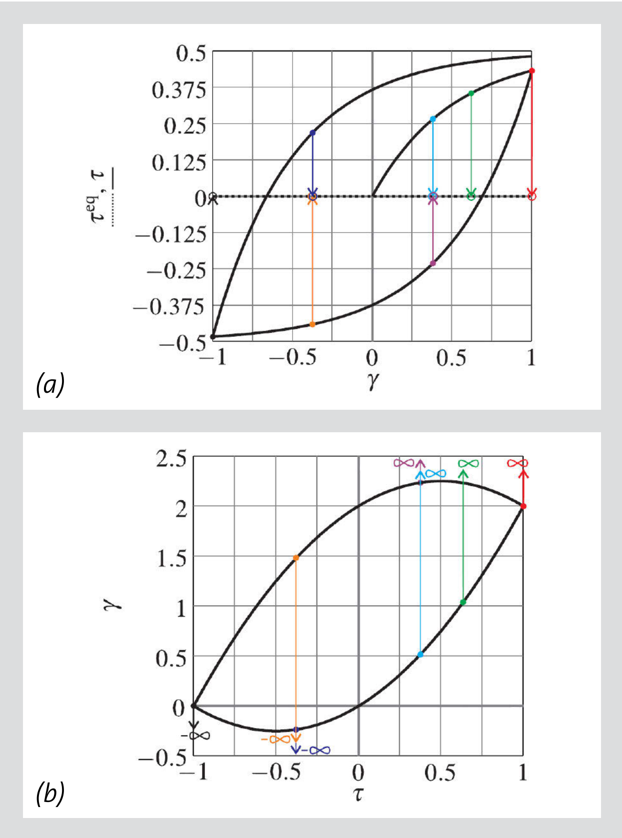

Figure 9: Investigation of the existence of an equilibrium relation for a viscoelastic fluid with G = 1 Pa, η = 0.5 Pas, T = 1 s, $\hat{\gamma}$ = 1 (a) and $\hat{\tau}$ = 1 Pa (b): (a) Total stress $\tau(\gamma)$ and equilibrium stress $\tau^{eq}(\gamma)$ of the viscoelastic fluid of Figure 8a according to cyclic strain controlled loading $\gamma(t)$ (colored arrows indicate the relaxation of the overstress to zero), (b) Total strain $\gamma(t)$ of the viscoelastic fluid of Figure 8b according to cyclic stress controlled loading $\tau(t)$, equilibrium strain $\gamma^{eq}(t)$ is not defined (colored arrows indicate the unlimited increase of γ for infinite holding times because $\tau = \tau^{ov}\ \forall\ \tau$.

controlled loading, defined by Figure 2a with $\beta = \tau$, is given in Figure 9b. Independent of at which point of the strain response $[\gamma(\tau);\tau]$ a creep process is started, the strain never reaches a timeconstant value. In case of a starting point with $\tau < 0$, the strain tends to $\gamma \rightarrow -\infty$, otherwise it goes to $\gamma \rightarrow \infty$. The analytical solution of the total stress response as result of cyclic strain controlled loading is identical with the overstress in Supplemental Information B.2 because the equilibrium stress of a Maxwell element is zero. The analytical solution for the strain response to a cyclic stress controlled loading is determined as solution of the differential equation [5, 6, 36, 43]

$$\eta\dot{\gamma} = \tau + \frac{\eta}{G}\dot{\tau} \tag{21}$$

and is documented in Supplemental Information B.4. Because of an existent flow function and a zero equilibrium relation according to relaxation the material behavior of the one-dimensional rheological element in Figure 8a is classified as fluid-like.

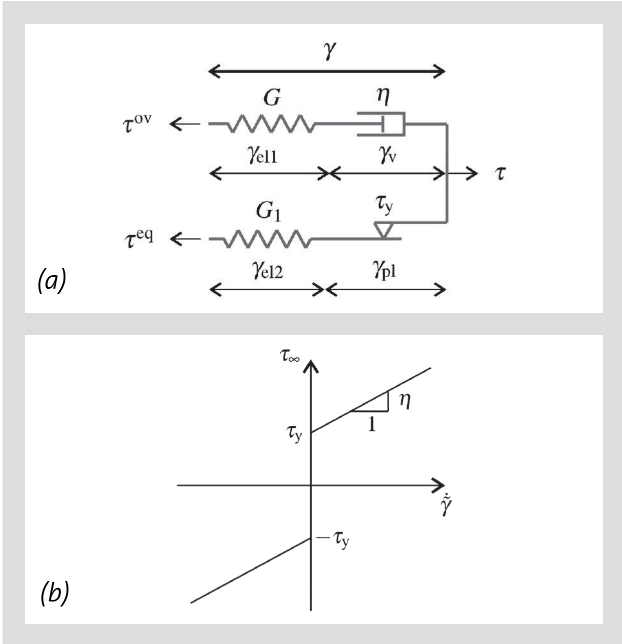

Figure 10: Example of a yield stress fluid (a) whose flow function is well defined (b).

5.4.3 Classification of a viscoplastic rheological element as yield stress fluid

To demonstrate the behavior of yield stress fluids, a viscoplastic material according to Figure 10a is considered. The one-dimensional constitutive equations are briefly summarised in Supplemental Information B.5. Its flow function (Figure 10b) is well defined and equals the one of a Bingham element. The stress response $\tau(\gamma)$ according to cyclic strain controlled loading (Figure 2a with $\beta = \gamma$) as well as the corresponding equilibrium relation $\tau^{eq}(\gamma)$ due to relaxation (Figure 1 with $\beta = \gamma$) are plotted in Figure 11a. The strain response $\gamma(\tau)$ related to a cyclic stress controlled loading (Figure 2a with $\beta = \tau$) is given in Figure 11b. The corresponding equilibrium relation $\gamma^{eq}(\tau)$ due to creep (Figure 1 with $\beta = \tau$) is not unique, because the plastic strain γ_{pl} strongly depends on the history of the entire creep process. If the condition for viscoplastic loading (Equation B.71) is fulfilled, the yield stress fluid of Figure 10a behaves in the postyield (Equation A.11b) and flows so that γ_{pl} changes as long as the condition of viscoelastic unloading (Equation B.69) is met again. That is why the equilibrium strain in Figure 11b can be uniquely determined only in the first preyield (see arrows in Figure) but not for the overall process. If creep is applied in the postyield at a point of the strain response $[\gamma(\tau);\tau]$, the strain tends to infinity for infinite holding times. The analytical solution of the total stress response and the equilibrium stress response as result of cyclic strain controlled loading are identical with the ones of Supplemental Information B.2 with respect to $G_2 = 0$. The analytical solution of the strain response is documented in Supplemental Information B.6. Because of an existent flow function and a non-zero equilibrium relation according to relaxation the one-dimensional rheological element in Figure 8a is a yield stress fluid.

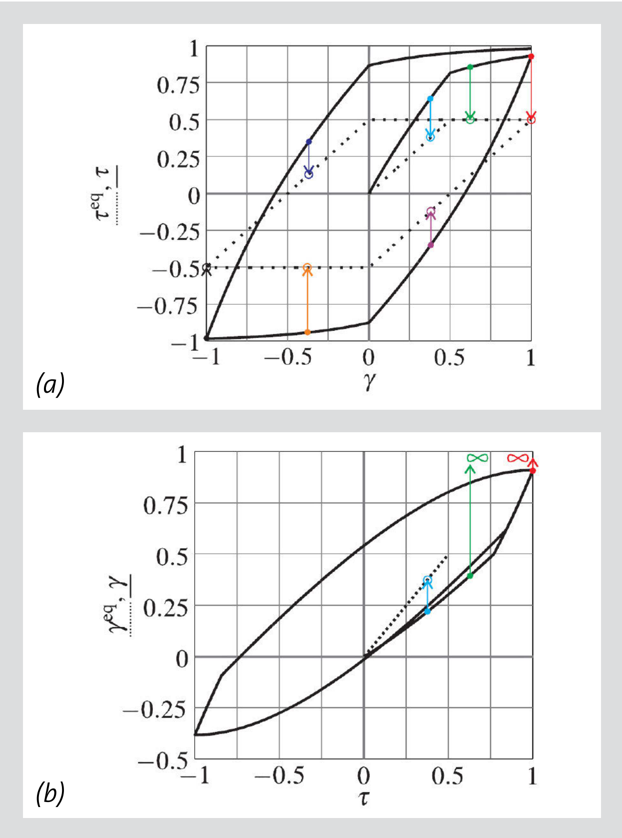

Figure 11: Investigation of the existence of an equilibrium relation for a viscoplastic yield stress fluid with G = 1 Pa, η = 0.5 Pas, G_1 = 1 Pa, ty = 0.5 Pa, T = 1 s, $\hat{\gamma}$ = 1 (a) and $\hat{\tau}$ = 1 Pa (b): (a) Total stress $\tau(\gamma)$ and equilibrium stress $\tau^{eq}(\gamma)$ of the viscoplastic yield stress fluid of Figure 10a according to cyclic strain controlled loading $\gamma(t)$ (colored arrows indicate the relaxation of the overstress, (b) Total strain $\gamma(t)$ of the viscoplastic yield stress fluid of Figure 10a according to cyclic stress controlled loading $\tau(t)$, equilibrium strain $\gamma^{eq}(t)$ is not uniquely defined for the overall process (blue arrow indicates the redistribution of the total stress as long as $\tau = \tau^{eq}$ and τ^{ov} = 0, Green and red arrow indicate the unlimited increase of g for infinite holding times because $|\tau^{ov}| > 0$).

6 SUMMARY AND CONCLUSION

This work is a contribution to aspects of yield stress fluids in the sense of phenomenological modeling. It introduced a terminology to define the three types of materials, ‘solid’, ‘liquid’, and ‘yield stress fluid’, and the terms ‘preyield’ and ‘postyield’. In this context a procedure was presented so that one can determine the class of a material behavior. Furthermore, the use of the differential viscosity instead of the dynamic viscosity was highlighted. The theory was proved by standard as well as extended rheological elements in Section 5. Here, a great emphasise was placed on the friction element since it is the basis of the yield stress concept.

The physical interpretation of viscosity was theoretically discussed in Section 3 for non-Newtonian fluids and was investigated by standard rheological elements in Section 5.2. Their constitutive equations were evaluated for constant positive and negative strain rates. The question: ”Does it makes sense, that the single friction element is connected with viscous properties?” directly led to the deficit of the dynamic viscosity in case of yield stress fluids. Because the dynamic viscosity is just the secant between a point on the flow function and the origin, it has no physical meaning for strongly non-linear fluids. For non-Newtonian fluids the application of the differential viscosity is advisable since it is defined as the tangent to the flow function.

To ensure a coherent concept of material classification, Section 4 was devoted to the general questions: ”What is a convenient definition of the term yield stress fluid in the sense of phenomenological modeling? How can it be distinguished from solids and liquids?” Here, three types of material behavior were classified by the equilibrium relation, introduced in Section 2, and the flow function (Section 3). The measuring of the flow function is clear, of course. But how is it possible to measure the equilibrium relation of yield stress fluids? In this sense, the determination of the equilibrium relation was considered related to relaxation and creep. The uniqueness between relaxation and creep is not given for all theoretically possible materials. Examples were given by the friction and Bingham element in Section 5.3, the Maxwell element (Section 5.4.2) as well as the viscoplastic yield stress fluid in Section 5.4.3. In case of fluids, one cannot determine the zero equilibrium relation by creep, because the material flows indefinitely. But it is possible to identify the zero equilibrium relation by relaxation. The determination of the equilibrium relation by creep and relaxation only gives the same result for solids. For yield stress fluids the identification of the equilibrium relation according to relaxation and creep is only possible in the preyield. In the postyield regime yield stress fluids flow indefinitely for infinite holding times so that no equilibrium relation can be identified by creep. It can be determined only through relaxation. The corresponding measurement procedure was given in Figure 1 with $\beta = \gamma$. By the application of the new material classification to standard and extended rheological elements in Sections 5.3 and 5.4, the working principle was verified because it produced the expected statements of literature for classical solids and liquids and dispeled ambiguity for yield stress fluids like it is denoted by Reiner [53].

As it is mentioned above, the investigation of the friction element was inevitably necessary for this work. Therefore, the first part of the constitutive equations of the friction element are worked out in Section 5.1, being valid for the preyield as well as the postyield with respect to positive and negative strain rates. This could be ensured by the introduction of a signum related function.

The second part of the constitutive equations of the friction element was presented in Supplemental

Information A and included the description in terms of the classical theory of plasticity. As consequence, the terms 'preyield' and 'postyield' were defined for the friction element by the loading and unloading conditions which in turn are connected with the Karush-Kuhn-Tucker and consistency conditions. In this work the latter two were illustrated in a convenient way by the evaluation of logical expressions in Supplemental Informationg A.1 and A.2 without considering the optimisation problem with constraints in form of inequalities.

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