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## Free energy and states of fractional-order hereditariness

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#### 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 cannot 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.

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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 (Nutting, 1921; Gemant, 1936; Jaishankar and McKinley, 2013; Nawaz et al., 2012). These materials show significative deviations of their mechanical and rheological features from well-studied 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 cannot 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 Baumgaertel and Winter, 1992) 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  $0 \le \beta \le 1$  (Lakes, 2009; Magin and Royston, 2010) yielding, by the Boltzmann superposition integral, constitutive equations in terms of fractional operators (Scott-Blair, 1947; Caputo, 1974; Mainardi, 2010; Samko et al., 1993). 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  $d^j f(x) \to d^\beta f(x)$ . 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 (Di Paola and Zingales, 2008; Cottone et al., 2009), thermodynamics (Povstenko, 2005; Povstenko, 2009; Sapora et al., 2013; Borino et al., 2011; Mongiovi and Zingales, 2013) beside rheology (Bagley and Torvik, 1983; Schiessel and Blumen, 1993).

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 a unique definition of the free

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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 Volterra (1940), Graffi and Fabrizio (1989) and DelPiero and Deseri (1996) and, as those properties are satisfied, several well-known different expressions for free energies may be found in scientific literature (Staverman and Schwarzl, 1952; Breuer and Onat, 1964; Gurtin and Hrusa, 1988; Morro and Vianello, 1989; Deseri et al., 2006). 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 cannot 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 (Di Paola et al., 2013a,b; Di Paola and Zingales, 2012). 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 (Deseri et al., 2013).

In this paper the model is further investigated to provide a 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 (Noll, 1972). 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 (Di Paola et al., 2013), and then, the correct definition of the material state is crucial for the general derivation of the free energy functional (Graffi and Fabrizio, 1989; DelPiero and Deseri, 1997). 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 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 Section 3 the rheological description of power-laws creep/relaxations will be briefly outlined for *EV* and *VE* material models. Section 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 Section 5 with some comments reported in Section 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. Mainardi, 2010): (*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 (Section 2.1). The definition of the material free energy and its relation with the stress measure are discussed in Section 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  $\sigma$  and  $\epsilon$ . Whenever either the strain or the stress are prescribed, the constitutive relations for the corresponding derived quantities read as follows:

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

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

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