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Free Energy and States of Fractional-Order Hereditariness

Luca Deseri[(1,2,3)], Mario Di Paola[(4)], Massimiliano Zingales*,[(4,5)]

[(1)] *Dipartimento di Ingegneria Civile, Ambientale e Meccanica, Via Mesiano n.77, 38123, Trento, Italy.*
[(2)] *Dept. of Civil Environmental Engineeering, Dept. of Mechanical Engineering, Carneige-Mellon University, 5000 Forbes Avenue, Pittsburgh PA, 15213-3890, USA.*
[(3)] *TMHRI-Department of Nanomedicine, The Methodist Hospital Research Institute, 6565 Fannin St. MS B-490, Houston TX 77030 USA.*
[(4)] *Dipartimento di Ingegneria Civile, Ambientale, Aerospaziale, dei Materiali, Viale delle Scienze Ed.8, 90128, Palermo, email:massimiliano.zingales@unipa.it*
[(5)] *Laboratory of Bio-NanoMechanics in Medicine (BNM2-LAB), Mediterranean Center of Human Health and Advanced Biotechnologies, Viale delle Scienze Ed.18, 90128, Palermo*

Abstract

Complex materials, often encountered in recent engineering and material sciences applications, show no complete separations between solid and fluid phases. This aspect is reflected in the continuous relaxation time spectra recorded in cyclic load tests. As a consequence the material free energy can not be defined in a unique manner yielding a significative lack of knowledge of the maximum recoverable work that can extracted from the material. The non-uniqueness of the free energy function is removed in the paper for power-laws relaxation/creep function by using a recently proposed mechanical analogue to fractional-order hereditariness.

Keywords: Fractional derivatives, Power-law creep/relaxation, Free energy, Dissipation rate, Material state.

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1. Introduction

Recent applications in engineering and physical sciences have involved reticulated polymers, foams, hydrogels soft matter as well as biological tissues as shown in several papers ((1),(2),(3),(4)). These materials show significative deviations of their mechanical and rheological features from well-studied

[1]corresponding author email:massimiliano.zingales@unipa.it; tel-fax:+39-091-238-967-63

conventional materials.

The differences are mainly due to the presence of a marked microstructure with material cross-links, Wan der Waals interactions and/or dipole-dipole bonds that lead to unpredictable behavior with the well-known tools of continuum mechanics, material rheology and thermodynamics . Indeed, in the rheology framework studied in the paper, complex materials involve different states of the matter that can not be separated at the macroscopic observation scale. Such a consideration suggests that a rheological description in terms of mechanical arrangements of linear springs and linear dashpots, representing the solid and the fluid phases, respectively, is not acceptable. This is confirmed by experimental relaxation spectra (see e.g. paper (5)) that shows a continuous distribution of relaxation times. Continuous relaxation times spectra are not described by single or multiple exponential-type relaxation functions, corresponding to arrangements of springs and dashpots, and they may be described, instead, by power-law relaxation functions.

As a matter of fact creep and relaxation tests show that experimental data may be fit with extraordinary precisions by power-laws with real-order exponents $0 \leq \beta \leq 1$ ((6),(7)) yielding, by the Boltzmann superposition integral, constitutive equations in terms of fractional operators ((8),(9)(10),(11)). Fractional differential calculus may be thought as a generalization of the well-known, integer-order, calculus operators that turns the order $j \in N$ of differentiation of a function $f(x)$ into a real-order value $j \leq \beta \leq j+1$ so that $d^j f(x) \rightarrow d^\beta f(x)$. Incorporating such fractional-order operators into the governing constitutive equations produces new viscoelastic models for Fractional Hereditary Materials (FHM) that exhibit power-law relaxations. Mechanical and geometrical representations of the fractional differentiation have been reported in several scientific fields, such as, mechanics ((12),(13)) , thermodynamics ((14),(15),(16),(17),(18)) beside rheology ((19),(20)).

The main gap in the use of alternative relaxation functions with respect to the widely used linear combination of exponential relaxations is related to the lack of an unique definition of the free energy function. This is the consequence to the incomplete separation of the materials' elastic and viscous phases at the scale of the tensile tests, yielding non-conventional creep and relaxation functions. Indeed if the two phases of the material may be clearly identified, as in conventional applications, then an exponential-type relaxation function is observed in creep/relaxation tests and

no differences among the existent versions of the free energies exist.

In this regard, free energies, defined upon phenomenological relaxation/creep functions must satisfy the fundamental properties described in classical papers by ((21),(22),(23)) and, as those properties are satisfied, several well-known different expressions for free energies may be found in scientific literature ((24),(25),(26),(27),(28)). Despite their equivalence in terms of the correspondent stress, very different configurations, each corresponding to the same material state, may have the same value of the free energy. Indeed the mechanical tests usually introduced to define the relaxation/creep function of the material do not allow for the evaluation of the material stored energy nor of the dissipated energy. This is a severe drawback since the maximum recoverable work from the material can not be evaluated using relaxation functions other than the exponential-type. The use of power-law relaxation functions $\propto t^{-\beta}$ with $0 \leq \beta \leq 1$ undergoes the same limitation, restricting the use of power-laws for the representation of creep and relaxations of unconventional materials.

In a general context, the aforementioned considerations may be overcome, allowing for only one definition of the material free energy, as the elastic (solid) and viscous (fluid) phases of the material may be separated. In this regard it has been recently shown that power-law creep/relaxations may be obtained with a mechanical analogue to the fractional-order springpot with a complete separation of solid and fluid phases ((29),(30),(31)). This model served, also, as a guide to introduce a fractal material microstructure yielding a macroscopic power-law relaxation with the order of the power-law corresponding to the Hausdorff dimension of the microstructure ((32)).

In this paper the model is further investigated to provide an unique expression of the free energy functional of FHM. It is shown that the evaluation of the dissipation rate of FHM, obtained with the mechanical analogue to the power-law relaxation, coincides after some algebraic manipulations, with the well-known dissipation rate of the Stavermann-Schwarzl formulation.

Additionally, the complete characterization of parameters of FHM, requires specific care in the definition of the equivalence classes of the stress (or strain) histories undergone by the material, namely to the knowledge of the *state* of the material ((33)). Indeed, it has been shown recently that the correct evaluation of FHM parameters requires the knowledge of the entire strain histories

undergone by the material specimen ((34)), and then, the correct definition of the material state is crucial for the general derivation of the free energy functional ((35),(36)). This latter problem is not so evident for relaxation functions given as linear combination of exponential laws since a discrete number of state variables are involved for the definition of the material state. The expression of the free energy in terms of the material state for the power-laws relaxation functions has also been provided in the paper.

The paper is organized as follows: In the next section the authors will provide some basic definitions involving power-laws creep/relaxations, the corresponding definitions of fractional integrals and derivatives and the definition of material free energy. In sec.(3) the rheological description of power-laws creep/relaxations will be briefly outlined for EV and VE material models. Sec.(4) will be devoted to the evaluation of the elastic energy stored in the mechanical model showing that it coalesces with a specific form of the free energy based upon the phenomenological power-law relaxation function. The expression for the material free energy in terms of the state of FHM is reported in sec.(5) with some comments reported in sec.(6). Mathematical details about fractional-order calculus have been introduced in appendix A and some additional informations concerning the mathematical derivations reported in the paper have been discussed in appendix B.

2. Power-law relaxation (creep) function: The free energy function of FHM

In the subsequent derivations we recall that two hypotheses are considered while analyzing viscoelastic materials (see e.g. (10)): *i*) invariance under time translation and *ii*) causality. With the first requirement we mean that a time shift in the input is reflected as the same shift in the output; with the second we mean that the material response depends on previous histories only, reflecting the memory of such materials. The discussion about the power-law relaxation (creep) function in integral material hereditariness is reported in this section with regards to the issues:

1. *Power-law relaxation (creep) functions* yielding the constitutive relations among the stress measure and the strain measure in terms of fractional-order operators.
2. *The free energy function of FHM* that represent a scalar measure of the stored material energy.

The section is devoted to the introduction of power-law relaxation/creep functions and their relations with fractional-order derivatives and integrals (sec.2.1). The definition of the material free energy and its relation with the stress measure are discussed in sec.(2.2)

2.1. Power-law relaxation (creep) function : Fractional-order calculus

Let us assume, in this section, to deal with *virgin* materials, namely either the strain or the stress are known from the very beginning of the observation of their behavior, conventionally set at $t = 0$, and hence no past histories with respect to such a time need to be taken into account.

Creep and relaxation tests are performed to detect the memory of the material: in the first case, the stress is held constant and the strain is measured, whereas in the second one the strain is held constant and the stress is measured. Whenever either a unit stress or a unit strain is utilized, the creep compliance $J(t)$ and relaxation modulus $G(t)$ are found as the strain and stress response to the imposed unit stress and strain respectively, i.e.

$$\epsilon(t) = U(t) \longrightarrow \sigma(t) = G(t) \tag{1a}$$

$$\sigma(t) = U(t) \longrightarrow \epsilon(t) = J(t), \tag{1b}$$

where $U(\circ)$ is the unit Heaviside step function. When either the creep or the relaxation function is known, the Boltzmann superposition principle allows writing convolution-type Riemann-Stieltjies integrals to express the relationships between σ and ϵ. Whenever either the strain or the stress are prescribed, the constitutive relations for the corresponding derived quantities read as follows:

$$\sigma(t) = \int_{0^+}^{t} G(t-\tau)d\epsilon(\tau), \tag{2a}$$

$$\epsilon(t) = \int_{0^+}^{t} J(t-\tau)d\sigma(\tau). \tag{2b}$$

Smoothness assumptions on $\epsilon(t)$ and $\sigma(t)$ will be discussed in the sequel.

Creep compliance and relaxation modulus are not independent. Indeed they are linked to each other by the relationship

$$\hat{J}_+(\omega)\hat{G}_+(\omega) = \frac{1}{(i\omega)^2}, \tag{3}$$

where the symbol $\hat{}$ denotes the right-sided Fourier transform (see eq.6, equation eq.(70).

Experiments on polymeric materials performed (see paper by Nutting, (1)) at beginning of the twentieth century showed that their relaxation function was well fitted by power-laws, i.e.

$$G(t) = \frac{C_\beta}{\Gamma(1-\beta)} t^{-\beta}, \tag{4}$$

where $\Gamma(\circ)$ is the Euler-Gamma function, C_β and β are characteristic constants of the material. The exponent β must be enclosed in the range $0 < \beta < 1$ because of thermodynamics restrictions ((37),(30)). At the extrema of the range, asymptotic behaviors are obtained: $\beta \to 0$ corresponds to purely elastic solid whereas $\beta \to 1$ to purely viscous fluid. The values of $0 < \beta < 1$ correspond to an intermediate behavior between elastic solid and viscous fluid, allowing it to describe both complex-structured materials and soft matter. As we expect, the creep compliance of the given material can be determined through (3) from the relaxation modulus assumed in (4). Furthermore, the right Fourier transform of the relaxation function (4) yields

$$\hat{G}_+(\omega) = C_\beta (i\omega)^{\beta-1}, \tag{5}$$

which may be replaced in (3) yielding, after inverse transform:

$$J(t) = \frac{1}{C_\beta \Gamma(1+\beta)} t^{\beta}. \tag{6}$$

It is worth analyzing the material behavior with the aid of normalized functions, $\overline{G}(t)$ and $\overline{J}(t)$ defined as follows $\overline{G}(t) := G(t)\left(C_\beta\right)^{-1}\Gamma(1-\beta) = t^{-\beta}$ and $\overline{J}(t) := J(t)C_\beta\Gamma(1+\beta) = t^{\beta}$ and plotted in fig.(1). A careful observer will notice immediately that all the curves share the common point (1,1), which represents a key value. Indeed, the blue curves ($0 < \beta \leq 1/2$) show that the elastic phase prevails on viscous one with decreasing β, whereas the red ones ($1/2 \leq \beta < 1$) show that the viscous phase prevails on elastic one as increases as β. This consideration provides a mean for identifying the former as *ElastoViscous* (EV) materials while the latter as *ViscoElastic* (VE) ones; the value $\beta = 1/2$ is clearly common to both kinds of materials, thus it may be obtained as a limiting case of both models described above. The constitutive equations of fractional hereditary materials may be obtained with the substitution of (4) and (6) into Boltzmann integrals in (2) and

in (2b) yielding:

$$\sigma(t) = \int_{0^+}^{\epsilon(t)} G(t-\tau)d\epsilon(\tau) = \frac{C_\beta}{\Gamma(1-\beta)} \int_{0^+}^{\sigma(t)} \frac{\dot{\epsilon}(\tau)}{(t-\tau)^\beta} d\tau = C_\beta \left({}^C\mathrm{D}_{0^+}^\beta \epsilon\right)(t)\,, \tag{7a}$$

$$\epsilon(t) = \int_{0^+}^{t} J(t-\tau)d\sigma(\tau) = \frac{(C_\beta)^{-1}}{\Gamma(1+\beta)} \int_{0^+}^{t} (t-\tau)^\beta \dot{\sigma}(\tau)d\tau = \frac{1}{C_\beta} \left(\mathrm{I}_{0^+}^\beta \sigma\right)(t)\,, \tag{7b}$$

which represent the constitutive relations in terms of fractional-order integrals and derivatives defined in the positive real axis $[0, \infty[$. In eq.(7) the terms $\left({}^C\mathrm{D}_{0^+}^\beta \epsilon\right)(t)$ and $\left(\mathrm{I}_{0^+}^\beta \sigma\right)(t)$ so-called Caputo's fractional derivative and Riemann-Liuville fractional integrals, respectively (see 6). We conclude that as soon as the creep and/or the relaxation function is assumed to be a power-law type, then the constitutive laws involves fractional operators. The constitutive equations in terms of fractional-order calculus assume that the considered material is at rest for $t = 0$ a condition that is seldom available in engineering and physical applications. It has been shown in previous paper that the presence of an history of strain/stress undergone by the material up to the time instant $\bar{t}$, that is the initial time of the strain/stress process must be included in the constitutive equations to provide accurate estimations of the decaying and the force coefficients of the material, namely β and C_β ((38)). In this latter case the complete stress measure in virgin FHM may be obtained by extending the initial time instant to $-\infty$ yielding:

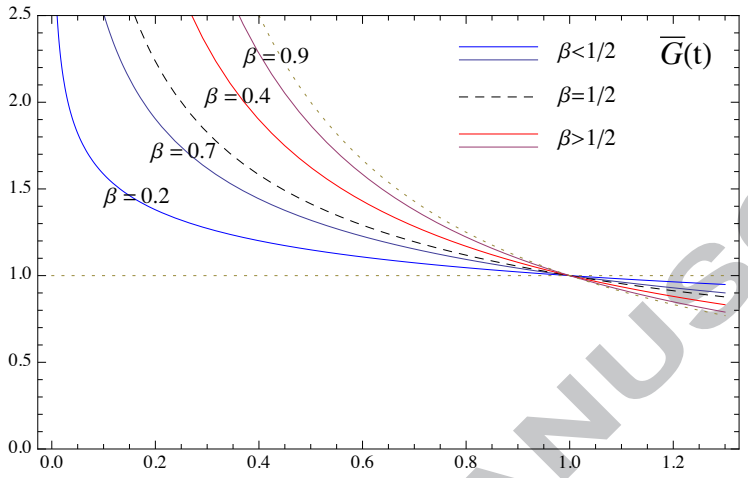
$$\sigma(t) = \int_{-\infty}^{t} G(t-\tau)d\epsilon(\tau) = \frac{C_\beta}{\Gamma(1-\beta)} \int_{-\infty}^{t} \frac{\dot{\epsilon}(\tau)}{(t-\tau)^\beta} d\tau = C_\beta \left({}^C\mathrm{D}_{+}^\beta \epsilon\right)(t)\,, \tag{8a}$$

$$\epsilon(t) = \int_{-\infty}^{t} J(t-\tau)d\sigma(\tau) = \frac{(C_\beta)^{-1}}{\Gamma(1+\beta)} \int_{-\infty}^{t} (t-\tau)^\beta \dot{\sigma}(\tau)d\tau = \frac{1}{C_\beta} \left(\mathrm{I}_{+}^\beta \sigma\right)(t)\,, \tag{8b}$$

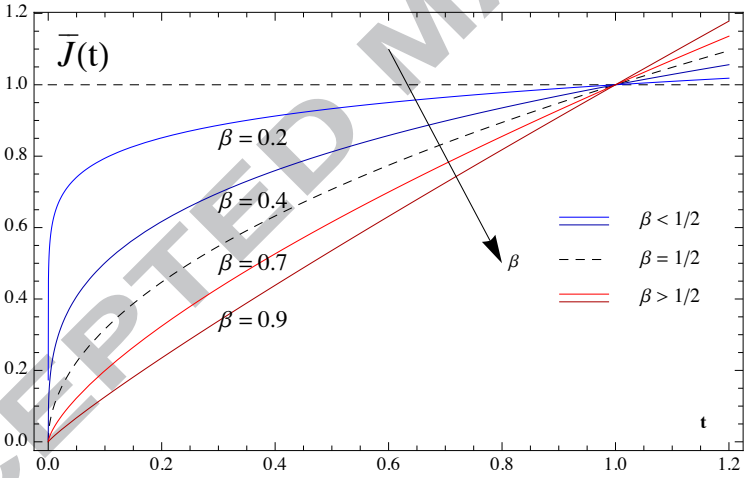
In the following section the definition of free energy of fractional hereditary materials will be discussed. The fundamental definitions necessary to evaluate the free energy associated to the power-law creep/relaxation from a mechanical perspective will be shortly summarized.

2.2. The free energy functionals of FHM

The specific Helmoltz free energy $\psi(\epsilon, \epsilon^t, T)$ with T the absolute temperature, $\epsilon^t(\tau) = \epsilon(t-\tau)$ with $-\infty \leq \tau \leq t$ the strain history, is, by definition, a thermodynamic state function whose gradient, with respect to the actual value of the deformation ϵ yields the measured stress. The rate of increments of the material free energy is expressed in terms of the rate of increments of the



(a) $\overline{G}(t)$



(b) $\overline{J}(t)$

Figure 1: Normalized (a) relaxation and (b) creep functions.

specific internal energy $\dot{u}$ and of the entropy production as:

$$\dot{\psi} := \dot{u} - T\dot{s} \tag{9}$$

where $\dot{s}$ is the specific entropy production rate that must satisfy the thermodynamic restriction provided by the second principle of thermodynamics $\dot{s} \geq \dot{q}/T$, where $\dot{q}$ is the rate of change of the specific thermal energy. The rate of change of the internal energy function is related to the specific mechanical work done on the system and to the thermal energy exchange as (first principle of thermodynamics) $\dot{u} = \dot{w}_{ext} + \dot{q}$. The free energy function in eq.(9) may be expressed in a more explicit form if we introduce the entropy production rate due to irreversible transformations, namely $\dot{s}^{(i)} \geq 0$ so that the second principle of thermodynamics may be written as: $\dot{s} = \dot{q}/T + \dot{s}^{i}$ yielding, upon substitution in the expression of the free energy in eq.(9)

$$\dot{\psi} = \dot{w}_{ext} - T\dot{s}^{(i)} = \dot{w}_{ext}(t) - D(t) \tag{10}$$

where we used the balance equation of the first principle and we introduced the specific dissipation rate $D(t) = T\dot{s}^{(i)} \geq 0$ due to irreversible thermodynamical transformations in the material.

Assuming isothermal processes, the free energy function ψ for simple hereditary material depends on the current value of the strain and on the past history undergone by the material $\psi(\epsilon\,(t)\,, \epsilon^t(\tau))$. The free energy function must be a state function and it must fulfill the following thermodynamical restrictions thermodynamic restrictions as (see paper (33)):

- For any time instant the following thermodynamic restriction holds true:

$$\sigma(t)\dot{\epsilon}(t) \geq \dot{\psi}(\epsilon(t), \epsilon^t(\tau)) \tag{11}$$

- For the entire set of strain histories $\epsilon(t-\tau)$ sharing the same value at the time instant t as $\epsilon(0) = \epsilon_0$ the minimum free energy must be achieved for the uniform strain history as $\epsilon(t-\tau) = const = \epsilon_0 \quad \forall t$

- The derivative of the free energy with respect to the actual value of the strain must correspond

to the measured stress as

$$\sigma(t) = \frac{\partial \psi(\epsilon(t), \epsilon^t(\tau))}{\partial \epsilon(t)} \tag{12}$$

- For any strain ϵ_0

$$\psi(\epsilon_0, \epsilon_0^t) - \psi(0, 0^t) = 1/2 G_\infty \epsilon_0^2 \tag{13}$$

where ϵ_0^t denotes a constant history of value ϵ_0.

- Any free energy is a state function and, therefore the following equality must be fulfilled for any pair of strain histories $\epsilon_A^t(\tau)$ and $\epsilon_B^t(\tau)$ corresponding to the same material state as:

$$\psi(\epsilon(t), \epsilon_A^t(\tau)) = \psi(\epsilon(t), \epsilon_B^t(\tau)) \tag{14}$$

Under the assumption of linear hereditary behavior the stress value $\sigma(t)$ is provided by the Boltzmann-Volterra integral as reported in the first equality of eq.(8). An explicit dependence of the stress measure on the actual value of the strain and of the strain histories may be easily introduced as we perform an integration by parts yielding the the following expression:

$$\sigma(\epsilon, \epsilon^t) = G(0)\epsilon(t) + \int_0^\infty \dot{G}(\tau)\epsilon^t(\tau) d\tau \tag{15}$$

The latter may be substituted in eq.(13) to yield a general expression of the free energy functional explicitly dependent on the current value of the strain as well as on the strain histories as follows:

$$\psi(\epsilon(t), \epsilon^t(\tau)) = \frac{1}{2} G(0)\epsilon(t)^2 + \epsilon(t) \int_0^\infty \dot{G}(\tau)\epsilon^t(\tau) d\tau + R(\epsilon^t(\tau)) \tag{16}$$

where $R(\epsilon^t(\tau))$ must be a functional of the past strain history only. The presence of the functional $R(\epsilon^t(\tau))$ is a source of indeterminacy for the free energy function; This leaves room to several choices for $\psi(t)$ the expressions proposed in papers ((24),(25),(26),(27),(28)), among others.

In other words the measure of the relaxation/creep function of the material is not sufficient, by itself, to predict the amount of energy available during any strain/stress process. If, instead, a rheological description of relaxation/creep function of a material may be identified accounting for viscous and elastic phases, then the dissipation rate (or the free energy rate) may be uniquely

evaluated.

Indeed, it is well-known that, for simple rheological models such as the Kelvin, the Maxwell as well as for their combinations, the expression of the specific free energy of the system may be obtained evaluating the specific energy stored in the elastic springs (see e.g. (24),(25)). Alternatively, the free energy function may be evaluated in terms of the specific dissipation rate of the viscous dashpots in the rheological model. The evaluation of the free energy function, in these cases, may be directly obtained by the physics of the model separating the fluid (purely viscous) and the solid (purely elastic) phases in the rheological model of the material relaxation function. It may be argued that, as an exact rheological representation of the relaxation function is involved, then the free energy of the material may be obtained in a mechanical setting evaluating either the elastic energy stored or the viscous dissipation rate. The choice of a specific form for the free energy function, although corresponding to the same representation for the stress function, may correspond to very different material state significantly affecting the mathematical predictions.

In the sequel of the paper we refer to a specific functional class of the free energy reported in eq.(16) that, assuming a linear material behavior, may be expressed as a quadratic functional of the strain measures for the free energy $\psi(t)$ (see ((25))) as :

$$\psi(t) = \int_{-\infty}^{t}\int_{-\infty}^{t} K(t-\tau_1, t-\tau_2)\dot{\epsilon}(\tau_1)\dot{\epsilon}(\tau_2)d\tau_1 d\tau_2 \tag{17}$$

with $K(\circ,\circ) \geq 0$ any non-negative and symmetric, i.e., $K(\tau_1,\tau_2) = K(\tau_2,\tau_1)$ continuous and differentiable function with respect the two arguments τ_1 and τ_2. In this setting, the explicit dependence of the energy from the current value of the strain and of the strain histories may be performing some straightforward integration by parts and variable substitutions in eq.(17) yielding:

$$\psi(\epsilon,\epsilon^t) = K(0,0)\epsilon(t)^2 + 2\epsilon(t)\int_{-\infty}^{t}\dot{K}(0,t-\tau)\epsilon(\tau)d\tau + \int_{-\infty}^{t}\int_{-\infty}^{t}\ddot{K}(t-\tau_1,t-\tau_2)\epsilon(\tau_1)\epsilon(\tau_2)d\tau_1 d\tau_2, \tag{18}$$

where we used the symmetry of the $K(\circ,\circ) \geq 0$. Inspection of the free energy functionals in eq.(16) and in eq.(18) shows that, under the assumption of $\psi(t)$ in eq.(17) the kernel function $K(\circ,\circ)$ may be related to the relaxation function obtained by stress measures as $2K(0,0) = G(0)$ and

$K(0,t) = K(t,0) = G(t)$. This identification yields the following expression for the integration term $R(\epsilon^t(\tau))$:

$$R(\epsilon^t(\tau)) = \int_0^\infty \int_0^\infty \ddot{K}(\tau_1,\tau_2)\epsilon^t(\tau_1)\epsilon^t(\tau_2)d\tau_1 d\tau_2 \tag{19}$$

This expression shows that the integration term is explicitely dependent on the choice of the Kernel function $K(\circ,\circ)$ whose restriction to the time axis τ_1 and τ_2 coincides with the relaxation function $G(t)$.

A more specific expression of the free energy functional is obtained as the kernel function is related to the relaxation function as ((24)):

$$K(\tau_1,\tau_2) = \frac{G(t-\tau_1+t-\tau_2)}{2} = \frac{G(2t-\tau_1-\tau_2)}{2} \tag{20}$$

then the free energy expression in eq.(17) is obtained in the form:

$$\psi_{ss}(t) = \frac{1}{2}\int_{-\infty}^{t}\int_{-\infty}^{t} G(2t-\tau_1-\tau_2)\dot{\varepsilon}(\tau_1)\,\dot{\varepsilon}(\tau_2)\,d\tau_1 d\tau_2 \tag{21}$$

where the suffix SS stands for Stavermann and Schwartzl free energy. In eq.(21) we made the replacement of the strain variable in terms of shear strain to be consistent with subsequent derivations. The introduction of the first principle of thermodynamics, yields that the specific mechanical work must equate the rate of increment of specific free energy added to the dissipation rate of the material as:

$$\begin{aligned}\sigma(t)\,\dot{\varepsilon}(t) &= 2\dot{\varepsilon}(t)\int_{-\infty}^{t} K(0,t-\tau)\dot{\varepsilon}(\tau)\,d\tau + \int_{-\infty}^{t}\int_{-\infty}^{t}\dot{K}(t-\tau_1,t-\tau_2)\dot{\varepsilon}(\tau_1)\,\dot{\varepsilon}(\tau_2)\,d\tau_1 d\tau_2 + D(t)\\ &= \dot{\varepsilon}(t)\int_{-\infty}^{t} G(t-\tau)\dot{\varepsilon}(\tau)\,d\tau + \frac{1}{2}\int_{-\infty}^{t}\int_{-\infty}^{t}\dot{G}(2t-\tau_1-\tau_2)\dot{\varepsilon}(\tau_1)\,\dot{\varepsilon}(\tau_2)\,d\tau_1 d\tau_2 + D(t)\end{aligned} \tag{22}$$

where we neglected non-essential dependency on the actual value of the strain only $\epsilon(t)$. The observation of the energy rate reported in eq.(22) shows that the dissipation rate associated to the

free energy in eq.(21) reads:

$$D\left(t\right)=-\frac{1}{2}\int_{-\infty}^{t}\int_{-\infty}^{t}\dot{G}\left(2t-\tau_1-\tau_2\right)\dot{\varepsilon}\left(\tau_1\right)\dot{\varepsilon}\left(\tau_2\right)d\tau_1 d\tau_2 \tag{23}$$

, that is, a quadratic form of the strain histories with measure provided by the relaxation function $G(t)$. Different choices of the kernel $K(\tau_1,\tau_2)$ in eq.(17) yield different choices for the dissipation rates, each corresponding to the same stress at time t. The explicit expressions of the free energy $\psi(t)$ and of the dissipation rate of $D(t)$, in eqs.(21, 23) for the power-law relaxation function is obtained as:

$$\psi_{ss}\left(t\right)=\frac{1}{2C_\beta\Gamma(\beta)}\int_{-\infty}^{t}\int_{-\infty}^{t}\left(2t-\tau_1-\tau_2\right)^{-\beta}\dot{\varepsilon}\left(\tau_1\right)\dot{\varepsilon}\left(\tau_2\right)d\tau_1 d\tau_2 \tag{24a}$$

$$D\left(t\right)=\frac{\beta}{2C_\beta\Gamma(\beta)}\int_{-\infty}^{t}\int_{-\infty}^{t}\left(2t-\tau_1-\tau_2\right)^{-(1+\beta)}\dot{\varepsilon}\left(\tau_1\right)\dot{\varepsilon}\left(\tau_2\right)d\tau_1 d\tau_2 \tag{24b}$$

Summing up, in this section we observed that the definition of the free energy in the presence of hereditary material behavior is affected by a pathological drawback: The presence of an integration term that is not known by relaxation (or creep) measures. In this regard, the residual term is related to the dissipation rate of the material during the strain history and, assuming a specific expression of the free energy functional it may be evaluated. Different kinds of expressions of the dissipation rate may be obtained for the different expressions of the free energy functional each corresponding to the same measures of stress relaxation, thus providing non-unique relations among stress measures and the material dissipation. In passing we observe that some thermodynamical restrictions about the fractional model of creep/relaxations have been reported in a recent paper (39).

In the following sections we show that for the power-law creep/relaxation function, a proper mechanical model that separates the elastic (solid) phase and the viscous (fluid) phase may be used to introduce a unique expression of the free energy for FHM. Thus, the exact amount of energy stored in the material for any strain process can be evaluated.

3. Rheological models of Fractional Hereditary Materials

The use of a mechanical model to pursue power-law hereditariness of material response is an old topic of rheology and it has been treated, in exact form, only for the case of power-law decay $\beta = 1/2$ ((19),(40)). Several other approximate expressions have been obtained in the scientific literature for $0 \leq \beta \leq 1$ holding for specific time intervals ((20),(41)).

In recent studies, the authors proposed an exact mechanical description of power-laws for decay exponent in the entire range $0 \leq \beta \leq 1$ possessing a clear separation between the viscous and the elastic material phases (30). The behavior of elasto-viscous material has been separated from visco-elastic one: Both are ruled by β-order differential equation, but in the former case $0 < \beta < 1/2$ while in the latter $1/2 \leq \beta < 1$. The different range of fractional-order involved in constitutive equations is linked to a different mechanical model. In this section we show two different mechanical arrangements to describe the material behavior. The rheological scheme of *Elasto-Viscous* material

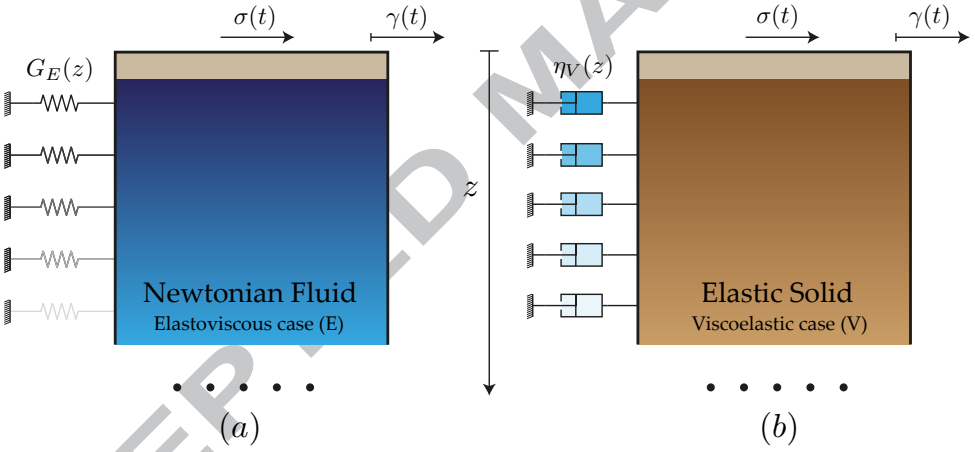


Figure 2: Rheological continuum models: (a) ElastoViscous (EV) and (b) ViscoElastic (VE).

(EV) is represented by an indefinite massless plate resting on a column of Newtonian fluid supported on a side by a "bed" by way of independent elastic springs, whereas the model of *Visco-Elastic* material (VE) is represented by an indefinite massless plate resting on a column of elastic solid linked to a rigid support through independent viscous dashpots, as depicted in fig.(2). In both cases, we consider a cross-section with area A; moreover, we assume that the material elastic modulus

$k(z)$ and viscous coefficient $c(z)$ spatially decay with a power-law, resembling a functionally graded microstructure. Thus, in the case of EV material, they read as follows:

$$k_{EV}(z) = AG_{EV}(z) = A\frac{G_0}{\Gamma(1+\alpha)}z^{-\alpha} \tag{25a}$$

$$c_{EV}(z) = A\eta_{EV}(z) = A\frac{\eta_0}{\Gamma(1-\alpha)}z^{-\alpha} \quad , \tag{25b}$$

whereas in the case of VE material they become:

$$k_{VE}(z) = AG_{VE}(z) = A\frac{G_0}{\Gamma(1-\alpha)}z^{-\alpha} \tag{26a}$$

$$c_{VE}(z) = A\eta_{VE}(z) = A\frac{\eta_0}{\Gamma(1+\alpha)}z^{-\alpha}. \tag{26b}$$

In equations eq.(25), eq.(26) the subscripts EV and VE indicate ElastoViscous and ViscoElastic case, respectively, and $0 \leq \alpha \leq 1$ is the decay parameter. In these models the equilibrium is governed by a differential equation in the following form:

$$\text{(EV)}: \quad \frac{\partial}{\partial z}\left[c_{EV}(z)\frac{\partial \dot{\gamma}}{\partial z}\right] = k_{EV}(z)\gamma(z,t) \tag{27a}$$

$$\text{(VE)}: \quad \frac{\partial}{\partial z}\left[k_{VE}(z)\frac{\partial \gamma}{\partial z}\right] = c_{VE}(z)\dot{\gamma}(z,t), \tag{27b}$$

where $\gamma(z,t)$ represent the transverse displacement imposed to the shear layer at depth z and $\dot{\gamma}(z,t) := \dfrac{\partial}{\partial t}\gamma(z,t)$ is its time rate of change. In order to solve the problems above, we make use of the boundary conditions related to the mechanical schemes in fig.(2), expressed in the form of limits as follows:

$$\begin{cases} \lim\limits_{z\to 0}\gamma(z,t) = \gamma(t) \\ \lim\limits_{z\to\infty}\gamma(z,t) = 0. \end{cases} \tag{28}$$

By using such boundary conditions it is shown in (30) that eq.(27a) (or eq.(27a)) delivers a relationship between the force σ arising in the top layer in both models and the Caputo fractional derivative of displacement γ, i.e.

$$\sigma(t) = C_\beta \left({}^C\mathrm{D}^\beta_{0^+}\gamma\right)(t) \tag{29}$$

where we assumed the parameters as:

$$C_\beta := C_\beta^{(EV)} = \frac{G_0\Gamma(\beta)}{\Gamma(2-2\beta)\Gamma(1-\beta)2^{1-2\beta}}\left(\tau_\alpha^{(EV)}\right)^\beta \tag{30a}$$

$$\tau_\alpha^{(EV)} = \frac{\eta_0}{G_0}\frac{\Gamma(1+\alpha)}{\Gamma(1-\alpha)} \tag{30b}$$

and $\alpha = 1-2\beta$ for the EV material, whereas:

$$C_\beta := C_\beta^{(VE)} = \frac{G_0\Gamma(1-\beta)}{\Gamma(2-2\beta)\Gamma(\beta)2^{2\beta-1}}\left(\tau_\alpha^{(VE)}\right)^\beta \tag{31a}$$

$$\tau_\alpha^{(VE)} = \frac{\eta_0}{G_0}\frac{\Gamma(1-\alpha)}{\Gamma(1+\alpha)} \tag{31b}$$

and $\alpha = 2\beta - 1$ for the VE materials. The terms $\tau_\alpha^{(EV)}$, $\tau_\alpha^{(VE)}$ are relaxation times. The result expressed by eq.(29) highlights that these rheological models are capable to yield a force on the top layer relaxing with a power-law, ultimately resembling the macroscopic material behavior. In this respect, the boundary of such rheological models reproduces the material response.

A different, mechanical-type perspective of the rheological models of EV and VE materials is related to the evaluation of the overall stress in the external springs $\sigma^{(EV)}(z,\tau+\bar{t})dz$ of the EV model and of the external dashpots in the VE model $\sigma^{(VE)}(z,\tau+\bar{t})dz$ (see fig.3) that reads:

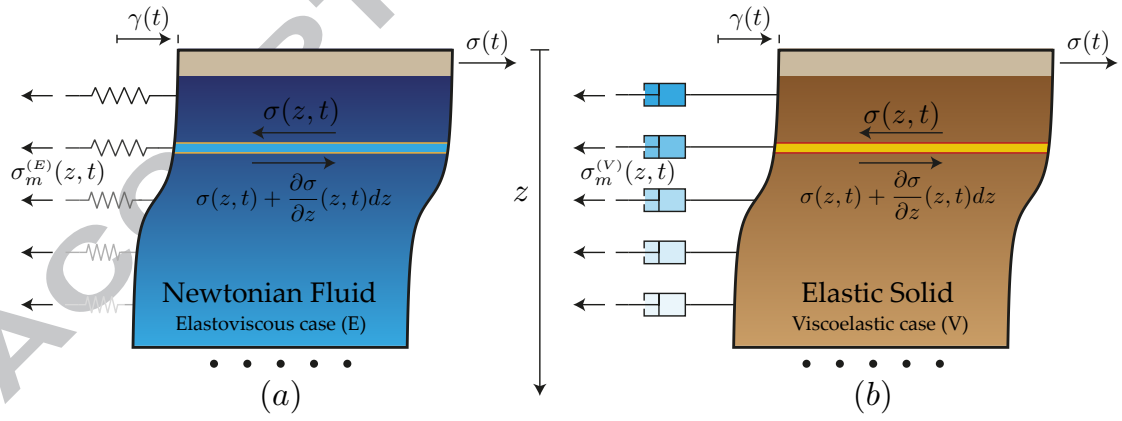


Figure 3: Elastoviscous (E) and Viscoelastic (V) deformed models and related external microstresses.

$$\sigma(t) = \int_0^\infty \sigma^{(EV)}(z, \tau + \bar{t}) dz = C_\beta \left({}^C\mathrm{D}_+^\beta \gamma \right)(t) \quad (0 \le \beta \le 1/2) \tag{32a}$$

$$\sigma(t) = \int_0^\infty \sigma^{(VE)}(z, \tau + \bar{t}) dz = C_\beta \left({}^C\mathrm{D}_+^\beta \gamma \right)(t) \quad (1/2 \le \beta \le 1), \tag{32b}$$

with the last equality obtained from the horizontal equilibrium conditions of the EV and VE models, respectively. The stress fields $\sigma^{(EV)}(z,t)$ and $\sigma^{(VE)}(z,t)$ may be obtained in terms of the Duhamel's integral of the displacement at the boundary $\gamma(t)$ as:

$$\sigma^{(EV)}(z, \tau + \bar{t}) = k(z)\gamma(z, \tau + \bar{t}) = k(z) \int_{-\infty}^{\tau + \bar{t}} H^{(EV)}(z, \tau + \bar{t} - r)\, \gamma(r)\, dr \tag{33a}$$

$$\sigma^{(VE)}(z, \tau + \bar{t}) = c(z)\dot{\gamma}(z, \tau + \bar{t}) = c(z) \int_{-\infty}^{\tau + \bar{t}} H^{(VE)}(z, \tau + \bar{t} - r)\, \dot{\gamma}(r) dr \tag{33b}$$

where the integral kernels in eqs.(33) may be obtained as the Right inverse Fourier transform of the frequency response fields, namely, $\hat{H}^{(EV)}(z,\omega)$ and $\hat{H}^{(VE)}(z,\omega)$, that reads (see Appendix B):

$$\hat{H}^{(EV)}(z,\omega) = \frac{G_0}{\Gamma(1-\beta)\Gamma(2-2\beta)2^{-\beta}} \left(\tau_\alpha^{EV} i\omega\right)^{\frac{\beta}{2}} z^\beta \, \mathrm{K}_\beta \left(\frac{z}{\sqrt{\tau_\alpha^{EV} i\omega}} \right) \tag{34a}$$

$$\hat{H}^{(VE)}(z,\omega) = \frac{G_0}{\Gamma(\beta)\Gamma(2-2\beta)2^{\beta-1}} \left(\tau_\alpha^{VE} i\omega\right)^{\frac{1+\beta}{2}} z^{1-\beta} \, \mathrm{K}_{1-\beta} \left(z\sqrt{\tau_\alpha^{VE} i\omega} \right) \tag{34b}$$

The achieved result is consistent with what is encountered in micromechanics, where a displacement (in this case γ) is prescribed on the boundary (namely the top plate) of a representative volume element of heterogeneous material and a boundary value problem is solved in the interior of such an element. This latter consideration is, in a broad sense, the result discussed in (30) where it was found that the response of the top plate, was the convolution between the rate of change of the imposed displacement and the relaxation functions $G^{(EV)}(\circ)$ or $G^{(VE)}(\circ)$ for (EV) or (VE) material models respectively.

4. The mechanical assessment of the free energy of FHM

The arguments in previous sections showed how the expression of free energies based on stress relaxations are not unique and this is a very serious drawback that may considerably affect the

mechanical derivations, such as studies about stability of either configurations or motions or both. On the other hand it has been assessed that the stored and the dissipated energy may be computed in unique way for Voigt, Maxwell or more refined models in which elastic springs and viscous dashpots are involved. On this solid ground, in sec.(3) we presented exact mechanical models corresponding to power-laws composed by springs and dashpots and in this section we aim to evaluate the stored (free) and the dissipated energy of fractional hereditary materials. Since for $0 \leq \beta \leq \frac{1}{2}$ and $\frac{1}{2} \leq \beta \leq 1$ the mechanical models are different from each another, the two cases will be discussed separately.

4.1. Derivation of the free energy for the Elasto-Viscous model

Let us consider a relaxation function $G(t) \propto t^{-\beta}$ with $0 \leq \beta \leq 1/2$ and, therefore, described by the Elasto-Viscous rheological model presented in sec.(3). In this case the overall dissipation rate of the material may be evaluated in terms of the energy rate dissipated by the internal dashpots as:

$$D_{EV}(t) = \frac{1}{2} \int_0^{\infty} c(z) \left(\frac{\partial \dot{\gamma}}{\partial z}(z,t) \right)^2 dz \tag{35}$$

and the rate of shear strain reads:

$$\frac{\partial \dot{\gamma}}{\partial z}(z,t) = \int_{-\infty}^{t} H_{,z}^{(EV)}(z, t-\tau) \dot{\gamma}(\tau)\, d\tau \tag{36}$$

The dissipation rate, involving the Green's function of the model reported in sec.3 may also be written in terms of the Fourier transforms $\hat{H}(z,\omega) = \Im_+ [H(z,t)]$ as:

$$\begin{aligned} D_{EV}(t) &= \frac{1}{2} \int_0^{+\infty} \frac{c(z)}{\left(\sqrt{2\pi}\right)^2} \int_0^{+\infty} e^{i\omega_1 t} \hat{H}_{,z}^{(EV)}(z,\omega_1) \hat{\dot{\gamma}}(\omega_1)\, d\omega_1 \int_0^{+\infty} e^{i\omega_2 t} \hat{H}_{,z}^{(EV)}(z,\omega_2) \hat{\dot{\gamma}}(\omega_2)\, d\omega_2 dz \\ &= \frac{1}{2(2\pi)^2} \int_0^{+\infty} \int_0^{+\infty} e^{i(\omega_1+\omega_2)t} \int_0^{+\infty} c(z) \hat{H}_{,z}^{(EV)}(z,\omega_1)\, \hat{H}_{,z}^{(EV)}(z,\omega_2)\, dz \hat{\dot{\gamma}}(\omega_2)\, \hat{\dot{\gamma}}(\omega_2)\, d\omega_2 d\omega_1 \\ &= \frac{1}{2} \frac{1}{\sqrt{2\pi}} \int_0^{+\infty} e^{i\omega_1 t} \frac{1}{\sqrt{2\pi}} \int_0^{+\infty} e^{i\omega_2 t} \hat{\hat{L}}^{(EV)}(\omega_1,\omega_2)\, \hat{\dot{\gamma}}(\omega_2)\, d\omega_2 \hat{\dot{\gamma}}(\omega_1)\, d\omega_1 \end{aligned} \tag{37}$$

where we defined

$$\hat{\hat{L}}^{(EV)}(\omega_1,\omega_2) := \int_0^{+\infty} c(z)\hat{H}^{(EV)}_{,z}(z,\omega_1)\,\hat{H}^{(EV)}_{,z}(z,\omega_2)\,dz \tag{38}$$

and as we denote the monodimensional inverse right Fourier transform as:

$$\begin{aligned} M^{(EV)}(\omega_1,t) :&= \frac{1}{\sqrt{2\pi}}\int_0^{+\infty} e^{i\omega_2 t}\hat{\hat{L}}^{(EV)}(\omega_1,\omega_2)\,\hat{\hat{\gamma}}(\omega_2)\,d\omega_2 \\ &= \Im_+^{-1}\left[\hat{\hat{L}}^{(EV)}(\omega_1,\circ)\,\hat{\hat{\gamma}}(\circ)\right](t) = \int_{-\infty}^{t}\hat{L}^{(EV)}(\omega_1,t-\tau_2)\,\hat{\hat{\gamma}}(\tau_2)d\tau_2 \end{aligned} \tag{39}$$

where, in the latter equality the convolution theorem has been applied. Substitution of Eq.(39) into eq.(37) allows for the dissipation rate in the form:

$$D_{EV}(t) = \frac{1}{2}\frac{1}{\sqrt{2\pi}}\int_0^{+\infty} e^{i\omega_1 t}\int_{-\infty}^{t}\hat{L}^{(EV)}(\omega_1,t-\tau_2)\,\hat{\hat{\gamma}}(\tau_2)d\tau_2\hat{\hat{\gamma}}(\omega_1)\,d\omega_1 = \frac{1}{2}\Im_+^{-1}\left[M^{(EV)}(t,\circ)\,\hat{\hat{\gamma}}(\circ)\right](t) \tag{40}$$

that, by means of the convolution theorem yields the dissipation rate in the form:

$$D_{EV}(t) = \frac{1}{2}\int_{-\infty}^{t} M^{(EV)}(t-\tau_1)\dot{\gamma}(\tau_1)\,d\tau_1 = \frac{1}{2}\int_{-\infty}^{t}\int_{-\infty}^{t} L^{(EV)}(t-\tau_1,t-\tau_2)\,\dot{\gamma}(\tau_2)\,d\tau_2\dot{\gamma}(\tau_1)\,d\tau \tag{41}$$

The expression of the dissiption rate provided in eq.(62) shows that, with the aid of the rheological model, a unique expression of the dissipation rate is obtained.

Summing up, the evaluation of the dissipation rate for the mechanical model of EV materials corresponding to power-laws relaxations $\propto t^{-\bar{\beta}}$ and $0 \le \bar{\beta} \le 1/2$ may be performed accounting for the overall energy dissipated in the purely viscous layers for applied transverse displacement $\gamma(t)$ at the top layer. In this regard the dissipation rate involves a double integral (see eq.62) with kernel provided by a two variable function, namley, $L^{(EV)}(\tau_1,\tau_2)$ that is obtained by means of an inverse double right Fourier transform of the operator $\hat{\hat{L}}^{(EV)}(\omega_1,\omega_2)$ reported in eq.(38).

The explicit evaluation of operator $\hat{\hat{L}}^{(EV)}(\omega_1,\omega_2)$ is a fundamental step to obtain an expression for the dissipation rate in the mechanical model and, substituting eq.(34 a) into eq.(38) it yields:

$$\hat{\hat{L}}^{(EV)}(\omega_1,\omega_2)=\frac{\left(\left(i\omega_1\tau_E\right)^{1-\beta}-\left(i\omega_2\tau_E\right)^{1-\beta}\right)\pi\mathrm{csc}\left(\pi\beta\right)}{2\tau_E i(\omega_1-\omega_2)} \tag{42}$$

that may be transformed to time domain yielding:

$$L^{(EV)}(t_1,t_2)=-\frac{2^{1-2\beta}(t_1+t_2)^{\beta-2}\tau_E^{-\beta}\eta_0\pi\mathrm{csc}\left(\pi\beta\right)}{\Gamma\left(2-\beta\right)\Gamma(\beta)^2\Gamma(\beta-1)} \tag{43}$$

The kernel operator in eq.(43), namely $L^{(EV)}(t_1,t_2)$ may be written, replacing the variables $t_1\rightarrow t-\tau_1$ and $t_2\rightarrow t-\tau_2$ and substituting the expression of the viscosity coefficient $\eta_0=\frac{\tau_E G_0\Gamma(2-2\beta)}{\Gamma(2\beta)}$, $\beta=1-\bar{\beta}$ with $0\leq\bar{\beta}\leq 1/2$, in the form:

$$L(t-\tau_1,t-\tau_2)=\frac{\bar{\beta}C_{EV}(\bar{\beta})(2t-\tau_1-\tau_2)^{-(1+\bar{\beta})}}{\Gamma(1-\bar{\beta})} \tag{44}$$

yielding, upon substitution in eq.(62), the dissipation rate $D_{EV}(t)$ in the form coalescing with eq.(23) assuming the relaxation function in eq.(4).

The result obtained in eq.(44) is of great importance since it states, clearly, that for power-law relaxation functions, the overall dissipation rate of the material for *any* strain history is always provided by the quadratic expression reported by (42). This result have been obtained by means of the rheological model correspoding to the relxation function of FHM that allows to separate,completely, the solid (elastic) phase and the fluid (viscous) phase of the material.

Summing up we showed that, despite the non-uniqueness of the free energy and of the dissipation rate associated to a particular functional class of the relaxation function, this is not the case of FHM. Indeed for such materials the relaxation function is expressed in terms of power-laws of real order and a rheological model that possess a power-law creep/relaxation function has been obtained in recent literature ((30)). The model has been used to define the state of FHM in aprevious paper ((43)) and in this paper it has been used to evaluate the dissipation rate. In this context has been shown that the dissipation rate obtained from the rheological model coincides with the dissipation rate involved in the Stavermann-Schwarzl expression of the free energy reported in(24) assuming a power-law relaxation function. We may conclude that the Stavermann-Schwarzl expression of the free energy function provided in (24) corresponds to the potential energy stored in the elastic solid

phase of fractional hereditary materials.

4.2. Derivation of the free energy for the Visco-Elastic material model

The arguments of previous section led the conclusion that, at least for the EV material model and, henceforth for decaying exponent in the range $0 \leq \beta \leq 1/2$ the energy stored in the linear elastic phase of a fractional hereditary material is provided by the Staverman-Schwartzl free energy function. In this section we show that a similar consideration may be withdrawn for the VE model of fractional hereditariness, that is for decaying exponent in the range $1/2 \leq \beta \leq 1$.

Indeed the evaluation of the dissipation rate $D_{VE}(t)$ for VE model is obtained by means of the overall energy dissipated by the external viscous dashpots of the VE model as:

$$D_{VE}(t) = \frac{1}{2}\int_0^{\infty} c(z)\left(\dot{\gamma}(z,t)\right)^2 dz \tag{45}$$

and, expressing the rate of displacement function in terms of function $H^{(VE)}(z,t)$ as:

$$\dot{\gamma}(z,t) = \int_{-\infty}^{t} H^{(VE)}(z,t-\tau)\dot{\gamma}(\tau)\,d\tau \tag{46}$$

the expression of the dissipation rate reads:

$$\begin{aligned} D_{VE}(t) &= \frac{1}{2}\int_0^{+\infty} \frac{c(z)}{\left(\sqrt{2\pi}\right)^2}\int_0^{+\infty} e^{i\omega_1 t}\hat{H}^{(VE)}(z,\omega_1)\hat{\dot{\gamma}}(\omega_1)\,d\omega_1 \int_0^{+\infty} e^{i\omega_2 t}\hat{H}^{(VE)}(z,\omega_2)\hat{\dot{\gamma}}(\omega_2)\,d\omega_2 dz \\ &= \frac{1}{2(2\pi)^2}\int_0^{+\infty}\int_0^{+\infty} e^{i(\omega_1+\omega_2)t}\int_0^{+\infty} c(z)\hat{H}^{(VE)}(z,\omega_1)\,\hat{H}^{(VE)}(z,\omega_2)\,dz\hat{\dot{\gamma}}(\omega_2)\,\hat{\dot{\gamma}}(\omega_2)\,d\omega_2 d\omega_1 \\ &= \frac{1}{2}\frac{1}{\sqrt{2\pi}}\int_0^{+\infty} e^{i\omega_1 t}\frac{1}{\sqrt{2\pi}}\int_0^{+\infty} e^{i\omega_2 t}\hat{\hat{L}}^{(VE)}(\omega_1,\omega_2)\,\hat{\dot{\gamma}}(\omega_2)\,d\omega_2\hat{\dot{\gamma}}(\omega_1)\,d\omega_1 \end{aligned} \tag{47}$$

where we defined:

$$\hat{\hat{L}}^{(VE)}(\omega_1,\omega_2) := \int_0^{+\infty} c(z)\hat{H}^{(VE)}(z,\omega_1)\,\hat{H}^{(VE)}(z,\omega_2)\,dz \tag{48}$$

and as we denote the monodimensional inverse right Fourier transform as:

$$\begin{aligned} M^{(VE)}(\omega_1,t) :&= \frac{1}{\sqrt{2\pi}}\int_0^{+\infty} e^{i\omega_2 t}\hat{\hat{L}}^{(VE)}(\omega_1,\omega_2)\,\hat{\dot{\gamma}}(\omega_2)\,d\omega_2 \\ &= \Im_+^{-1}\left[\hat{\hat{L}}^{(VE)}(\omega_1,\circ)\,\hat{\dot{\gamma}}(\circ)\right](t) = \int_{-\infty}^{t}\hat{L}^{(VE)}(\omega_1,t-\tau_2)\,\hat{\dot{\gamma}}(\tau_2)d\tau_2 \end{aligned} \tag{49}$$

yielding the form of the dissipation rate for the ViscoElastic material model as:

$$D_{VE}(t) = \frac{1}{2}\int_{-\infty}^{t} M^{(VE)}(t-\tau_1)\dot{\gamma}(\tau_1)\,d\tau_1 = \frac{1}{2}\int_{-\infty}^{t}\int_{-\infty}^{t} L^{(VE)}(t-\tau_1,t-\tau_2)\,\dot{\gamma}(\tau_2)\,d\tau_2\dot{\gamma}(\tau_1)\,d\tau \tag{50}$$

The explicit expression for the operator $\hat{\hat{L}}^{(VE)}(\omega_1,\omega_2)$ may be obtained in the form:

$$\hat{\hat{L}}^{(VE)}(\omega_1,\omega_2) = \frac{\left((i\omega_1\tau_V)^{\beta} - (i\omega_2\tau_V)^{\beta}\right)\pi\csc(\pi\beta)}{2\tau i(\omega_1-\omega_2)} \tag{51}$$

that may be transformed back in time domain, yielding for the VE case:

$$L^{(VE)}(t_1,t_2) = \frac{2^{1-2\beta}(t_1+t_2)^{\beta-1}\eta_0\tau_V^{-(\beta+1)}\Gamma(1-\beta)}{\Gamma(\beta)^2\Gamma(2\beta)} \tag{52}$$

The expression in eq.(52) may be still manipulated, accounting for the transformations $t_1 = t-\tau_1$ and $t_2 = t-\tau_2$ and $\eta_0 = \dfrac{\tau_V G_0\Gamma(2\beta)}{\Gamma(2-2\beta)}$ yielding:

$$L^{(VE)}(t-\tau_1,t-\tau_2) = \frac{\beta C_{VE}(\beta)(2t-\tau_1-\tau_2)^{-(\beta+1)}}{\Gamma(1-\beta)} \tag{53}$$

that may be substituted into eq.(50) to yield the dissipation rate in the form provided in eq.(23) for stress relaxation measures in the form of eq.(4). This result shows that, also with the VE model of FHM, the expression of the free energy function coincides with the functional in (24) for *any* strain histories undergone by the material.

The expression of the free energy function in term of the state of FHM for the VE material model is analogous to that provided in previous section and it has not been repeated for brevity.

5. The state-based free energy function of FHM

The arguments provided in previous sections showed that, for the case of power-law relaxation/creep measures of stress and strain, respectively, an unique expression of the free energy function that corresponds to the stored energy for any strain and stress history may be withdrawn from the mechanical equivalence of FHM.

The knowledge of the strain or stress history in a material sample, as required for *virgin* materials, is an ideal condition that is seldom available in applications where unknown strain histories $\varepsilon^{(\bar{t})}(\tau)$ may have been applied to the material; We refer to these cases as *non-virgin* materials. The stress measure in *non-virgin* materials involves the presence of the residual stress due to the unknown strain histories as well as the contribution of the strain process used to measure the stress. The presence of the residual stress may introduce important deviations among the measured value and the current value of the stress for materials with long-tails relaxation function as the power-law of FHM

In these cases we refer to the notion of the *state* of simple materials ((33), (35),(44)) yielding the definition of the equivalence class of strain histories that corresponds to the same material state. In this regard two histories are *equivalent* if any arbitrary continuation (often called either process or segment) of finite arbitrary duration yields either the same stress and/or the same mechanical work measures.

In this section we introduce the definition the state of FHM and we will define the the material free energy in (24) that represent the energy stored in the elastic phase of the material, defined in the following ψ_{SS}, in terms of material state.

The specific functional classes of equivalent strain histories for power-law relaxation functions, is obtained as we introduce two different strain histories $\epsilon^0_{(i)}(r)$ with $(i = 1, 2)$ up to time instant $\bar{t} = 0$ and then continued with the null history up to time t as:

$$\epsilon^0_{(i)}(r) := \epsilon_{(i)}(\bar{t} - r)U(\bar{t} - r) = \epsilon_{(i)}(-r)U(-r) \quad i = 1, 2 \quad r \leq t. \tag{54}$$

where $U(\circ)$ is the unit step function and the apex denotes the usual definition of the strain history up to the time instant $\bar{t} = 0$ that is continued with the null history up to time t. The definition

of material state up to time t does not depend on the specific strain process $\epsilon^p(r)$ for $0 \leq r \leq t$ following the application of the strain histories $\epsilon^{(0)}_{(i)}(r)$ with $(i = 1, 2)$ on the material sample. The strain function up to time t in the definition of the state may then be expressed as the superposition:

$$\epsilon_{(i)}(r) := \epsilon^0_{(i)}(r) + U(-r)\epsilon^p(r) \quad i = 1, 2 \text{ and } r \in (-\infty, t) \tag{55}$$

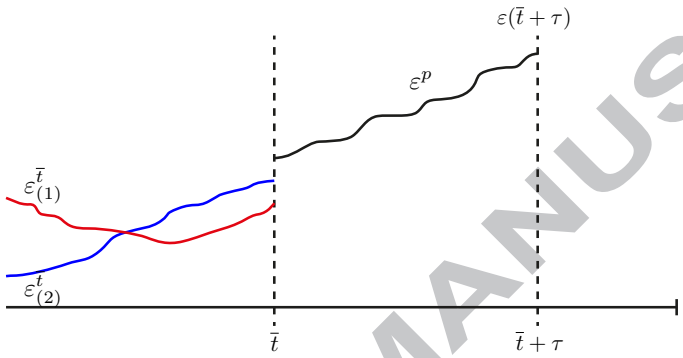


Figure 4: Past histories $\epsilon^{\bar{t}}_{(1)}$, $\epsilon^{\bar{t}}_{(2)}$ and deformation process ϵ^p

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Following the definition of state, we say that the strain histories $\epsilon^0_{(1)}(r)$ and $\epsilon^0_{(2)}(r)$ are equivalent if they correspond to the same stress measure at any time instant t with $t \geq 0$ whatever continuation process $\epsilon^p(r)$ is considered:

$$\sigma_{(1)}(t) = \sigma_{(2)}(t) \quad \forall t \geq 0 \tag{56}$$

where $\sigma_{(i)}(t)$ is the measured stress at time instant t corresponding to the strain histories $\varepsilon^0_{(i)}(r)$. The condition expressed in eq.(56), corresponds, to the condition:

$$\int_{-\infty}^{t} G(t-r)\dot{\epsilon}_{(1)}(r)dr = \int_{-\infty}^{t} G(t-r)\dot{\epsilon}_{(2)}(r)dr \tag{57}$$

that for the power-law relaxation of FHM is rewritten as:

$$C_\beta \left({}^C\mathrm{D}^\beta_+ \epsilon_{(1)} \right)(t) = C_\beta \left({}^C\mathrm{D}^\beta_+ \epsilon_{(2)} \right)(t) \tag{58}$$

In other words, as we define the history $\epsilon_*(t) := \epsilon_{(2)}(t) - \epsilon_{(1)}(t)$ condition in eq.(58) implies that:

$$C_\beta \left({}^C\mathrm{D}_+^\beta \epsilon_* \right)(t) = 0 \quad \forall t \geq 0 \tag{59}$$

It has been shown (see (43)) that the equivalence class of the extended strain histories does not contain the trivial case $\epsilon_* = 0$ only. In this setting the state of the material, denoted as $\mathcal{I}^0(t)$, is obtained as difference among the stress at time t evaluated for different histories but belonging to the same equivalence class, namely, $\varepsilon^{(eq)}(t) = \varepsilon_*(t)$ and the stress obtained for the actual value of the strain as:

$$\mathcal{I}^0(t) := C_\beta \left({}^C\mathrm{D}_+^\beta \epsilon^{(eq)} \right)(t) - \frac{C_\beta \epsilon(0)}{\Gamma(1-\beta) t^\beta}. \tag{60}$$

that corresponds to the generic element of the equivalence class as:

$$\epsilon^{(eq)}(t) = \frac{\left(\mathrm{I}_+^\beta \, \mathcal{I}^0 \right)(t)}{C_\beta} = \frac{1}{C_\beta \Gamma(\beta)} \int_{-\infty}^t \mathcal{I}^0(r)(t-r)^{\beta-1} dr. \tag{61}$$

The knowledge of the residual stress for virgin material, as well as of the state of FHM for non-virgin materials, does not allow one, in principle, to evaluate the free energy stored in the material as observed in sec.(2.2). The main reason is that the presence of the pure elastic (solid) phase and viscous (fluid) phase in a non-separable form for the power-law creep/relaxation functions, namely $J(t)$ and $G(t)$ does not allow for the evaluation of the dissipation rate or the elastic energy rate. In the previous section it has been shown that the indeterminacy may be removed if we use the rheological model of power-law relaxation function of FHM presented in sec.(3). Indeed, the clear separation of the viscous (dissipated) and elastic (stored) phases of the rheological model yields an expression of the dissipation rate of the material that corresponds exactly with free energy in ((24)), namely, $\psi_{ss}(t)$ (see eqs.44,52).

Under these circumstances the expression of the dissipation rate of the material may be also obtained in terms of the state of FHM, that is in terms of the equivalence class of strain histories

corresponding to the same stress measure as:

$$D_{EV}(t) = D_{VE}(t) = D(t) = \frac{1}{2}\frac{\beta+1}{C_\beta \Gamma(\beta)} \int_{-\infty}^{t}\int_{-\infty}^{t} (2t-\tau_1-\tau_2)^{-(\beta+1)} \dot{I}_+^\beta\left(\mathcal{I}^0\right)(\tau_1)\, \dot{I}_+^\beta\left(\mathcal{I}^0\right)(\tau_2)\, d\tau_1 d\tau_2 \tag{62}$$

that correspond to the dissipation rate replacing the generic strain history with eq.(61) yielding:

$$\psi_{ss}(t) = \frac{1}{2}\frac{1}{C_\beta \Gamma(\beta)} \int_{-\infty}^{t}\int_{-\infty}^{t} (2t-\tau_1-\tau_2)^{-\beta} \dot{I}_+^\beta\left(\mathcal{I}^0\right)(\tau_1)\, \dot{I}_+^\beta\left(\mathcal{I}^0\right)(\tau_2)\, d\tau_1 d\tau_2 \tag{63}$$

that is expressed by a quadratic form of a linear operator, namely the Riemann-Liouville fractional integral $I^\beta(\circ)$, applied to the state measure $\mathcal{I}^0(t)$ as it is expected for the linear measure of strain and stress used in the paper.

6. Conclusions

In this paper the authors discuss the free energy function for fractional hereditary materials in terms of a recently proposed mechanical model (29). Indeed the specific expression of the free energy for a given relaxation/creep function is not unique. This problem is well-known and several expression of the free energy have been provided satisfying the thermodynamic restrictions about the minimum energy rate or the maximum entropy function. However for relaxation function, characterized by a single exponential (corresponding to an arrangement of elastic spring and a viscous dashpot) the expressions of the free energy coincides. In these cases it has been proved, that the expression of the free energy in (24) is the elastic potential energy stored in the solid phase of the the material. In case of other expressions of the relaxation function, however, no information about the physics beyond the free energy expressions have been presented in scientific literature.

The authors showed that a physical context to the free energy in (24) may be withdrawn, also, for the case of power-law relaxation function possessed by fractional hereditary materials. This equivalence has been obtained by means of the exact mechanical description of power-laws $\propto t^\beta$ provided in previous papers involving either the case of Elasto-Viscous material models $0 \leq \beta \leq 1/2$ or the Visco-Elastic material models $1/2 \leq \beta \leq 1$ (see for instance (29),(30)). Indeed, as the equivalence among a power-law relaxation and a mechanical model represented by a proper

setting of linear elastic springs (solid phase) and linear viscous dashpots (fluid phase) has been established then the evaluation of the stored and dissipated energy is unique. In this paper we showed that the dissipation rate of the material must be evaluated by means of the overall dissipation of the viscous dashpots of the rheological model. Hence the energy rate dissipated during *any* strain/stress history is provided, only, by the overall dissipation rate of the viscous elements of the fluid phase. Therefore, the rheological model equivalent to power-law creep/relaxation functions is a crucial point to evaluate the mechanical energy stored and dissipated in the material during any strain/stress history. It has been shown that the energy rate of FHM coincides with the energy rate in (24) for the power-law relaxation function. The free energy function corresponding to FHM has also been formulated in terms of the material state that represents the residual stress in non-virgin materials.

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Appendix A

In this Appendix we address some basic notions about fractional calculus. The Euler-Gamma function $z \mapsto \Gamma(z)$ may be considered as the generalization of the factorial function since, as z assumes integer values as $\Gamma(z+1) = z!$ and it is defined as the result of the integral

$$\Gamma(z) = \int_0^\infty e^{-x} x^{z-1} dx. \tag{64}$$

Riemann-Liouville fractional integrals and derivatives with $0 < \beta < 1$ of functions defined on the entire real axis $\mathbb{R}$ have the following forms:

$$\left(\mathrm{I}_+^\beta f\right)(t) = \frac{1}{\Gamma(\beta)} \int_{-\infty}^t \frac{f(\tau)}{(t-\tau)^{1-\beta}} d\tau \tag{65a}$$

$$\left(\mathrm{D}_+^\beta f\right)(t) = \frac{1}{\Gamma(1-\beta)} \frac{d}{dt} \int_{-\infty}^t \frac{f(\tau)}{(t-\tau)^{\beta}} d\tau, \tag{65b}$$

whereas their counterparts defined over the whole real axis take the following forms:

$$\left(\mathrm{I}_a^\beta f\right)(t) = \frac{1}{\Gamma(\beta)} \int_a^t \frac{f(\tau)}{(t-\tau)^{1-\beta}} d\tau \tag{66a}$$

$$\left(\mathrm{D}_a^\beta f\right)(t) = \frac{f(a)}{\Gamma(1-\beta)(t-a)^{\beta}} + \frac{1}{\Gamma(1-\beta)} \int_a^t \frac{\dot{f}(\tau)}{(t-\tau)^{\beta}} d\tau. \tag{66b}$$

The relation (66b) is a direct consequence of Corollary of Lemma 2.1 in (11) (p.32). Besides Riemann-Liouville fractional operators defined above, another class of fractional derivatives that is often used in the context of fractional viscoelasticity is represented by Caputo fractional derivatives defined as:

$$\left({}^{C}\mathrm{D}^{\beta}_{a^+} f\right)(t) := \mathrm{I}^{m-\beta}_{a^+}\left(\mathrm{D}^{m}_{a^+} f\right)(t) \quad m-1<\beta<m \tag{67}$$

and whenever $0<\beta<1$ it reads as follows:

$$\left({}^{C}\mathrm{D}^{\beta}_{a^+} f\right)(t) = \frac{1}{\Gamma(1-\beta)}\int_a^t \frac{\dot{f}(\tau)}{(t-\tau)^{\beta}} d\tau \tag{68}$$

A closer analysis of (66b) and (68) shows that Caputo fractional derivative coincides with the integral part of the Riemann-Liouville fractional derivative in bounded domain. Moreover, the definition in (67) implies that the function $f(t)$ has to be absolutely integrable of order m (in (68) $m=1$). Whenever $f(a)=0$ Caputo and Riemann-Liouville fractional derivatives coalesce.

Similar considerations hold true also for Caputo and Riemann-Liouville fractional derivatives defined on the entire real axis. Caputo fractional derivatives may be considered as the interpolation among the well-known, integer-order derivatives, operating over functions $f(\circ)$ that belong to the class of Lebesgue integrable functions. As a consequence, they are very useful in the mathematical description of complex systems evolution.

The right-Fourier Transform of Caputo fractional derivative and Riemann-Liouville fractional integral read as follows:

$$\mathcal{F}_+\left\{\left(\mathrm{I}^{\beta}_{+} f\right)(\circ)\right\}(\omega) = (-i\omega)^{-\beta}\hat{f}_+(\omega) \tag{69a}$$

$$\mathcal{F}_+\left\{\left({}^{C}\mathrm{D}^{\beta}_{+} f\right)(\circ)\right\}(\omega) = (-i\omega)^{\beta}\hat{f}_+(\omega) \tag{69b}$$

where

$$\mathcal{F}_+\left\{f(\circ)\right\}(\omega) := \hat{f}_+(\omega) := \int_0^{\infty} f(t)\mathrm{e}^{-i\omega t} dt. \tag{70}$$

Appendix B

The displacement functions along the depth of the continuum column of the mechanical models displayed in Figure 2 (see (29) for more details) analyzed in the right Fourier domain assume the following forms

$$\hat{\gamma}^{(E)}(z,\omega) = \hat{\gamma}(\omega)\frac{(\tau_\alpha^{(E)} i\omega)^{-\frac{\overline{\beta}}{2}}}{\Gamma(\overline{\beta})2^{\overline{\beta}-1}} z^{\overline{\beta}} \mathrm{K}_{\overline{\beta}}\left(\frac{z}{\sqrt{\tau_\alpha^{(E)} i\omega}}\right) \tag{71a}$$

$$\hat{\gamma}^{(V)}(z,\omega) = \hat{\gamma}(\omega)\frac{(\tau_\alpha^{(V)} i\omega)^{\frac{\beta}{2}}}{\Gamma(\beta)2^{\beta-1}} z^{\beta} \mathrm{K}_{\beta}\left(z\sqrt{\tau_\alpha^{(V)} i\omega}\right), \tag{71b}$$

for the (E) and (V) case respectively, where $\hat{\gamma}(\omega)$ is the right Fourier transform of the imposed displacement at the top plate, $\mathrm{K}_\nu(\circ)$ is the modified Bessel function of the second kind of order ν, $\tau_\alpha^{(E)}$ and $\tau_\alpha^{(V)}$ have been defined in equations(30b) and (31b) respectively and $\overline{\beta} = 1-\beta$. The relationships above are preparatory to enable us evaluating $\hat{\sigma}_m^{(E)}$ and $\hat{\sigma}_m^{(V)}$, namely the time-(right)-Fourier transforms of the microstress, arising in the external devices for both models.

Bearing in mind that $\overline{\alpha} = 2\overline{\beta} - 1$, the right-Fourier Transform of the microstress related to the external springs for the (E) case may be written as follows:

$$\begin{aligned}
\hat{\sigma}_m^{(E)}(z,\omega) &= \frac{1}{A} k_{\scriptscriptstyle E}(z)\hat{\gamma}^{(E)}(z,\omega) \\
&= \frac{G_0}{\Gamma(1+\overline{\alpha})} z^{-\overline{\alpha}}\ \hat{\gamma}(\omega)\frac{(\tau_\alpha^{(E)} i\omega)^{-\frac{\overline{\beta}}{2}}}{\Gamma(\overline{\beta})2^{\overline{\beta}-1}} z^{\overline{\beta}} \mathrm{K}_{\overline{\beta}}\left(\frac{z}{\sqrt{\tau_\alpha^{(E)} i\omega}}\right) \\
&= \hat{\gamma}(\omega)\frac{G_0\,(\tau_\alpha^{(E)} i\omega)^{\frac{\beta-1}{2}}}{\Gamma(2-2\beta)\Gamma(1-\beta)2^{-\beta}} z^{\beta} \mathrm{K}_{1-\beta}\left(\frac{z}{\sqrt{\tau_\alpha^{(E)} i\omega}}\right).
\end{aligned} \tag{72}$$

The resultant of such quantities can be computed as the integral across the external devices (the springs for the (E) case). In order to evaluate this quantity, we recall that following result holds for a modified Bessel function of second kind integral:

$$\int_0^\infty z^{\mu-1}\mathrm{K}_\nu\left(Az\right)dz = 2^{\mu-2}A^{-\mu}\Gamma\left(\frac{\mu-\nu}{2}\right)\Gamma\left(\frac{\mu+\nu}{2}\right). \tag{73}$$

By assuming $\mu - 1 = \beta$ and $\nu = 1-\beta$ in (73) and with the aid of (72), the resultant of the

right-Fourier Transform of the microstress for the (E) case arises in the following form:

$$\begin{aligned}\int_0^\infty \hat{\sigma}_m^{(E)}(z,\omega)dz &= \hat{\gamma}(\omega)\frac{G_0\left(\tau_\alpha^{(E)}i\omega\right)^{\frac{\beta-1}{2}}}{\Gamma(2-2\beta)\Gamma(1-\beta)2^{-\beta}}\int_0^\infty z^\beta \mathrm{K}_{1-\beta}\left(\frac{z}{\sqrt{\tau_\alpha^{(E)}i\omega}}\right)dz \\ &= \hat{\gamma}(\omega)\frac{G_0\left(\tau_\alpha^{(E)}i\omega\right)^{\frac{\beta-1}{2}}}{\Gamma(2-2\beta)\Gamma(1-\beta)2^{-\beta}}\ 2^{\beta-1}\left(\frac{1}{\tau_\alpha^{(E)}i\omega}\right)^{-\frac{(\beta+1)}{2}}\Gamma(\beta) \\ &= \hat{\gamma}(\omega)\underbrace{\frac{G_0\left(\tau_\alpha^{(E)}\right)^\beta\Gamma(\beta)}{\Gamma(2-2\beta)\Gamma(1-\beta)2^{1-2\beta}}}_{C_\beta^E}(i\omega)^\beta\left(\frac{i\omega}{i\omega}\right) \\ &= (i\omega)\hat{\gamma}(\omega)\ C_\beta^E(i\omega)^{\beta-1}.\end{aligned} \tag{74}$$

Hence, by taking the inverse right-Fourier transform of both sides and by using Fubini's Theorem on the left-hand side, we have:

$$\int_0^\infty \sigma_m^{(E)}(z,t)dz = \int_{-\infty}^t G(t-\tau)\dot{\gamma}(\tau)d\tau, \tag{75}$$

since the right Fourier transform of the assumed relaxation function in (4) takes the form:

$$\mathcal{F}_+\left\{\frac{C_\beta}{\Gamma(1-\beta)}t^{-\beta}\right\}(\omega) = C_\beta(i\omega)^{\beta-1}. \tag{76}$$

Similarly, whenever it is assumed that $\alpha = 2\beta - 1$ the microstress for the external dashpots for the (V) case can be evaluated as follows:

$$\begin{aligned}\hat{\sigma}_m^{(V)}(z,\omega) &= \frac{1}{A}c_V(z)(i\omega)\hat{\gamma}^{(V)}(z,\omega) \\ &= \frac{\eta_0}{\Gamma(1+\alpha)}z^{-\alpha}\ (i\omega)\hat{\gamma}(\omega)\frac{\left(\tau_\alpha^{(V)}i\omega\right)^{\frac{\beta}{2}}}{\Gamma(\beta)2^{\beta-1}}z^\beta \mathrm{K}_\beta\left(z\sqrt{\tau_\alpha^{(V)}i\omega}\right) \\ &= \frac{G_0\tau_\alpha^{(V)}}{\Gamma(1+\alpha)}\frac{\Gamma(1+\alpha)}{\Gamma(2-2\beta)}\ (i\omega)\hat{\gamma}(\omega)\frac{\left(\tau_\alpha^{(V)}i\omega\right)^{\frac{\beta}{2}}}{\Gamma(\beta)2^{\beta-1}}z^{1-\beta}\mathrm{K}_\beta\left(z\sqrt{\tau_\alpha^{(V)}i\omega}\right) \\ &= \hat{\gamma}(\omega)\frac{G_0\left(\tau_\alpha^{(V)}i\omega\right)^{\frac{\beta+2}{2}}}{\Gamma(\beta)\Gamma(2-2\beta)2^{\beta-1}}z^{1-\beta}\mathrm{K}_\beta\left(z\sqrt{\tau_\alpha^{(V)}i\omega}\right)\end{aligned} \tag{77}$$

where equation (31b) has been used in the following form:

$$\tau_\alpha^{(V)} = \frac{\eta_0}{G_0}\frac{\Gamma(1-\alpha)}{\Gamma(1+\alpha)} \quad\Longrightarrow\quad \eta_0 = \tau_\alpha^{(V)}G_0\frac{\Gamma(1+\alpha)}{\Gamma(2-2\beta)}. \tag{78}$$

The use of equation (77) and the assumptions $\mu - 1 = 1 - \beta$ and $\nu = \beta$ in equation (73) allow for

writing the overall microstress for the (V) case in the following form:

$$\begin{aligned}\int_0^\infty \hat{\sigma}_m^{(V)}(z,\omega)dz &= \hat{\gamma}(\omega)\frac{G_0\left(\tau_\alpha^{(V)}i\omega\right)^{\frac{\beta+2}{2}}}{\Gamma(\beta)\Gamma(2-2\beta)2^{\beta-1}}\int_0^\infty z^{1-\beta}\mathrm{K}_\beta\left(z\sqrt{\tau_\alpha^{(E)}i\omega}\right)dz \\ &= \hat{\gamma}(\omega)\frac{G_0\left(\tau_\alpha^{(V)}i\omega\right)^{\frac{\beta+2}{2}}}{\Gamma(\beta)\Gamma(2-2\beta)2^{\beta-1}}\ 2^{-\beta}\left(\tau_\alpha^{(V)}\right)^{\frac{\beta-2}{2}}\Gamma(1-\beta) \\ &= \hat{\gamma}(\omega)\underbrace{\frac{G_0\left(\tau_\alpha^{(V)}\right)^{\beta}\Gamma(1-\beta)}{\Gamma(\beta)\Gamma(2-2\beta)2^{2\beta-1}}}_{C_\beta^V}\ (i\omega)^\beta\left(\frac{i\omega}{i\omega}\right) \\ &= (i\omega)\hat{\gamma}(\omega)\ C_\beta^V(i\omega)^{\beta-1}.\end{aligned} \tag{79}$$

Now, proceeding like in equation (75) we get:

$$\int_0^\infty \sigma_m^{(V)}(z,t)dz = \int_{-\infty}^t G(t-\tau)\dot{\gamma}(\tau)d\tau \tag{80}$$

The results addressed by equations (74)-(75) and equations (79)-(80) show that it is possible to compute the resultant of the microstresses in both models without knowing explicitly the transfer function $H(z,t)$ in the time domain.