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Abstract

In this paper the authors deals with the heredity behavior of hydroxyapatite-based composite used for cranioplastic surgery. It is shown that biomimetic prostheses, for their microstructural morphology, have a mechanical behavior that can be well described by an isotropic fractional-order hereditary model. The three-axial isotropic behavior is framed in the context of fractional-order calculus and same details about thermodynamical restrictions of memory functions used in the formulation of the three-axial isotropic constitutive equations. A mechanical model that corresponds, exactly, to the three-axial isotropic hereditariness is also introduced in the paper.

Keywords	Biomimetic materials,cranioplasty; fractional calculus; power-law hereditariness; isotropic hereditariness.

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Dear Editor,

Enclosed please found a manuscript for possible publication on International Journal of Non-Linear Mechanics by Bologna, Graziano, Deseri and myself.

Looking forward to hear from at your earliest convenience.

Massimiliano Zingales

Power-Laws hereditariness of biomimetic ceramics for cranioplasty neurosurgery

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Abstract

In this paper the hereditary behavior of hydroxyapatite-based composites used for cranioplastic surgery is discussed in the context of material isotropy. Mixtures of collagen and hydroxiapatite composites are classified as biomimetic ceramic composites with hereditary properties modeled in the paper fractional-order calculus. Isotropy of the biomimetic ceramic is assumed and the thermodynamic of restrictions among material parameters are provided. The proposed formulation of the fractional-order isotropic hereditariness has been further exploited by means of a novel mechanical hierarchy that corresponds, exactly, to the three-dimensional fractional-order constitutive model introduded in the paper.

Keywords: `Biomimetic materials,cranioplasty, fractional calculus, power-law hereditariness, isotropic hereditariness.`
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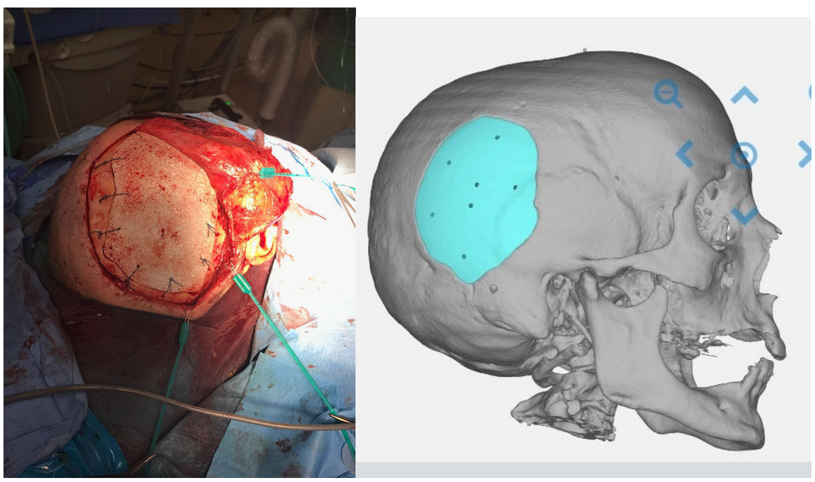
1. Introduction

Cranioplastic neurosurgery is nowadays an important issue worldwide since it is necessary both in traumatic therapies or in presence of specific oncologic pathology. Cranioplasty is a surgical procedure that aims to re-establish the skull integrity following a previous craniotomy due to the occurrence of traumas, tumors and/or congenital malformations. In all cases cranioplasty can be considered as the conclusive action of a surgery initiated by the removal of a bone operculum fig.1.



(a) (b)

Figure 1: cranioplasty surgery

Ideally, cranioplasty procedures should provide restoration of the protective functions of the skull with maintenance of the original aesthetics and long-term mechanical performance [1] The ideal material for cranioplasty should be chemically inert, biocompatible, biomechanically reliable, easily manufactured, individually shaped, safe, and able to promote osteoblast migration. Today synthetic implants based on metallic (mainly titanium) or acrylic plaques (mainly polymethylmeta-crylate or polyetheretherketone) are widely used in cranioplasty procedures. These are bioinert materials with good biocompatibility, resistance to infections, ease of sterilization, ability to be subjected to imaging diagnostics, and the capacity to undergo flexible design for adaptation to different clinical cases. They exhibit good mechanical strength, which offers adequate brain protection from external shocks. However, they present poor osteogenic and osteoconductive ability, thus resulting in a foreign body functioning as a shell expected to provide brain protection, but connected to the surrounding bone only by its perimeter contact surface. In order to overcome many limitations an Hydroxyapatite (HA)-based material has been widely considered for decades as the gold standard for bone scaffolds, as its composition is very close to that of bone mineral, thus exhibiting excellent biocompatibility, a low inflammatory reaction as well as good osteogenic ability and osteoconductivity. The hydrophilic character of HA favors cell attachment and tight adhesion of bone to the scaffold surface, which is a key target for the stability of the bone/implant interface. Therefore, HA scaffolds presenting wide, open and interconnected multiscale porosity can induce extensive bone ingrowth and penetration throughout the whole scaffold, partly thanks to the possibility of massive fluid perfusion, which triggers and assists neovascularization. Hence, cranial reconstruction using synthetic porous HA has recently become the subject of intense debate among surgeons, and it now represents a new concept in cranioplasty procedures. The custom-made concept was first applied to porous hydroxyapatite because of the need to overcome the fragility of the material itself. Among the advantages of HA-based prosthesis there is the important issue of customization.

Indeed, in presence of cranioplasty the morphology of the bone to be replaced with a synthetic prosthesis must match, completely, the original bone to accelerate the osteointegration of the prosthesis [2, 3, 4] in the surgical hole. In fig.(2 a-d) an human parietal bone and its synthetic prosthesis fig.(3 a-c) have been obtained from at universitary neurosurgery hospital in Palermo. The synthetic bone used for replacement is a CustomBone® (Finceramica Faenza), namely custom-made, porous hydroxyapatite scaffolds with total porosity in the range of 60 to 70 % and pore architecture based on macro-pores (> 100 micron

) interconnected with micro-pores (5-10 micron). CustomBone® scaffolds were obtained by reproduction of the patients bone defect as modeled by 3D CT scan and its represented as a composite ceramics material obtained from chemical deposition of hydroxyiapatite within a small fraction of collagen type I (see fig.1a).

The use of biomimetic ceramics to replace cortical as well as trabecular bone is well defined to technique in bone surgery [5]. Indeed the mechanical feature of the prosthesis in terms of elastic moduli and the strength of the biomimetic composite of integration are very similar. However, the use of ceramic materials to replace the bones of human head may involve different behavior in terms of energy dissipation. Indeed biologic tissues show marked hereditariness that is due to the reptation of the collagen chains of the material as well as to the fiber recruitment in the tissues. Material hereditariness involves additional stresses that may be applied to the grafted ceramics prosthesis and may lead to fracture propagation during patient follow-up [6].

The hereditary properties of bone in uniaxial test are represented by creep $J(t)$ and relaxation $G(t)$ function that formulated in terms of power-law $J(t) \propto t^\beta$ and $G(t) \propto t^{-\beta}$ with $0 \leq \beta \leq 1$, yields accurate description of experimental data [5, 7, 8, 9, 10]. Power-laws hereditariness in conjuction with Boltzmann superposition yields constitutive behavior in terms of the so-called fractional integrals and derivatives. Fractional calculus may be considered as generalization of the classical differential calculus to real-order integration and differentiation $\left(i.e.df/dt \to d^\beta f/dt^\beta\right)$ with $\beta \in [0, 1]$ as reported in classical references [11, 12, 13, 14, 15]. In such a context, uniaxial hereditariness [16, 17, 18, 19] involving fractional order stress-strain relations have been reported since the beginning of the twentyth century [20, 13] defining the so-called springpot element [21, 22].

In presence of tensorial stress/strain state, as in continuum mechanic description of biomimetic prosthesis, no generalities have been reported in scientific literature to capture multiaxial hereditariness with fractional-order calculus, at the best of authors' knowledge. Indeed, in several cases, recently discussed

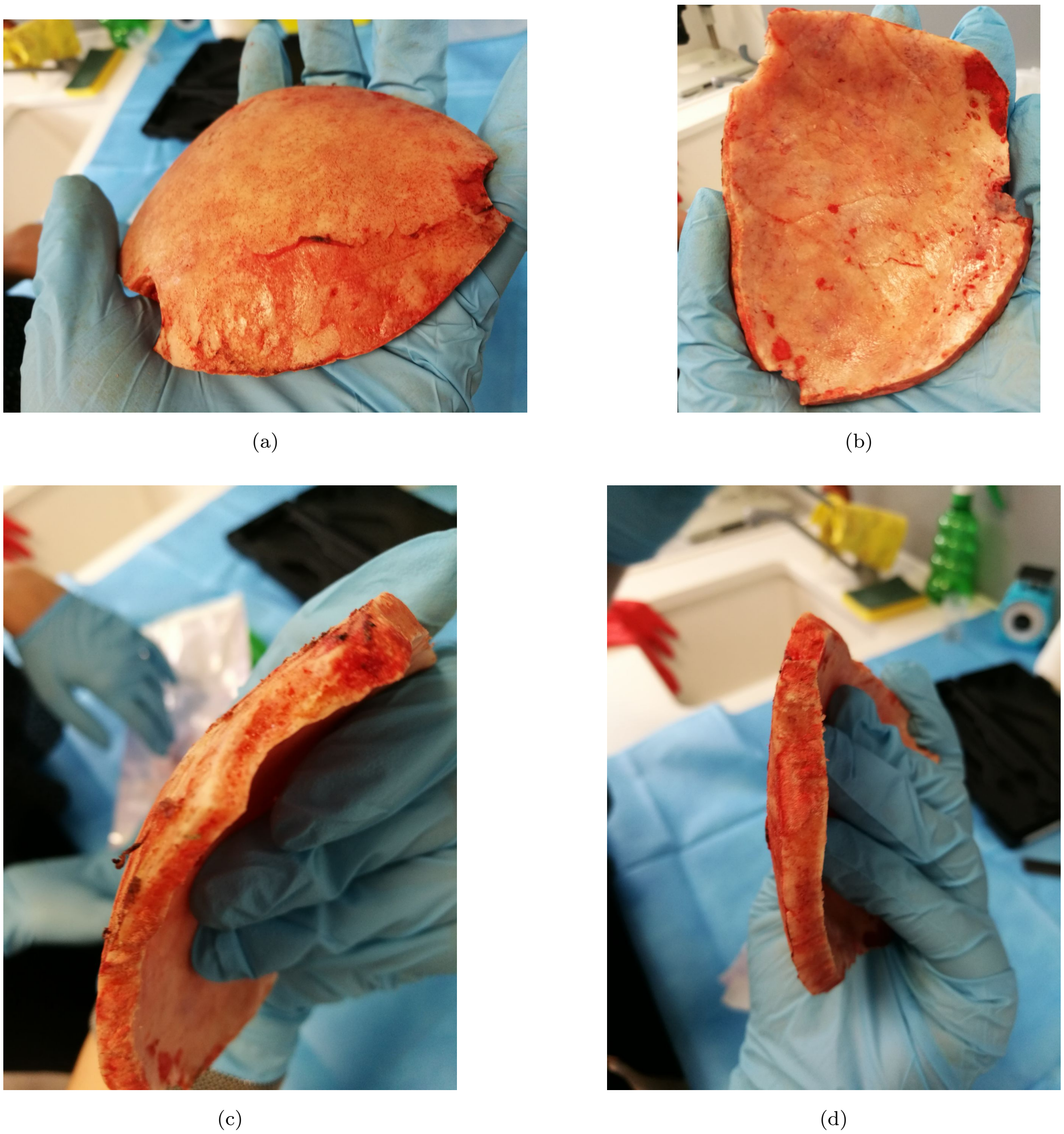
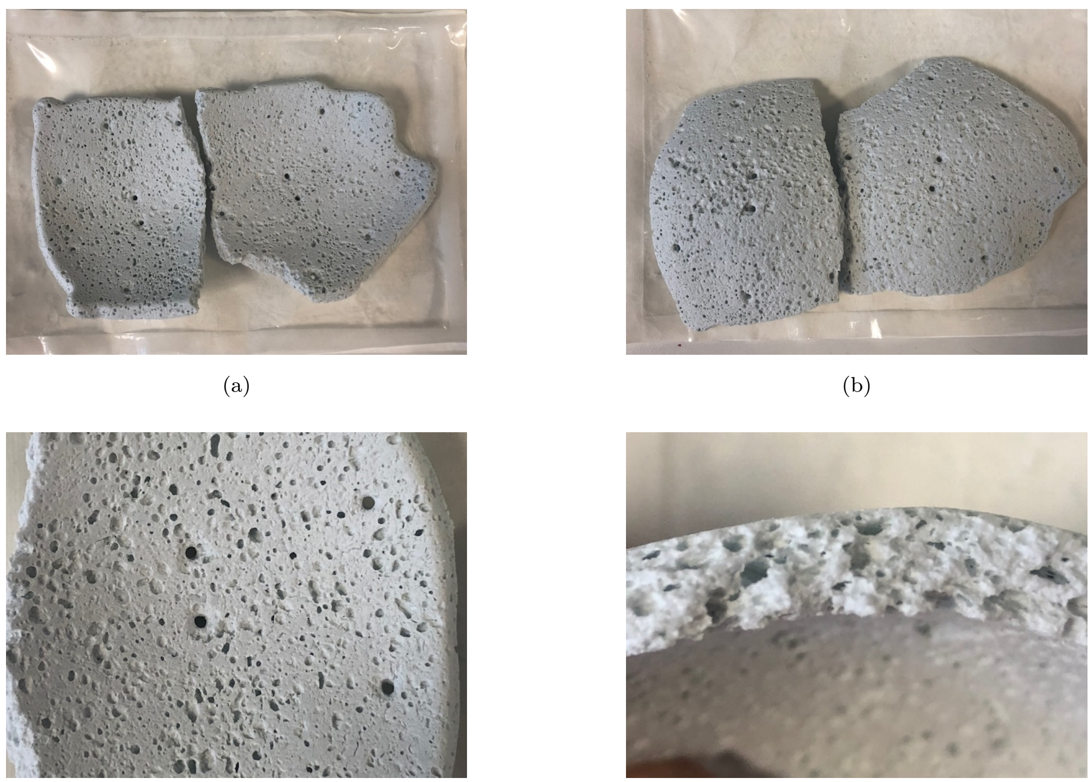


Figure 2: human parietal bone



(a) (b) (c) (d)

Figure 3: CustomBone® prosthesis morphology

in scientific literature [23, 24] the use of power-laws without thermodynamic restrictions to the parameters do not guarantee positive entropy rate for any strain/stress process involved by material.

In this paper a three axial constitutive relation describing material hereditariness is discussed in the context of power-laws functional classes of the relaxation/creep functions. It will be shown that, under the assumption of material isotropy, thermodynamical restrictions on the constitutive parameters allows to formulate the constitutive behavior in terms of a Caputo fractional derivative that is formally analogous to the constituitve behavior in uniaxial state of stress/strain. Additionally a novel mechanical hierarchy

The paper is organized as follows: Generalities about fractional-order calculus and isotropic hereditariness are provided in sec.2; In sec.3 a mechanical hierarchy that corresponds exactly to the isotropic fractional-order hereditariness is reported. Some conclusions about the proposed model of isotropic hereditariness and the influence on the mechanics of the biomimetic ceramics prosthesis have been withdrawn in sec.4

2. Power-law hereditariness of isotropic biomimetic ceramics

In this section the constitutive relations in presence of power-law hereditariness are outlined. In sec.2.1 main arguments of power-law hereditariness under uniaxial stress/strain are shortly outlined. Generalization to the isotropic case is defined in sec.2.2 and thermodynamic restrictions on the material parameters is introduced in sec.2.3.

2.1. Uniaxial power-law hereditariness: The fractional order constitutive equation

The constitutive behavior of materials in long-standing mechanical tests is described by means of the well-known creep and relaxation functions, dubbed $J(t)$ and $G(t)$, respectively. The linear superposition applied to a generic stress/strain history, namely $\sigma(\tau)$ and $\varepsilon(\tau)$ with $\tau \leq t$, yields:

$$\sigma(t) = \int_0^t G(t-\tau) d\varepsilon(\tau) = \int_0^t G(t-\tau)\dot{\varepsilon}(\tau) d\tau \tag{1a}$$

$$\varepsilon(t) = \int_0^t J(t-\tau) d\sigma(\tau) = \int_0^t J(t-\tau)\dot{\sigma}(\tau) d\tau \tag{1b}$$

Eqs.(1a, b) are defined in terms of Boltzman superposition with $d\sigma = \dot{\sigma} dt$ and $d\varepsilon = \dot{\varepsilon} dt$ increments with $\dot{[\cdot]} = \frac{d}{dt}$. Creep and relaxation functions characterize the material behavior and they must satisfy the conjugation relation $\hat{J}(s)\hat{G}(s) = 1/s^2$, where s indicates the Laplace parameter and $\hat{f}(s) = \mathcal{L}[f(t)]$ the Laplace transform of the generic function $f(t)$. In the context of materials hereditariness, power-law representation of creep and relaxation functions, i.e. $J(t)$ and $G(t)$, was introduced at the beginning of the last century [20],

$$G(t) = \frac{C_\beta}{\Gamma(1-\beta)} t^{-\beta}, \tag{2a}$$

$$J(t) = \frac{1}{C_\beta \Gamma(\beta+1)} t^{\beta} \tag{2b}$$

where $\Gamma(\cdot)$ is the Euler-Gamma function, $\beta \in [0,1]$ and $C_\beta > 0$, are material parameters, that may be estimated through a best-fitting procedure of experimental data [25, 26]. Straightforward manipulations show that the power-law functional class in eqs.(2a, b), satisfies the conjugation relation and it yields, upon substitution in eqs. (1a, b) the constitutive relations:

$$\sigma(t) = \frac{C_\beta}{\Gamma(1-\beta)} \int_0^t (t-\tau)^{-\beta} \dot{\varepsilon}(\tau) d\tau = C_\beta \left(D_{0^+}^{\beta} \varepsilon \right)(t) \tag{3a}$$

$$\varepsilon(t) = \frac{1}{C_\beta \Gamma(\beta+1)} \int_0^t (t-\tau)^{\beta} \dot{\sigma}(\tau) d\tau = \frac{1}{C_\beta} \left(I_{0^+}^{\beta} \sigma \right)(t) \tag{3b}$$

in terms of the Caputo fractional derivative and Riemann-Liouville fractional integral, respectively.

Use of power-laws and, as a consequence, of fractional-order operators is usually referred, in a rheological context [27], to the introduction of the springpot element.

Springpot is a one-dimensional element defined in terms of two parameters, i.e. C_β and β, $0 \leq \beta < 1$ and $C_\beta > 0$ whose constitutive relation is reported in eqs.(3a,b). Such element with an intermediate behavior among elastic springs and viscous dashpots, is widely used nowadays to define several types of materials and as limiting cases, elastic ($\beta = 0$) and viscous elements ($\beta = 1$) may be obtained. More precisely, a simple spring corresponds to $\beta = 0$ and $\dfrac{d^\beta f}{dt^\beta} = \dfrac{d^0 f}{dt^0} = f$; whilst, case of $\beta = 1$ corresponds to a first order derivative, i.e. $\dfrac{d^\beta f}{dt^\beta} = \dfrac{df}{dt} = \dot{f}$, which is a Newtonian dashpot.

2.2. Constitutive relation for isotropic power-law hereditariness

The extension of the constitutive relation presented in sec.2.1 to case and tensorial strain/stress state is discussed in this section by means of effect superposition.

Let us consider a 2nd-order stress tensor $\boldsymbol{\sigma}$ with component σ_{ij} represented in fig.(4) are with the respective symmetries namely $\sigma_{ij} = \sigma_{ji}$ for $i \neq j$.

In the following we introduce the Voigt representation of the state variables of the material in terms of vector representation of stress and strains tensors as:

$$\boldsymbol{\sigma}^T(t) = [\sigma_{11}(t)\, \sigma_{22}(t)\, \sigma_{33}(t)\, \sigma_{32}(t)\, \sigma_{31}(t)\, \sigma_{12}(t)] \tag{4}$$

$$\boldsymbol{\varepsilon}^T(t) = [\varepsilon_{11}(t)\, \varepsilon_{22}(t)\, \varepsilon_{33}(t)\, 2\varepsilon_{32}(t)\, 2\varepsilon_{31}(t)\, 2\varepsilon_{12}(t)] \tag{5}$$

where t is the current time and the mixed index stress and strain components, namely $\sigma_{ij}(t)$ and $\varepsilon_{ij}(t)$ with $i \neq j$ denote shear stress and strain, respectively. Let us assume that $\sigma_{ij}(t) = \delta_{ij}$ with δ_{ij} the Kronecker tensor $\delta_{ij} = 1$ for $i = j$, $\delta_{ij} = 0$, for $i \neq j$ and let us consider a single normal stress $\sigma_{ii} = 1$ for (i = 1,2,3) reported in fig.4 a,b,c):

In such a context the evolution of the strain $\varepsilon_{ii}(t)$ along the stress direction $\sigma_{ii}(t)$ and in the orthogonal planes reads:

$$\varepsilon_{ii}(t) = J_L(t)\, \sigma_{ii} = J_L(t) \tag{6a}$$

$$\varepsilon_{kk}(t) = \varepsilon_{jj}(t) = -J_v(t)\, \sigma_{ii} \tag{6b}$$

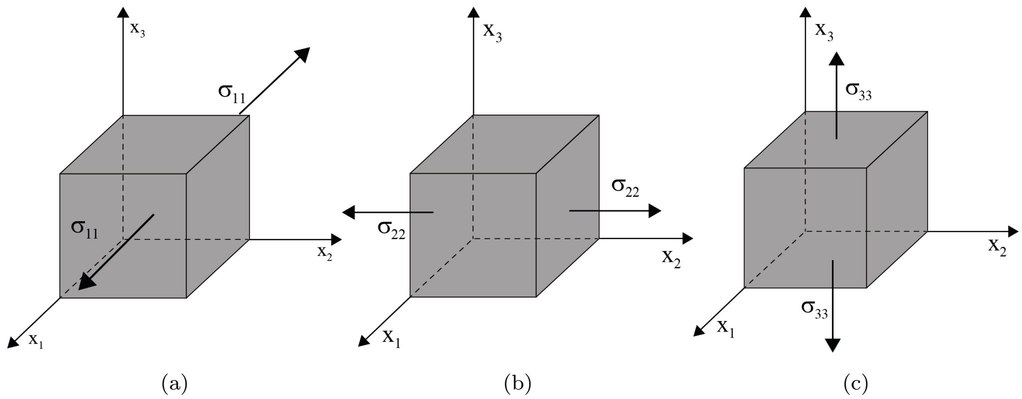


Figure 4: elementary representative cube

with $i \neq j \neq k$ and $i,\ j,\ k = 1, 2, 3$.

In eqs.(6a-b) the function of $J_L\,(t)$ and $J_\upsilon\,(t)$ are the axial and the transverse creep functions with respect to the stress direction, respectively. Under the assumption of smooth load process $\sigma_{ij}(t)$ the presence of contemporaneous stress $\sigma_{ij}(t) = \sigma_{ij}(t)\delta_{ij}$, with $i = 1, 2, 3$, may be account for by the integral.

$$\varepsilon_{ii}\,(t) = \int_0^t J_L\,(t-\tau)\,\dot{\sigma}_{ii}\,(\tau) - J_\upsilon\,(t-\tau)\,[\dot{\sigma}_{jj}\,(\tau) + \dot{\sigma}_{kk}\,(\tau)]\,d\tau \qquad (7)$$

with $i \neq j \neq k$ and i,j,k=1,2,3, respectively.

In the context of material isotropy shear stains $2\varepsilon_{ij}(t)$, $(i \neq j)$, are not involved by axial stress $\sigma_{ii}\,(t)$, but only by shear stress as $\sigma_{ij}(t)$ with $i \neq j$. The evolution of the shear strain $2\varepsilon_{ij}(t)$ due to a generic shear stress history $\sigma_{ij}(t)$ may be obtained by superposition integrals by means of the shear creep function $J_T(\cdot)$ as:

$$2\varepsilon_{ij}\,(t) = \int_0^t J_T\,(t-\tau)\dot{\sigma}_{ij}\,(\tau)\,d\tau \qquad (8)$$

with $i \neq j$ and $i, j = 1, 2, 3$. The constitutive equations reported in eqs.(7),(8) may be reported in Voigt' notation as:

$$\boldsymbol{\varepsilon}\,(t) = \int_0^t \mathbf{J}\,(t-\tau)\dot{\boldsymbol{\sigma}}\,(\tau)\,d\tau \qquad (9)$$

where $\mathbf{J}(t)$ is creep functions matrix that is described as:

$$\mathbf{J}\left(t\right)=\left[\begin{array}{cc}\mathbf{J}^{(A)}\left(t\right) & \mathbf{0}\\ \mathbf{0} & \mathbf{J}^{(T)}\left(t\right)\end{array}\right] \tag{10}$$

where the elements of the axial creep matrix $\mathbf{J}^{(A)}(t)$ are:

$$J_{ij}^{(A)}\left(t\right)=J_L\left(t\right)\delta_{ij}-\left(1-\delta_{ij}\right)J_\upsilon\left(t\right) \tag{11}$$

with $i,j=1,2,3$. The shear creep matrix $\mathbf{J}^{(T)}(t)$ is a diagonal matrix gathering the shear creep functions $J_T\left(t\right)$ as:

$$J_{ij}^{(T)}\left(t\right)=J_T\left(t\right)\delta_{ij} \tag{12}$$

The three creep functions $J_L(t)$,$J_\upsilon(t)$ and $J_T(t)$ are related by a linear relation that reads:

$$J_T(t)=2J_L(t)-J_\upsilon(t) \tag{13}$$

that may be obtained with straightforward manipulations introducing a shear stress state $\sigma_{ij}(t)$ that involves a shear strain state under isotropy assumption, namely $\gamma_{ij}=2\varepsilon_{ij}\left(t\right)$ and evaluating the elongation and the stress along the principal axes at angles of $\pi/4$.

Under the assumption of linear elasticity the creep functions coincides with the material compliance that reads $J_T=1/G$, $J_L=1/E$ and $J_\upsilon=\upsilon/E$ yielding, after substitution in eq.(13):

$$\frac{1}{G}=2\left(\frac{1}{E}+\frac{\upsilon}{E}\right)=\frac{2\left(1+\upsilon\right)}{E} \tag{14}$$

that is the well-known relation among elasticity moduli.

The knowledge of the creep function matrix, namely, $\mathbf{J}(t)$ in eq.(10) allows for the definition of the relaxation matrix $\mathbf{G}(t)$ by means of the coniugation relation as:

$$\hat{\mathbf{G}}(s)\hat{\mathbf{J}}(s)=\frac{1}{s^2}\mathbf{I} \tag{15}$$

where $\mathbf{I}$ is the identity matrix and $\hat{\boldsymbol{G}}(s)$, $\hat{\boldsymbol{J}}(s)$ are the Laplace transforms of the relaxation $\mathbf{G}(t)$ and the creep functions $\mathbf{J}(t)$ matrices.

Straightforward manipulations of eq.(15) and inverse Laplace transform the relaxation matrix may be written as:

$$\mathbf{G}\left(t\right)=\left[\begin{array}{cc}\mathbf{G}^{(A)}\left(t\right) & \mathbf{0}\\ \mathbf{0} & \mathbf{G}^{(T)}\left(t\right)\end{array}\right] \tag{16}$$

where:

$$G_{ij}^{(A)}\left(t\right)=\mathcal{L}^{-1}\left[\frac{1}{s^2\left(\hat{J}_L+\hat{J}_v\right)\left(\hat{J}_L-2\hat{J}_v\right)}\right]\left[\left(\hat{J}_L-\hat{J}_v\right)\delta_{ij}+\left(1-\delta_{ij}\right)\hat{J}_v\right] \tag{17a}$$

$$G_{ij}^{(T)}\left(t\right)=\mathcal{L}^{-1}\left[\frac{1}{s^2\left(\hat{J}_L+\hat{J}_v\right)}\right]\delta_{ij} \tag{17b}$$

The observation of eqs.(17a),(17b) shows that in presence of material fading memory the relaxation matrix $\mathbf{G}(t)$ is obtained in terms of a combination of creep functions obtained by uniaxial creep tests. Similar considerations may be also withdrawn from the observation that in uniaxial relaxation tests, the relaxation functions $G_L(t)$ is obtained in lateral free conditions, that is the strain state involves $\varepsilon_{11}\neq\varepsilon_{22}\neq 0$ and $\varepsilon_{33}=1$ and measuring only $\sigma_{33}(t)=G_L\left(t\right)$ relaxation with $\sigma_{11}=\sigma_{22}=0$.

The knowledge of the relaxation matrix of the material $\mathbf{G}(\mathbf{t})$ allow to evaluate the stress vector as:

$$\sigma\left(t\right)=\int_0^t\mathbf{G}\left(t-\tau\right)\dot{\varepsilon}\left(\tau\right)d\tau \tag{18}$$

The shear longitudinal and transverse relaxation functions namely $G_T(t)$,$G_L(t)$ and $G_v(t)$ are related by a linear equation similar to that involving creep functions in eq.(13) that reads:

$$G_T\left(t\right)=\frac{1}{2}\left(G_L\left(t\right)-G_v\left(t\right)\right) \tag{19}$$

allowing for the evaluation of the transverse relaxation $G_v(t)$ as:

$$G_v\left(t\right)=2G_T\left(t\right)-G_v\left(t\right) \tag{20}$$

In the following section the thermodynamic restrictions among the material parameters used in power-law representation of isotropic material hereditariness are outlined.

2.3. Power-law isotropic hereditariness: Thermodynamic restrictions

Let us assume that relaxation functions in laterally restrained axial and torsion shear tests, respectively, may be captured by power-laws with different order $(\alpha \neq \beta)$ as:

$$G_L\left(t\right) = G_L^{(\alpha)} t^{-\alpha} + \bar{G}_L; \quad G_T\left(t\right) = G_T^{(\beta)} t^{-\beta} + \bar{G}_T \tag{21a}$$

$$G_v\left(t\right) = 2\left({G_T}^{(\beta)} t^{-\beta} + \bar{G}_T\right) - \left({G_L}^{(\alpha)} t^{-\alpha} + \bar{G}_L\right) \tag{21b}$$

with eq.(21b) obtained from the application of eq.(16). Physical dimensions of the coefficients are $[C_L] = [C_T] = F/L^2$, $\left[C_L^{(\alpha)}\right] = \dfrac{F}{L^2 T^{-\alpha}}$, $\left[C_L^{(\alpha)}\right] = \dfrac{F}{L^2 T^{-\beta}}$.

Expression of the relaxation functions in eqs.(21a),(21b) yields the relaxation matrix of the material in eq.(16) with elements in the block matrices $\mathbf{G^{(A)}}\left(\mathbf{t}\right)$ and $\mathbf{G^{(T)}}\left(\mathbf{t}\right)$ reads:

$$G_{ij}^{(A)} = G_L\left(t\right)\delta_{ij} + \left(1 - \delta_{ij}\right) G_v\left(t\right) \tag{22a}$$

$$G_{ij}^{(T)}\left(t\right) = G_T\left(t\right)\delta_{ij} \tag{22b}$$

The observation of eqs.(22a), (22b) shows that the relaxation matrix involves elements decaying with different power-law order β and α $(\alpha, \beta \in [0, 1])$ as the functional classes in eqs. (21a), (21b) are replaced in eqs.(22a), (22b).

Coefficients and parameters involved in the power-law descriptions of the material relaxation, namely, $G_L(t)$, $G_v(t)$ and $G_T(t)$ are related by thermodynamic restrictions to ensure the requirement of positive entropy rate increment [28]. Indeed, a dissipative simple solid is defined only if the restrictions:

$$\mathbf{G}\left(0\right) \geq \mathbf{G}\left(\infty\right) \geq 0 \tag{23}$$

$$\dot{\mathbf{G}}\left(0\right) \geq 0 \tag{24}$$

are fulfilled by the relaxation matrix of the material as reported in basic references on material hereditariness [29, 30, 31, 32].

Eqs.(24,25) are always satisfied assuming positive values of coefficients $\bar{G}_L$, $\bar{G}_T$ and $G_L^{(\alpha)}$ and $G_T^{(\beta)}$, whereas the latter eq.(25) is satisfied, only, as the eigenvalues of the first derivative of the matrix, namely, $\dot{\mathbf{G}}(0)$ are all negative. This requirement may be verified introducing a one-parameter family of relaxation matrices defined on a real parameter δ as $\mathbf{G}_\delta(t) = \mathbf{G}(t+\delta)$ and investigating the behavior of the matrix family $\dot{\mathbf{G}}_\delta(t)$ for limiting case of the parameter $\delta \to 0$.

The parameter-dependent matrix family $\dot{\mathbf{G}}_\delta(t)$ is defined as:

$$\dot{\mathbf{G}}_\delta(t) = \begin{bmatrix} \dot{\mathbf{G}}_\delta^{(A)}(t+\delta) & \mathbf{0} \\ \mathbf{0} & \dot{\mathbf{G}}_\delta^{(T)}(t+\delta) \end{bmatrix} \tag{25}$$

where the elements of the matrix reads:

$$\dot{G}_\delta^{(A)}(t+\delta) = -G_L^{(\alpha)}\alpha(t+\delta)^{-\alpha-1} \tag{26a}$$

$$\dot{G}_\delta^{(T)}(t+\delta) = -G_T^{(\beta)}\beta(t+\delta)^{-\beta-1} \tag{26b}$$

$$\dot{G}_\delta^{(\upsilon)}(t+\delta) = -2G_T^{(\beta)}\beta(t+\delta)^{-\beta-1} + G_L^{(\alpha)}\alpha(t+\delta)^{-(\alpha+1)} \tag{26c}$$

Observing that the one-parameter family $\dot{\mathbf{G}}_\delta(t)$ leads, to the limit to:

$$\lim_{\delta\to 0} \dot{\mathbf{G}}_\delta(t) = \dot{\mathbf{G}}(t) \tag{27}$$

We can infer the behavior of $\dot{\mathbf{G}}(t)$ from those of the family $\mathbf{G}_\delta(t)$ and letting $\delta \to 0$. In this regard, the requirement in eq.(25) may be obtained as:

$$-\dot{G}(0) = -\lim_{\delta\to 0} \dot{G}_\delta(t) \geq 0 \tag{28}$$

that is we evaluate the eigenvalues $\lambda_i(\delta)$ $(i = 1, 2, ...6)$ of the matrix $\dot{\mathbf{G}}_\delta(0)$ and with the additional restraint $-\lambda_i(\delta) \geq 0$ $(i = 1, 2, ...6)$ as $\delta \to 0$.

The evaluation of the eigenvalues $\lambda_i(\delta)$ reads:

$$-\lambda_1(\delta) = -\lambda_2(\delta) = -2\left(\dot{G}_L(\delta) - \dot{G}_T(\delta)\right) \geq 0 \tag{29a}$$

$$-\lambda_3(\delta) = -\lambda_4(\delta) = -\lambda_5(\delta) = -\dot{G}_T(\delta) \geq 0 \tag{29b}$$

$$-\lambda_6(\delta) = -4\dot{G}_T(\delta) + \dot{G}_L(\delta) \geq 0 \tag{29c}$$

Substitution of eq.(26a),(26b) into eq.(29b) shows that the inequality is fulfilled for $C_T^{(\beta)} \geq 0$ and $0 \leq \beta \leq 1$. Inequalities in eqs.(29a),(29c) read, after substitution:

$$\alpha G_\alpha \delta^{-(\alpha+1)} - \beta G_\beta \delta^{-(\beta+1)} \geq 0 \tag{30a}$$

$$4\beta G_\beta \delta^{-(\beta+1)} - \alpha G_\alpha \delta^{-(\alpha+1)} \geq 0 \tag{30b}$$

that, after some straightforward manipulation, may be cast in a more suitable form, taking natural logarithms as:

$$\ln(A_{\alpha\beta}) \geq (\alpha - \beta)\ln\delta \tag{31a}$$

$$\ln\left[\frac{(A_{\alpha\beta})}{4}\right] \leq (\alpha - \beta)\ln\delta \tag{31b}$$

where $A_{\alpha\beta} = \alpha G_L^{(\alpha)} / \left(\beta G_T^{(\beta)}\right)$. Inequalities in eqs.(30a)(30b) must be fulfilled for any value of the parameter δ yielding that $\alpha = \beta$. Moreover, in this latter case the additional thermodynamical restriction holds true.

$$G_T^{(\beta)} \leq C_L^{(\beta)} \leq 3C_T^{(\beta)} \tag{32}$$

In passing we observe that the condition $\alpha = \beta$ holds true only for the two terms (or one terms) description of the relaxation function in eq.(22a). Indeed, as we assume that the relaxation functions $G_L(t)$ and $G_T(t)$ involve linear combinations of power-laws as:

$$G_L(t) = \sum_{j=1}^{n} G_L^{(\alpha_j)} t^{-\alpha_j}; G_T(t) = \sum_{i=1}^{m} G_T^{(\beta_i)} t^{-\beta_i} \tag{33}$$

with n and m the number of power-laws involved. Under such circumstances thermodynamical arguments proposed this study yield proper same conditions among the order of the power-laws as:

$$\max_{j=1,N}(\alpha_j) = \max_{i=1,M}(\beta_j) \tag{34a}$$

$$\min_{j=1,N}(\alpha_j) = \min_{i=1,M}(\beta_j) \tag{34b}$$

The use of eq.(22a),(22b) substituted into the constitutive equations for the three-axial hereditariness yields a relation among the stress vector and history of the strain vector $\boldsymbol{\varepsilon}(t)$ as:

$$\boldsymbol{\sigma}(t) = \mathbf{G}_\beta \int_0^t (t-\tau)^{-\beta} \dot{\boldsymbol{\varepsilon}}(\tau)\, d\tau + \bar{\mathbf{G}} = \mathbf{G}_\beta \left(D_{0^+}^{\beta} \boldsymbol{\varepsilon}\right)(t) + \bar{\mathbf{G}} \tag{35}$$

where we assumed the Voigt representation of the relaxation tensor $\mathbf{G}(\mathbf{t})$ in matrix form and we used the notation:

$$\mathbf{G}(t) = \mathbf{G}_\beta \frac{t^{-\beta}}{\Gamma(1-\beta)} + \bar{\mathbf{G}} \tag{36}$$

with the matrices:

$$\mathbf{G}_\beta(t) = \begin{bmatrix} G_\beta^{(L)} & G_\beta^{(v)} & G_\beta^{(v)} & 0 & 0 & 0 \\ G_\beta^{(v)} & G_\beta^{(L)} & G_\beta^{(v)} & 0 & 0 & 0 \\ G_\beta^{(v)} & G_\beta^{(v)} & G_\beta^{(L)} & 0 & 0 & 0 \\ 0 & 0 & 0 & G_\beta^{(T)} & 0 & 0 \\ 0 & 0 & 0 & 0 & G_\beta^{(T)} & 0 \\ 0 & 0 & 0 & 0 & 0 & G_\beta^{(T)} \end{bmatrix} \tag{37a}$$

$$\overline{\mathbf{G}} = \begin{bmatrix} \bar{G}_L & \bar{G}_v & \bar{G}_v & 0 & 0 & 0 \\ \bar{G}_v & \bar{G}_L & \bar{G}_v & 0 & 0 & 0 \\ \bar{G}_v & \bar{G}_v & \bar{G}_L & 0 & 0 & 0 \\ 0 & 0 & 0 & \bar{G}_T & 0 & 0 \\ 0 & 0 & 0 & 0 & \bar{G}_T & 0 \\ 0 & 0 & 0 & 0 & 0 & \bar{G}_T \end{bmatrix} \tag{37b}$$

The stress vector obtained as functional of the strain vector $\boldsymbol{\varepsilon}(t)$ in eq.(35) is the generalization of the constitutive equation reported in eq.(3a) under the assumption of material isotropy.

In the next section the multiaxial fractional-order hereditariness will be further discussed introducing a mechanical hierarchy that yields the constitutive model reported in eq.(35)

3. Exact mechanical description of fractional-order isotropic hereditariness

The stress/strain tensor outlined in sec.(2) requires a multiaxial constitutive relation, as in eq.(35), that under the assumption of $\bar{\mathbf{G}} = 0$ generalizes eq.(3a). The rheological element, namely the springpot, corresponding to eq.(3a) can not, however, be defined also for the isotropic description in sec.(2), namely for the presence of shear stress/strain. A mechanical model that may be involved in presence of normal and shear stress to be used in experimental test is represented in fig.5

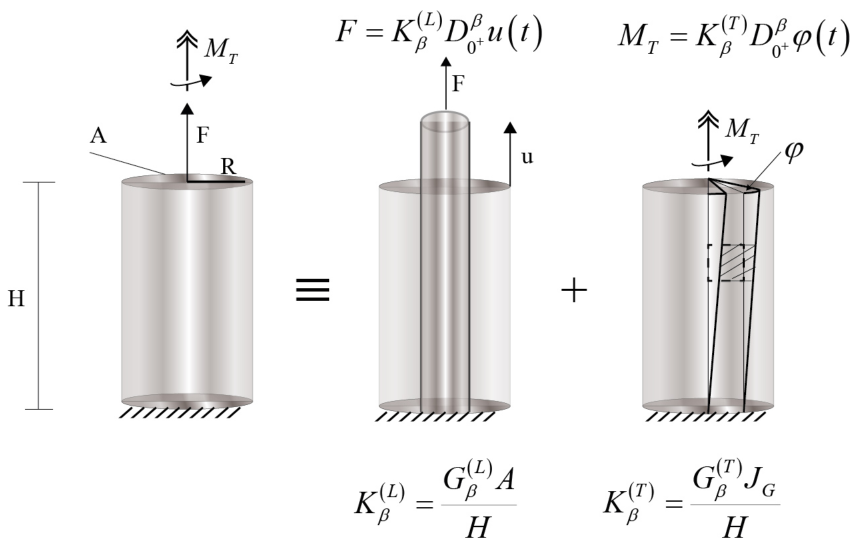


Figure 5: Rheologic elements

In such condition, the circular column of length, cross section A and radius R under axial stress and shear stress related to the measured relative displacements

$u(t)$ and twist angle $\varphi(t)$ as:

$$\begin{aligned} F &= K_\beta^{(L)} (D_{0+}^\beta u)(t) \\ M_T &= K_\beta^{(T)} (D_{0+}^\beta \varphi)(t) \end{aligned} \tag{38}$$

where $A = \pi R^2$ and $J_G = \pi R^4/4$ are the cross section and the polar moment of inertia of the circular cross-section represented in fig.5. The constitutive equations in eq.(38) involves respectively for limiting cases: i) a linear elastic spring ($\beta = 0$) ; ii) a linear viscous element ($\beta = 1$).

In the following we introduce a hierarchic mechanical model to capture the axial and shear hereditariness assuming power-law description of the creep and relaxation functions for axial and shear stress/strain, respectively [33, 17, 34, 16]. The obtained mechanical hierarchy corresponds exactly to an axial and shear springpots with the same order of time evolution/decay.

To this aim let us introduce an elastic column of unbounded length with circular cross section of radius R. The elastic features of the column are non-costant along the column axis and vary with the coordinate as:

$$E(z) = \frac{E_\alpha}{\Gamma(1-\alpha)} z^{-\alpha}; \quad G(z) = \frac{G_\alpha}{\Gamma(1-\alpha)} z^{-\alpha} \quad -1 \leq \alpha \leq 1 \tag{39}$$

The column is externally restrained by a set of torsional and axial viscous dashpots fig.(5) with non-homogeneous viscosity $\eta(z)$ as:

$$\eta(z) = \frac{\eta_\alpha}{\Gamma(1+\alpha)} z^{-\alpha} \quad -1 \leq \alpha \leq 1 \tag{40}$$

Axial and torsional equilibrium along the column axis reads:

$$\frac{\eta_\alpha}{\Gamma(1+\alpha)} z^{-\alpha} 2\pi R \Delta z \dot{u}(z,t) = \frac{E_\alpha \pi R^2 s (z+\Delta z)^{-\alpha}}{\Gamma(1-\alpha)} \left[u(z+\Delta z,t) - u(z,t)\right] + \\ + \frac{E_\alpha \pi R^2 s z^{-\alpha}}{\Gamma(1-\alpha)} \left[u(z,t) - u(z-\Delta z,t)\right] \tag{41}$$

$$\frac{\eta_\alpha}{\Gamma(1+\alpha)} z^{-\alpha} 2\pi R^2 \Delta z \dot{\varphi}(z,t) = G_\alpha \pi R^4 (z+\Delta z)^{-\alpha} \left[\varphi(z+\Delta z,t) - \varphi(z,t)\right] + \\ + G_\alpha \pi R^4 (z+\Delta z)^{-\alpha} \left[\varphi(z,t) - \varphi(z-\Delta z,t)\right] \tag{42}$$

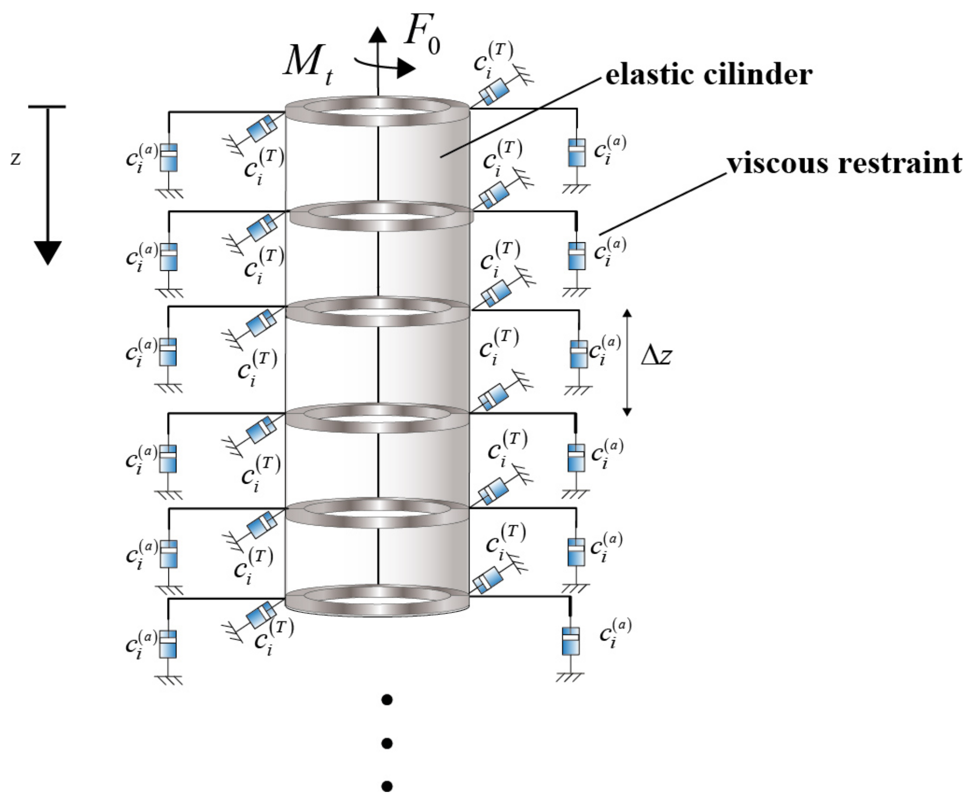


Figure 6: column with non-homogeneous viscosity

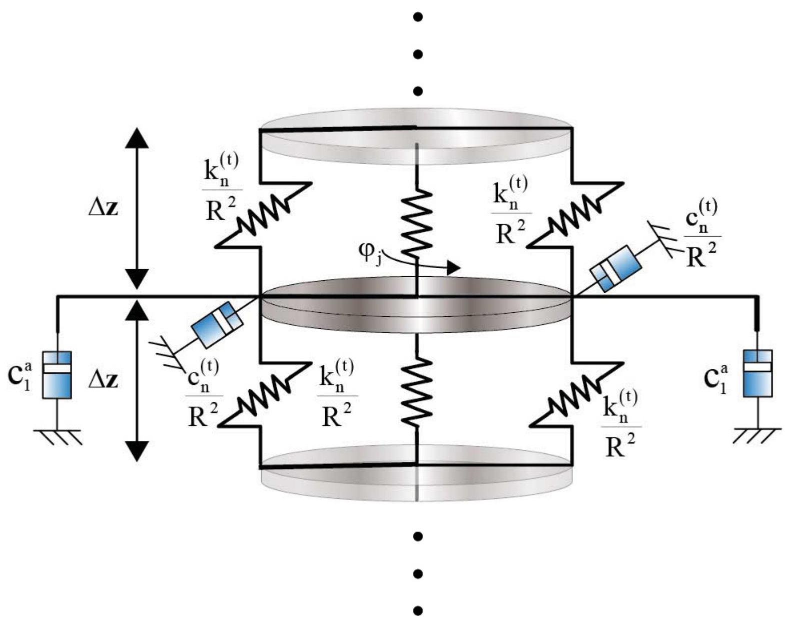


Figure 7: elements of the column with non-homogeneous viscosity

that can be rewritten in differential form, letting $\Delta z \to 0$ as:

$$\frac{\eta_\alpha z^{-\alpha}}{\Gamma(1+\alpha)}\frac{\partial u(z,t)}{\partial t} = \frac{E_\alpha R s}{\Gamma(1-\alpha)}\frac{\partial}{\partial z}\left(z^{-\alpha}\frac{\partial u(z,t)}{\partial z}\right) \tag{43a}$$

$$\frac{\eta_\alpha z^{-\alpha}}{\Gamma(1+\alpha)}\frac{\partial \varphi(z,t)}{\partial t} = \frac{G_\alpha R}{\Gamma(1-\alpha)}\frac{\partial}{\partial z}\left(z^{-\alpha}\frac{\partial \varphi(z,t)}{\partial z}\right) \tag{43b}$$

Boundary conditions involving the differential fields $u(z,t)$ and $\varphi(z,t)$ in eqs.(43a),(43b) read, respectively.

$$\lim_{z\to\infty} u(z,t) = 0 \tag{44a}$$

$$\lim_{z\to 0} \frac{E_\alpha}{\Gamma(1-\alpha)} z^{-\alpha}\frac{\partial u}{\partial z} = F_0 \tag{44b}$$

$$\lim_{z\to\infty} \varphi(z,t) = 0 \tag{45a}$$

$$\lim_{z\to 0} \frac{G_\alpha}{\Gamma(1-\alpha)} z^{-\alpha}\frac{\partial \varphi}{\partial z} = M_0 \tag{45b}$$

Mathematical operators and boundary conditions in eqs.(46a,b) are completely equivalent to those of a previous differential problem that has been solved resorting to a non- linear mapping followed by Laplace transform [17, 35]. Such a procedure yields a Bessel differential equation of second kind in terms of the anomalous Laplace parameters. Position of the boundary conditions and inverse Laplace transform provides solution in the form:

$$u_0(t) = u_0(z,t) = \lim_{z\to\infty} u(z,t) = \frac{t^{-\beta}}{k_\beta^{(L)}} F_0 = J_L(t) F_0 \tag{46}$$

$$\varphi_0(t) = \varphi_0(z,t) = \lim_{z\to\infty} \varphi(z,t) = \frac{t^{-\beta}}{k_\beta^{(T)}} M_0 = J_T(t)_0 \tag{47}$$

with:

$$k_\beta^{(L)} = \frac{\Gamma(2\beta)\left(\tau_L^\beta\right)}{E_\alpha 2^{1-2\beta}\Gamma(\beta)\Gamma(1-\beta)} \tag{48}$$

$$k_\beta^{(T)} = \frac{\Gamma(2\beta)\left(\tau_L^\beta\right)}{G_\alpha 2^{1-2\beta}\Gamma(\beta)\Gamma(1-\beta)} \tag{49}$$

with $\beta = \frac{1+\alpha}{2}$ and the relaxation times:

$$\tau_L = \frac{\eta_\alpha}{E_\alpha}\frac{\Gamma(1-\alpha)}{\Gamma(1+\alpha)} \tag{50}$$

$$\tau_T = \frac{\eta_\alpha}{G_\alpha}\frac{\Gamma(1-\alpha)}{\Gamma(1+\alpha)} \tag{51}$$

Effect superpositions provides, resorting to the fundamental equations of linear viscoelasticity, the constitutive equations of the macroscopic variables as:

$$F_0(t) = k_\beta^{(L)}\left(D_{0+}^\beta u_0\right)(t) \tag{52}$$

$$M_T(t) = k_\beta^{(T)}\left(D_{0+}^\beta \varphi_0\right)(t) \tag{53}$$

Eqs.(52),(53) are the constitutive equation at the macro-scale and, recalling that $F_0 = \sigma_{33}A$ and $|\tau| = \sqrt{|t_{31}|^2 + |t_{32}|^2} = \frac{M_0}{2As}$ the constitutive equation of the material reads:

$$\sigma_{33} = G_\beta^{(L)}\left(D^\beta \varepsilon_{33}\right)(t) \tag{54}$$

$$|\tau| = G_\beta^{(T)}\left(D^\beta |\gamma|\right)(t) \tag{55}$$

with the coefficients $G_\beta^{(L)}$ and $G_\beta^{(T)}$ that read:

$$G_\beta^{(L)} = \frac{\bar{k}_\beta^{(L)}\bar{l}}{A} \qquad G_\beta^{(T)} = \frac{\bar{k}_\beta^{(T)}}{2As}\frac{R}{\bar{l}} \tag{56}$$

and $\bar{l}$ an internal length of the material. Eqs.(54),(55) the multiaxial constitutive relations of the isotropic material and, henceforth, they proposed hierarchy correspondent to the fractional-order isotropy introduce in the paper.

4. Conclusions

The mathematical structure of the fractional-order isotropic hereditariness has been discussed in the paper. The study has been framed in the context of biomimetic ceramics used in cranioplasty neurosurgery (i.e. CustomBone® "prosthesis"). The creep and relaxation functions of isotropic linear hereditarinnes have been particularized for power-law decays yielding a multi-axial constitutive model in terms of fractional-order operators. Additionally a specific mechanical model has been introduced that correspond to the fractional-order isotropic hereditariness. In future studies experimental campaigns involving creep and relaxation of biomimetic ceramics will be reported to assess the validity of material isotropy. Additionally, the proposed hierarchy will be further extended to deal with non-linear hereditariness as those observed in creep and relaxations of tendons and ligaments.

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Power-Laws hereditariness of biomimetic ceramics for cranioplasty neurosurgery

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Abstract

In this paper the authors deal with the hereditary behavior of hydroxyapatite-based composites used for cranioplastic surgery. It is shown that biomimetic prosthesis, possess an isotropic fractional-order material hereditariness due to their microstructure architecture. The three-axial hereditariness is framed in the context of fractional-calculus providing details about thermodynamical restrictions of memory functions used in the formulation. A mechanical model that corresponds, exactly, to the three-axial fractional-order hereditariness is also introduced in the paper.

Keywords: `Biomimetic materials,cranioplasty, fractional calculus, power-law hereditariness, isotropic hereditariness.`

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1. Introduction

The cranioplastic neurosurgery is nowadays an important issue worldwide since it is necessary both in traumatic therapies or in presence of specific oncologic

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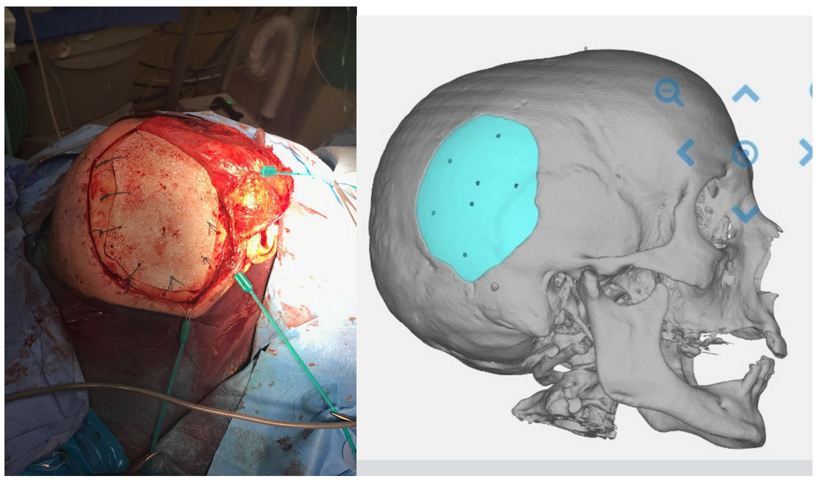
pathology. Cranioplasty is a surgical procedure that aims to re-establish the skull integrity following a previous craniotomy due to the occurrence of traumas, tumors and/or congenital malformations. In all cases cranioplasty can be considered as the conclusive action of a surgery initiated by the removal of a bone operculum fig.1.



(a) (b)

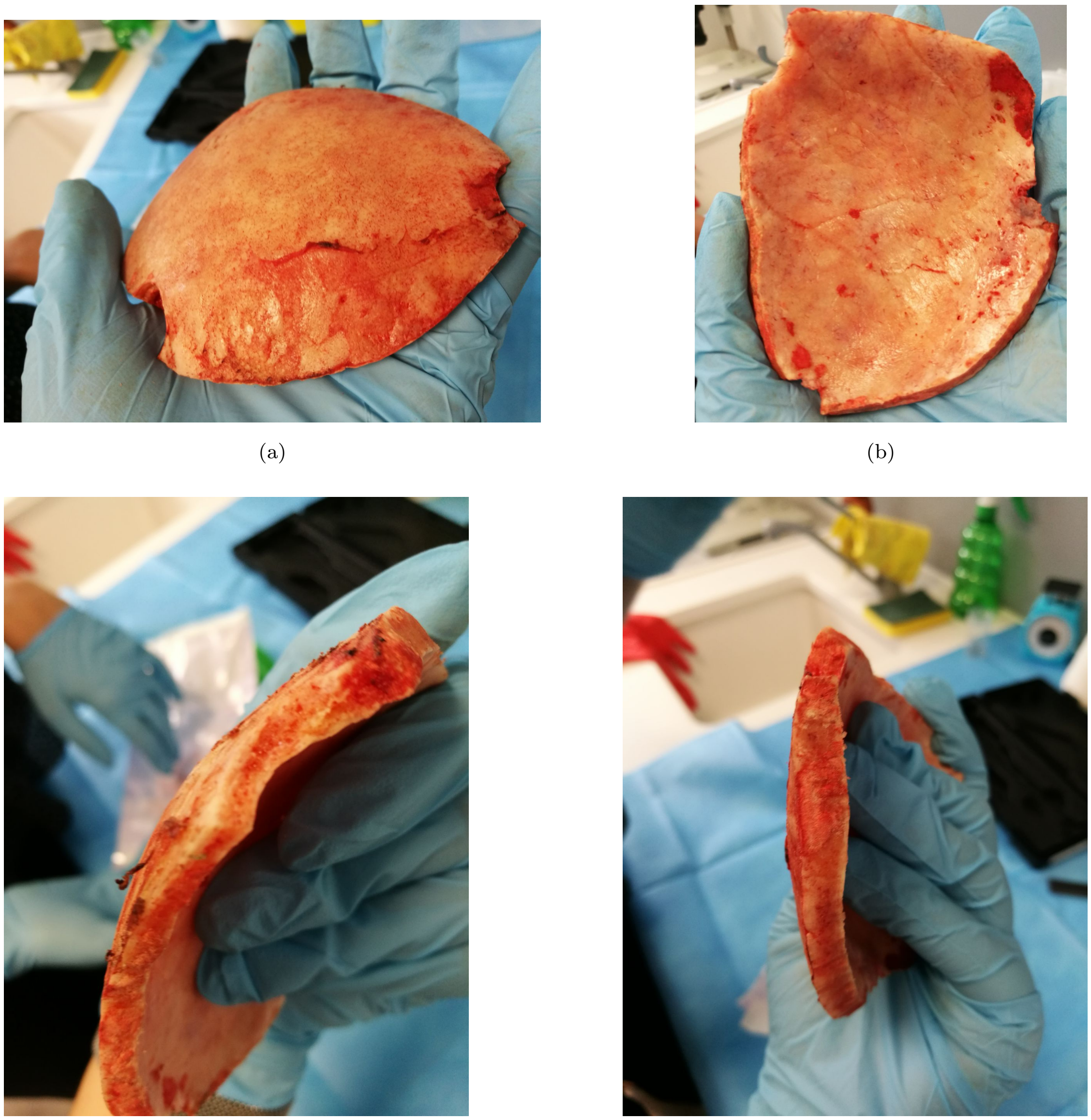
Figure 1: cranioplasty surgery

Ideally, cranioplasty procedures should provide restoration of the protective functions of the skull with maintenance of the original aesthetics and long-term mechanical performance [1] The ideal material for cranioplasty should be chemically inert, biocompatible, biomechanically reliable, easily manufactured, individually shaped, safe, and able to promote osteoblast migration. Today synthetic implants based on metallic (mainly titanium) or acrylic plaques (mainly polymethylmeta-crylate or polyetheretherketone) are widely used in cranioplasty procedures. These are bioinert materials with good biocompatibility, resistance to infections, ease of sterilization, ability to be subjected to imaging diagnostics, and the capacity to undergo flexible design for adaptation to different clinical cases. They exhibit good mechanical strength, which offers adequate brain protection from external shocks. However, they present poor osteogenic and osteoconductive ability, thus resulting in a foreign body functioning as a shell expected to provide brain protection, but connected to the surrounding bone only by its perimeter contact surface. In order to overcome many limitations an Hydroxyapatite (HA)-based material has been widely considered for decades as the gold standard for bone scaffolds, as its composition is very close to that of bone mineral, thus exhibiting excellent biocompatibility, a low inflammatory reaction as well as good osteogenic ability and osteoconductivity. The hydrophilic character of HA favors cell attachment and tight adhesion of bone to the scaffold surface, which is a key target for the stability of the bone/implant interface. Therefore, HA scaffolds presenting wide, open and interconnected multiscale porosity can induce extensive bone ingrowth and penetration throughout the whole scaffold, partly thanks to the possibility of massive fluid perfusion, which triggers and assists neovascularization. Hence, cranial reconstruction using synthetic porous HA has recently become the subject of intense debate among surgeons, and it now represents a new concept in cranioplasty procedures. The custom-made concept was first applied to porous hydroxyapatite because of the need to overcome the fragility of the material itself. Among the advantages of HA-based prosthesis there is the important issue of customization.

Indeed, in presence of cranioplasty the morphology of the bone to be replaced with a synthetic prosthesis must match, completely, the original bone to accelerate the osteointegration of the prosthesis [2, 3, 4] in the surgical hole. In fig.(2 a-d) an human parietal bone and its synthetic prosthesis fig.(3 a-c) have been obtained from at universitary neurosurgery hospital in Palermo. The synthetic bone used for replacement is a CustomBone® (Finceramica Faenza), namely custom-made, porous hydroxyapatite scaffolds with total porosity in the range of 60 to 70 % and pore architecture based on macro-pores ($>$ 100 micron) interconnected with micro-pores (5-10 micron). CustomBone® scaffolds were obtained by reproduction of the patients bone defect as modeled by 3D CT scan and its represented as a composite ceramics material obtained from chemical deposition of hydroxyiapatite within a small fraction of collagen type I (see fig.1a).

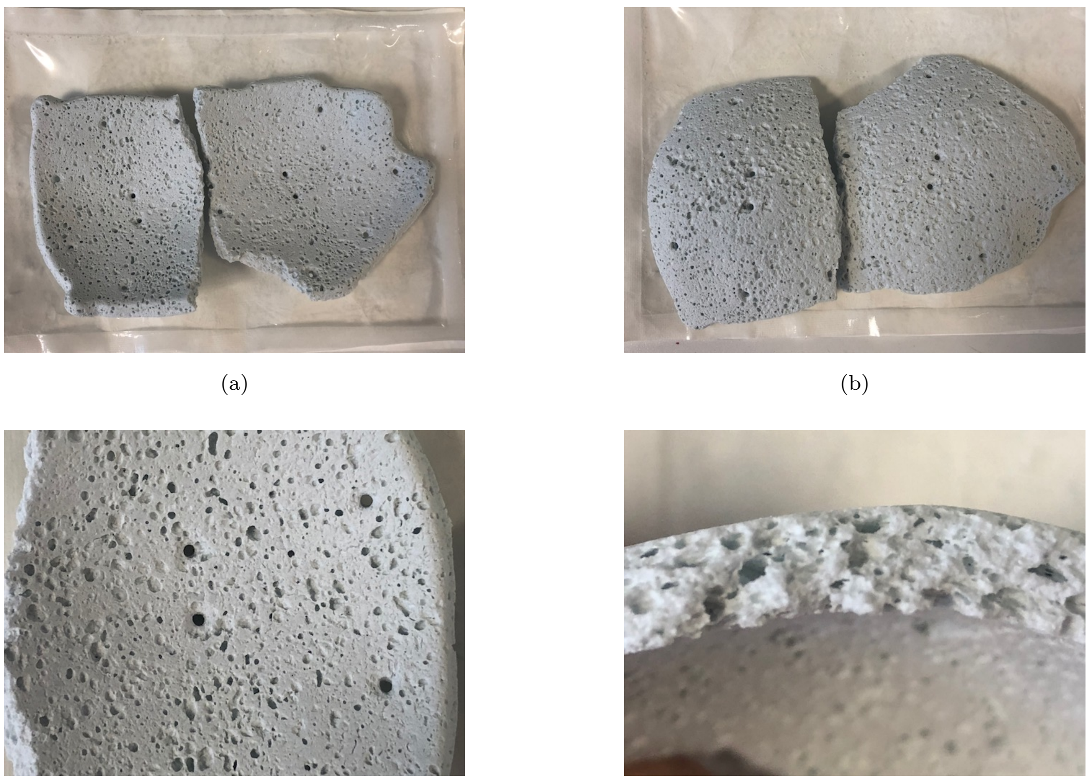
The use of biomimetic ceramics to replace cortical as well as trabecular bone is as well defined to technique in bone surgery [5]. Indeed the mechanical feature of the prosthesis in terms of elastic moduli and the strength of the biomimetic composite of integration are very similar. However, the use of ceramic materials to replace the bones of human head may involve in different behavior in terms of energy dissipation loss. This feature is related to material hereditariness that depends on the movements of the organic collagen chains in the real bone. In such a case additionally long-term stress are applied to the grafted ceramics prosthesis and this may lead to fracture propagation during patient follow up [6]. The mechanical hereditary behavior of the material bone is intrinsically orthotropic, due to the self-organization of the bone tissues into osteons in periostial bone and trabecular in the sponges bone.

The hereditary properties of bone in uniaxial test is to be described by the creep $J(t)$ and the relaxation $G(t)$ functions that are well represented by power-law $J(t) \propto t^{\beta}$ and $G(t) \propto t^{-\beta}$ with $0 \leq \beta \leq 1$ [5, 7, 8, 9]. In presence of multiaxial state of stress different relations for creep and relaxations may be observed. The introduction of power-laws in the description of creep and relaxations yields that the constitutive behavior of the material is expressed in terms of the so-called fractional calculus[10, 11, 12, 13, 14] that is a generalization of the classical differential calculus to real-order integration and differentiation $df/dt \rightarrow d^{\beta}f/dt^{\beta}$ with $\beta \in [0,1]$. In such a context, uniaxial hereditariness involving fractional order stress-strain relations have been reported since the beginning of the twentyth century [15, 12] defining the so-called springpot element [16, 17]. The introduction of the 3D constitutive relation for power-law hereditariness, as those shows by biomimetic prosthesis, has not been, however, sufficiently, investigated in scientific literature. Indeed, in several cases, recently discussed in scientific literature [18] the use of power-laws without thermodynamic restriction on the parameters does not guarantee positive entropy increment for any strain/stress process.



(a) (b) (c) (d)

Figure 2: human parietal bone



(a) (b) (c) (d)

Figure 3: CustomBone® prosthesis morphology

In this paper the authors aim to formulate a thermodynamically consistent three-axial constitutive relations involving power-laws in the context of material isotropy. The proposed constitutive relation discussed in sec. 2.3 will be analyzed in sec.3 providing the exact mechanical description of the three-axial isotropic fractional-order hereditariness. Some conclusions about the proposed model of isotropic hereditariness and the influence on the mechanics of the biomimetic ceramics prosthesis are withdrawn in sec.4

2. Power-laws hereditariness of isotropic biomimetic ceramics

In this section the thee-axial isotropic constitutive relations in presence of power-laws hereditariness is outlined. In sec.2.1 the main arguments of power-law hereditariness under uniaxial condition are shortly outlined. The three-axial isotropic constitutive relation are defined in sec.2.2.

2.1. Uniaxial power-law hereditariness: The fractional order constitutive equation

The constitutive behavior of materials in long-standing mechanical tests is described by means of the well-known creep and relaxation functions, dubbed $J(t)$ and $G(t)$, respectively. The linear superposition applied to a generic stress/strain history, namely $\sigma(\tau)$ and $\varepsilon(\tau)$ with $\tau \leq t$, yields,

$$\sigma(t) = \int_0^t G(t-\tau) d\varepsilon(\tau) = \int_0^t G(t-\tau)\dot{\varepsilon}(\tau) d\tau \tag{1a}$$

$$\varepsilon(t) = \int_0^t J(t-\tau) d\sigma(\tau) = \int_0^t J(t-\tau)\dot{\sigma}(\tau) d\tau \tag{1b}$$

Eqs.(1a, b) are defined in terms of Boltzman superposition, where $\dot{f}(t) = df(t)/dt$ denotes the increment of the generic function $f(t)$ as well as the stress $d\sigma = \dot{\sigma}dt$. and the strain $d\varepsilon = \dot{\varepsilon}dt$ increments, respectively.

Convolution integrals in eqs.(1a, b) are completely described introducing the functional class of creep and relaxation functions an phenomenological based experimental data.

Creep and relaxation functions characterize the material behavior and they must satisfy the conjugation relation $\hat{J}(s)\hat{G}(s) = 1/s^2$, where s indicates the Laplace parameter and $\hat{f}(s) = \mathcal{L}[f(t)]$ the Laplace transform of the generic function $f(t)$.

In the context of materials hereditariness, power-law representation of creep and relaxation functions, i.e. $J(t)$ and $G(t)$, was introduced at the beginning of the last century [15],

$$G(t) = \frac{C_\beta}{\Gamma(1-\beta)} t^{-\beta}, \tag{2a}$$

$$J(t) = \frac{1}{C_\beta \Gamma(\beta+1)} t^{\beta} \tag{2b}$$

where $\Gamma(\cdot)$ is the Euler-Gamma function, $\beta \in [0, 1]$ and C_β, are positive real parameters, that may be estimated through a best-fitting procedure of experimental data [19, 20]. Straightforward manipulations show that the power-law functional class in eqs.(2a, b), satisfies the conjugation relation and it yields, upon substitution in eqs. (1a, b) the constitutive relations:

$$\sigma(t) = \frac{C_\beta}{\Gamma(1-\beta)} \int_0^t (t-\tau)^{-\beta} \dot{\varepsilon}(\tau) d\tau = C_\beta \left(D_{0^+}^{\beta} \varepsilon \right)(t) \tag{3a}$$

$$\varepsilon(t) = \frac{1}{C_\beta \Gamma(\beta+1)} \int_0^t (t-\tau)^{\beta} \dot{\sigma}(\tau) d\tau = \frac{1}{C_\beta \Gamma(\beta)} \int_0^t (t-\tau)^{\beta-1} \sigma(\tau) d\tau =$$
$$= \frac{1}{C_\beta} \left(I_{0^+}^{\beta} \sigma \right)(t) \tag{3b}$$

in terms of the Caputo fractional derivative and Riemann-Liouville fractional integral, respectively.

Use of power-laws and, as a consequence, of fractional-order operators is usually referred, in a rheological context [21], to the introduction of the spring-pot element. Springpot is a one-dimensional element that is defined by means of two parameters, i.e. C_β and β, $0 \leq \beta < 1$ and $C_\beta > 0$ whose constitutive relation is obtained by eqs.(3a,b). More precisely, a simple spring corresponds to $\beta = 0$ and $\frac{d^\beta f}{dt^\beta} = \frac{d^0 f}{dt^0} = f$; whilst, case of $\beta = 1$ corresponds to a first order derivative, i.e. $\frac{d^\beta f}{dt^\beta} = \frac{df}{dt} = \dot{f}$, which is a Newtonian rheological element.

2.2. Multiaxial constitutive relation for isotropic power-law hereditariness

The extension of the constitutive relation presented in sec.2.1 to case of multiaxial state of stress and/or of strain is analyzed here by means of the effects superposition.

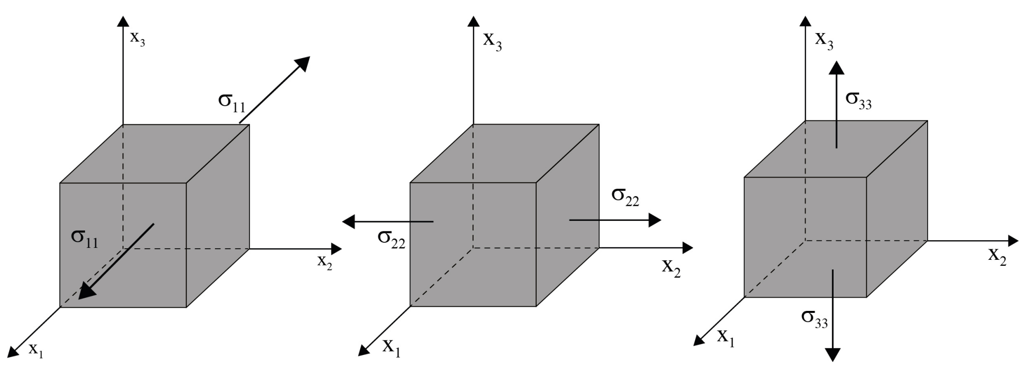
Let us consider a three-dimensional stress tensor $\boldsymbol{\sigma}$ with component σ_{ij} represented in fig.(4) with the respective symmetries namely $\sigma_{ij} = \sigma_{ji}$ for $i \neq j$.

In the following we introduce the Voigt representation of the state coordinates of the material considered that involves vector representation of stress and strains as:

$$\boldsymbol{\sigma}^T(t) = [\sigma_{11}(t)\, \sigma_{22}(t)\, \sigma_{33}(t)\, \sigma_{32}(t)\, \sigma_{31}(t)\, \sigma_{12}(t)] \tag{4}$$

$$\boldsymbol{\varepsilon}^T(t) = [\varepsilon_{11}(t)\, \varepsilon_{22}(t)\, \varepsilon_{33}(t)\, 2\varepsilon_{32}(t)\, 2\varepsilon_{31}(t)\, 2\varepsilon_{12}(t)] \tag{5}$$

where t is the current time and the mixed index stress ans strain components, namely $\sigma_{ij}(t)$ and $\varepsilon_{ij}(t)$ with $i \neq j$ denotes shear stress and strains, respectively. Let us assume that $\sigma_{ij}(t) = \delta_{ij}$ with δ_{ij} the kroneker, and considering one single stress $\sigma_{ii} = 1$ acting (i = 1,2,3) it yields (fig.4 a,b,c):



(a) (b) (c)

Figure 4: elementary representative cube

$$\varepsilon_{ii}\left(t\right) = J_L\left(t\right)\sigma_{ii} = J_L\left(t\right) \tag{6a}$$

$$\varepsilon_{kk}\left(t\right) = \varepsilon_{jj}\left(t\right) = -J_\upsilon\left(t\right)\sigma_{ii} \tag{6b}$$

with $i \neq j \neq k$ and $i,\, j,\, k = 1, 2, 3$. In eqs.(6a-b) the function of $J_L\left(t\right)$ and $J_\upsilon\left(t\right)$ are the axial and the transverse creep functions with respect to the stress direction, respectively. Under the assumption of smooth load process $\sigma_{ij}(t)$ the presence of contemporaneous stress $\sigma_{ij}(t) = \sigma_{ij}(t)\delta_{ij}$, with $i = 1, 2, 3$, may be account for by the integral.

$$\varepsilon_{ii}\left(t\right) = \int_0^t J_L\left(t-\tau\right)\dot{\sigma}_{ii}\left(\tau\right) - J_\upsilon\left(t-\tau\right)\left[\dot{\sigma}_{jj}\left(\tau\right) + \dot{\sigma}_{kk}\left(\tau\right)\right]d\tau \tag{7}$$

with $i \neq j \neq k$ and i,j,k=1,2,3, respectively. The assumption of isotropic hereditariness yields that the shear stress $2\varepsilon_i j(t)$ $i \neq j$, is not caused by axial stress, and it involves only the shear stress $\sigma_i j(t)$. Under these assumption, introducing the shear creep function $J_T(\cdot)$ the constitutive relation for the shear strain reads:

$$2\varepsilon_{ij}\left(t\right) = \int_0^t J_T\left(t-\tau\right)\dot{\sigma}_{ij}\left(\tau\right)d\tau \tag{8}$$

with $i \neq j$ and $i, j = 1, 2, 3$.

Direct form of constitutive equations in eqs.(7, 8) may be represented in Voigt formulation as:

$$\boldsymbol{\varepsilon}\left(t\right) = \int_0^t \mathbf{J}\left(t-\tau\right)\dot{\boldsymbol{\sigma}}\left(\tau\right)d\tau \tag{9}$$

where $\mathbf{J}(t)$ is creep functions matrix that may be reported in block matrix formulation as:

$$\mathbf{J}\left(t\right) = \begin{bmatrix} \mathbf{J}^{(A)}\left(t\right) & \mathbf{0} \\ \mathbf{0} & \mathbf{J}^{(T)}\left(t\right) \end{bmatrix} \tag{10}$$

where the elements of the axial creep matrix $\mathbf{J}^{(A)}(t)$ are:

$$J_{ij}^{(A)}\left(t\right) = J_L\left(t\right)\delta_{ij} - \left(1-\delta_{ij}\right)J_\upsilon \tag{11}$$

with $i, j = 1, 2, 3$. The shear creep matrix $\mathbf{J}^{(T)}(t)$ is diagonal matrix elements:

$$J_{ij}^{(T)}\left(t\right) = J_T\left(t\right)\delta_{ij} \tag{12}$$

The three creep functions $J_L(t)$,$J_\upsilon(t)$ and $J_T(t)$ are related by a linear relation that reads:

$$J_T(t) = 2J_L(t) - J_\upsilon(t) \tag{13}$$

that may be obtained with straightforward manipulation among a pure shear stress $\sigma_{ij}(t)$ corresponding to shear strain $\gamma_{ij} = 2\varepsilon_{ij}(t)$ and correlating the elongation along principals directions at angles of $\pi/4$.

Under the assumption linear elasticity the creep functions coincides with the material compliance that reads $J_T = 1/G$, $J_L = 1/E$ and $J_\upsilon = \upsilon/E$ yielding, after substitution in eq.(13):

$$\frac{1}{G} = 2\left(\frac{1}{E} + \frac{\upsilon}{E}\right) = \frac{2(1+\upsilon)}{E} \tag{14}$$

that is the well-known relation among elasticity moduli. The knowledge of the creep function matrix, namely, $\mathbf{J}(t)$ in eq.(10) allows for the definition of the relaxation matrix $\mathbf{G}(t)$ by means of the coniugation relation as:

$$\hat{\mathbf{G}}(s)\hat{\mathbf{J}}(s) = \frac{1}{s^2}\mathbf{I} \tag{15}$$

where $\mathbf{I}$ is the identity matrix and $\widehat{\boldsymbol{G}(s)}$, $\widehat{\boldsymbol{J}(s)}$ are the Laplace transforms of the relaxation $\mathbf{G}(t)$ and the creep $\mathbf{J}(t)$ matrices. Straightforward manipulations of eq.(15) and inverse Laplace transform the relation matrix may be written as:

$$\mathbf{G}(t) = \begin{bmatrix} \mathbf{G}^{(A)}(t) & \mathbf{0} \\ \mathbf{0} & \mathbf{G}^{(T)}(t) \end{bmatrix} \tag{16}$$

where:

$$G_{ij}^{(A)}(t) = \mathcal{L}^{-1}\left[\frac{1}{s^2\left(\hat{J}_L + \hat{J}_\upsilon\right)\left(\hat{J}_L - 2\hat{J}_\upsilon\right)}\right]\left[\left(\hat{J}_L - \hat{J}_\upsilon\right)\delta_{ij} + (1-\delta_{ij})\hat{J}_\upsilon\right] \tag{17a}$$

$$G_{ij}^{(T)}(t) = \mathcal{L}^{-1}\left[\frac{1}{s^2\left(\hat{J}_L + \hat{J}_\upsilon\right)}\right]\delta_{ij} \tag{17b}$$

The observation of eqs.(17 a,b) shows that in presence of material fading memory, that is material hereditariness there is a need to obtain the relaxation functions in the relaxation matrix $\mathbf{G}(t)$ from a combination of creep functions obtained by uniaxial creep tests. Similar considerations hold true also assuming that from uniaxial traction relaxation tests, the relaxation functions $G_L(t)$ is obtained in lateral free conditions that is the specific state of strain involves $\varepsilon_{11} \neq \varepsilon_{22} \neq 0$ and $\varepsilon_{33} = 1$ measuring only the decay of the axial stress $\sigma_{33}(t)$ but with applied lateral stress $\sigma_{11} = \sigma_{22} = 0$.

Summing up, the aforementioned considerations show that the relaxation matrix $\mathbf{G}(t)$ cannot be obtained from uniaxial traction/torsion relaxation.

Similar but less intuitive arguments hold about the relation among the shear, $G_T(t)$, longitudinal and transverse relaxation functions namely $G_L(t)$ $G_\upsilon(t)$ that reads:

$$G_T(t) = \frac{1}{2}\left(G_L(t) - G_\upsilon(t)\right) \tag{18}$$

allowing, the evaluation of the transverse relaxation $G_\upsilon(t)$ as:

$$G_\upsilon(t) = 2G_T(t) - G_\upsilon(t) \tag{19}$$

The relations among the creep and relation functions involved in isotropic material hereditariness in eqs.(13, 19) do not depend on the specific functional class used to capture experimental data.

In the following section, the introduction of power-law hereditariness for the 3D constitutive equation is found in the context of thermodynamical restriction to introduce some constrains on material parameters.

2.3. The isotropic fractional-order hereditariness

In this section we assume that relaxation functions are expressed as:

$$G_L(t) = \frac{C_\alpha^{(L)} t^{-\alpha}}{\Gamma(1-\alpha)} + \overline{C}_L \tag{20a}$$

$$G_T(t) = \frac{C_\alpha^{(T)} t^{-\alpha}}{\Gamma(1-\alpha)} + \overline{C}_T \tag{20b}$$

where $C_\alpha^{(v)} = 2C_\alpha^{(T)} - C_\alpha^{(L)}$ and $\overline{C}^{(v)} = 2\overline{C}_T - \overline{C}_L$ and $G_v(t)$ according to eq.(19). Under these circumstances, the relaxation matrix of the material may be represented as

$$\mathbf{G}(t) = \mathbf{C}_\alpha \frac{t^{-\alpha}}{\Gamma(1-\alpha)} + \bar{\mathbf{C}} \tag{21}$$

Where the matrix:

$$\mathbf{C}_\alpha = \begin{bmatrix} \mathbf{C}_\alpha^{(A)} & \mathbf{0} \\ \mathbf{0} & \mathbf{C}_\alpha^{(T)} \end{bmatrix} \tag{22a}$$

$$\bar{\mathbf{C}} = \begin{bmatrix} \underline{\bar{C}}^{(A)} & \mathbf{0} \\ \mathbf{0} & \bar{\mathbf{C}}^{(T)} \end{bmatrix} \tag{22b}$$

and

$$C_{ij}^{(A)} = C_\alpha^{(L)}\delta_{ij} + (1-\delta_{ij})\, C_\alpha^{(v)}; \quad \bar{C}_{ij} = \bar{C}^{(A)}\delta_{ij} + (1-\delta_{ij})\,\bar{C}^{(v)} \tag{23}$$

The use of eq.(21) substituted into the constitutive equations for the tree-axial hereditariness yields a relation among the value of the stress vector and history of the strain vector $\boldsymbol{\varepsilon}(t)$ as:

$$\boldsymbol{\sigma}(t) = \mathbf{C}_\alpha \int_0^t (t-\tau)^{-\alpha} \dot{\boldsymbol{\varepsilon}}(\tau)\, d\tau + \bar{\mathbf{C}} = \mathbf{C}_\alpha \left(D_{0^+}^\alpha \boldsymbol{\varepsilon}\right)(t) + \bar{\mathbf{C}} \tag{24}$$

The stress vector in eq.(24) in terms of the strain vector $\boldsymbol{\varepsilon}(t)$ is the three-axial generalization of the constitutive equation reported in eq.(3a).

In the following section we show that, under the assumption of material isotropy, there are some restrictions on the expressions of the power-laws used for $G_L(t)$ and $G_T(t)$.

2.4. Power-laws 3D hereditariness: Thermodynamic restrictions

In this section, we assume that the relaxation tests conducted for the restrained axial and shear relaxations may be captured by power-laws with dotted decay order $(\alpha \neq \beta)$ as:

$$G_L(t) = C_L^{(\alpha)} t^{-\alpha} + \bar{G}_L; \quad G_T(t) = C_T^{(\beta)} t^{-\beta} + \bar{G}_T \tag{25a}$$

$$G_v(t) = 2\left(C_T t^{-\beta} + \bar{G}_T\right) - \left(C_L t^{-\alpha} + \bar{G}_L\right) \tag{25b}$$

with eq.(24b) obtained from the application of eq.(18). Physical dimensions of the coefficients are $[C_L] = [C_T] = F/L^2$, $\left[C_L^{(\alpha)}\right] = \dfrac{F}{L^2 T^{-\alpha}}$, $\left[C_L^{(\alpha)}\right] = \dfrac{F}{L^2 T^{-\beta}}$. The observation of eq.(24a, b) yields a relaxation matrix containing two different power-laws with orders β and α ($\alpha, \beta \in [0, 1]$). It may be observed that the relaxation matrix must satisfy some thermodynamic restrictions [22] about the functional class of the elements collected in the matrix, namely $G_L(t)$, $G_\upsilon(t)$ and $G_T(t)$. Indeed, a dissipative material is guaranteed, only if the restrictions:

$$\mathbf{G}(0) \geq \mathbf{G}(\infty) \geq 0 \tag{26}$$

$$\dot{\mathbf{G}}(0) \geq 0 \tag{27}$$

are fulfilled by the relaxation matrix [23, 24, 25, 26]. It can be verified that eqs.(26,27) are always satisfied assuming positive values of coefficients $\bar{G}_L$, $\bar{G}_T$ and $C_L^{(\alpha)}$ and $C_T^{(\beta)}$, whereas the latter eq.(26) is satisfied, only, as the eigenvalues of the first derivative of the matrix, namely, $\dot{\mathbf{G}}(0)$ are all negative. In order to check this requirement we introduce a one-parameter family of relaxation matrices defined on a real parameter δ as $\mathbf{G}_\delta(t) = \mathbf{G}(t+\delta)(t)$ and we investigated the behavior of the matrix family $\dot{\mathbf{G}}_\delta(t)$.

In this regard matrix $\dot{\mathbf{G}}(t)$ reads:

$$\dot{\mathbf{G}}_\delta(t) = \begin{bmatrix} \dot{\mathbf{G}}_\delta^{(A)}(t+\delta) & \mathbf{0} \\ \mathbf{0} & \dot{\mathbf{G}}_\delta^{(T)}(t+\delta) \end{bmatrix} \tag{28}$$

where the elements of the matrix reads:

$$\dot{G}_\delta^{(A)}(t+\delta) = -C_L^{(\alpha)} \alpha (t+\delta)^{-\alpha-1} \tag{29a}$$

$$\dot{G}_\delta^{(T)}(t+\delta) = -C_T^{(\beta)} \beta (t+\delta)^{-\beta-1} \tag{29b}$$

$$\dot{G}_\delta^{(\upsilon)}(t+\delta) = -2C_T^{(\beta)} \beta (t+\delta)^{-\beta-1} + C_L^{(\alpha)} \alpha (t+\delta)^{-(\alpha+1)} \tag{29c}$$

Observing that the one-parameter family $\dot{\mathbf{G}}_\delta(t)$ leads, to the limit to:

$$\lim_{\delta \to 0} \dot{\mathbf{G}}_\delta(t) = \dot{\mathbf{G}}(t) \tag{30}$$

We can infer the behavior of $\dot{\mathbf{G}}(t)$ from those of the family $\mathbf{G}_\delta(t)$ and letting $\delta \to 0$. In this regard, the requirement in eq.(27) may be obtained as:

$$-\dot{G}\left(0\right) = -\lim_{\delta\to 0} \dot{G}_\delta\left(t\right) \geq 0 \tag{31}$$

that is we evaluate the eigenvalues $\lambda_i(\delta)$ $(i = 1, 2, ...6)$ of the matrix $\dot{\mathbf{G}}_\delta(0)$ and we require that simultaneously $-\lambda_i(\delta) \geq 0$ $(i = 1, 2, ...6)$ as $\delta \to 0$. Evaluation of the eigenvalues $\lambda_i(\delta)$ reads:

$$-\lambda_1\left(\delta\right) = -\lambda_2\left(\delta\right) = -2\left(\dot{G}_L\left(\delta\right) - \dot{G}_T\left(\delta\right)\right) \geq 0 \tag{32a}$$

$$-\lambda_3\left(\delta\right) = -\lambda_4\left(\delta\right) = -\lambda_5\left(\delta\right) = -\dot{G}_T\left(\delta\right) \geq 0 \tag{32b}$$

$$-\lambda_6\left(\delta\right) = -4\dot{G}_T\left(\delta\right) + \dot{G}_L\left(\delta\right) \geq 0 \tag{32c}$$

Substitution of eq.(29a,b) into eq.(32 b) shows that the inequality is fulfilled for $C_T^{(\beta)} \geq 0$ and $0 \leq \beta \leq 1$. The inequalities in eqs.(32 a,c) read, after substitution:

$$\alpha C_\alpha \delta^{-(\alpha+1)} - \beta C_\beta \delta^{-(\beta+1)} \geq 0 \tag{33a}$$

$$4\beta C_\beta \delta^{-(\beta+1)} - \alpha C_\alpha \delta^{-(\alpha+1)} \geq 0 \tag{33b}$$

that, after some straightforward manipulation, may be cast in a more suitable form, taking natural logarithms.

$$\ln\left(A_{\alpha\beta}\right) \geq \left(\alpha - \beta\right)\ln\delta \tag{34a}$$

$$\ln\left[\frac{\left(A_{\alpha\beta}\right)}{4}\right] \leq \left(\alpha - \beta\right)\ln\delta \tag{34b}$$

where $A_{\alpha\beta} = \alpha C_L^{(\alpha)}/\beta\left(C_T^{(\beta)}\right)$. The two inequalities in eqs.(34 a,b) must be fulfilled for any value of the parameter δ yielding that $\alpha = \beta$. Moreover, in this latter case the additional thermodynamical restriction holds true.

$$C_T \leq C_L \leq 3C_T \tag{35}$$

In passing we observe that the condition $\alpha = \beta$ holds true only for the two terms (or one terms) description of the relaxation function in eq.(25 a).

Indeed, as assume that the relaxation functions $G_L(t)$ and $G_T(t)$ involve linear combinations of power-laws:

$$G_L\left(t\right) = \sum_{j=1}^{N} C_L^{(j)} t^{-\alpha_j}; G_T\left(t\right) = \sum_{i=1}^{M} C_T^{(i)} t^{-\beta_i} \tag{36}$$

with N and M the number of power-laws involved. The thermodynamical arguments proposed this study yields that:

$$\max_{j=1,N}\left(\alpha_j\right) = \max_{i=1,M}\left(\beta_j\right) \tag{37a}$$

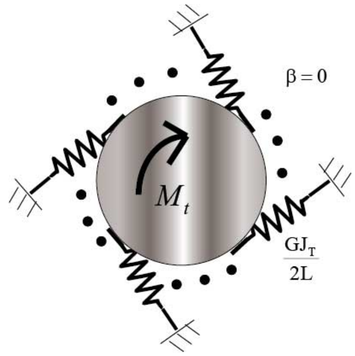
$$\min_{j=1,N}\left(\alpha_j\right) = \min_{i=1,M}\left(\beta_j\right) \tag{37b}$$

In the next section the multiaxial fractional-order hereditariness will be further discussed introducing a mechanical hierarchy that corresponds exactly to the multiaxial constitutive model expressed in eq.(24).

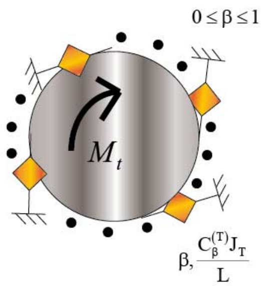
3. The exact mechanical representation of three-axial fractional-order isotropic hereditariness

Relaxation functions in eq.(25b) corresponds to an exact rheological model that generalizes the 1D springpot reported in eq.(7). The mechanical model generalizing the one-dimensional springpot has been recently reported in recent scientific literature [27, 28, 29, 30]. The main difference among the uniaxial springpot and multiaxial case the presence of pure shear stress and strain in the 3D rheology.

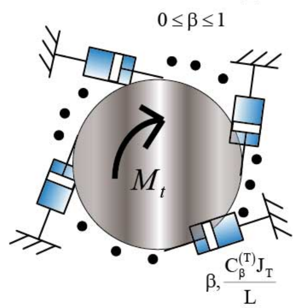
These effect may be construed as we introduce a torsional rheologic element reported in the correspondent figures of the eqs.(38-40) that are represented by the constitutive relation.



$$
\begin{aligned}
F &= k_L u = \frac{EA}{L} u \\
M_t &= k_T \varphi = \frac{GJ_T}{L} \varphi
\end{aligned}
\tag{38}
$$



$$
\begin{aligned}
F &= \frac{C_\beta^{(L)} A}{L} \left(D^\beta u\right)(t) \\
M_t &= \frac{C_\beta^{(T)} J_T}{L} \left(D^\beta \varphi\right)(t)
\end{aligned}
\tag{39}
$$



$$
\begin{aligned}
F &= \frac{\eta_L A}{L} \dot{u} \\
M_t &= \frac{\eta_T J_T}{L} \dot{\varphi}
\end{aligned}
\tag{40}
$$

where we denoted J_t and A the torsional inertia and the cross-section of the model, respectively.

The constitutive equations in eqs.(38,40) involves respectively: i) a linear elastic spring ; ii) a linear viscous element and iii) a linear shear springpot. In the following we introduced a hierarchic mechanical model to capture the axial and shear hereditariness assuming power-law description of the creep and relaxation functions for axial and shear stress/strain, respectively [31, 28, 32, 27]. The obtained mechanical hierarchy corresponds exactly to an axial and shear springpots with the same order of time evolution/decay.

To this aim let us introduce an elastic column of unbounded length with circular cross section of radius R. The elastic features of the column are non-costant along the column axis and vary with the coordinate as:

$$E\left(z\right)=\frac{E_{\alpha}}{\Gamma\left(1-\alpha\right)}z^{-\alpha};\quad G\left(z\right)=\frac{G_{\alpha}}{\Gamma\left(1-\alpha\right)}z^{-\alpha}\qquad -1\leq\alpha\leq 1 \tag{41}$$

The column is externally restrained by a set of torsional and axial viscous dashpots fig.(5) with non-homogeneous viscosity $\eta(z)$ as:

$$\eta\left(z\right)=\frac{\eta_{\alpha}}{\Gamma\left(1+\alpha\right)}z^{-\alpha}\qquad -1\leq\alpha\leq 1 \tag{42}$$

Axial and torsional equilibrium along the column axis reads:

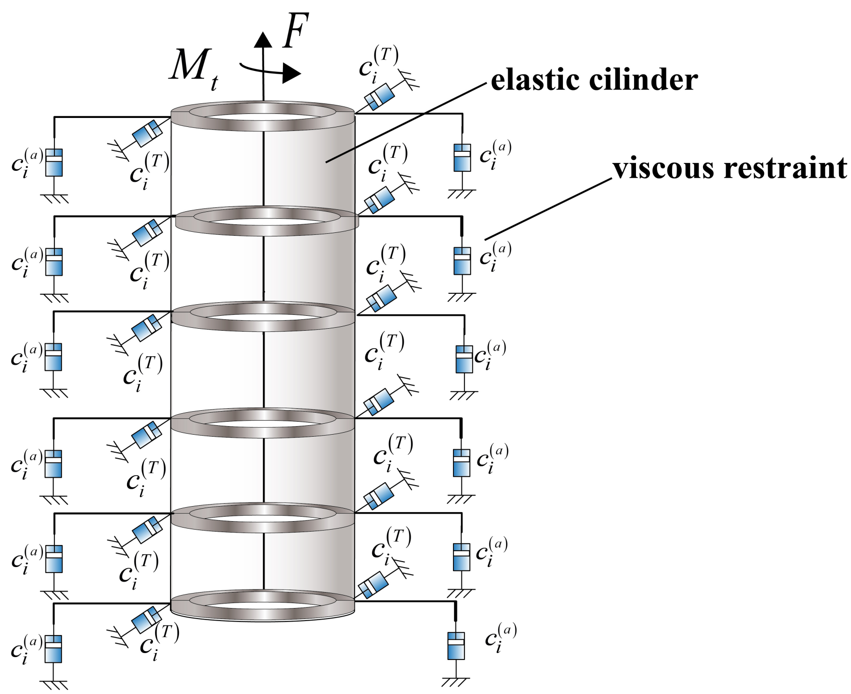


Figure 5: column with non-homogeneous viscosity

$$\frac{\eta_{\alpha}}{\Gamma\left(1+\alpha\right)}z^{-\alpha}2\pi R\Delta z\dot{u}\left(z,t\right)=\frac{E_{\alpha}\pi R^{2}s(z+\Delta z)^{-\alpha}}{\Gamma\left(1-\alpha\right)}\left[u\left(z+\Delta z,t\right)-u\left(z,t\right)\right]+$$

$$+\frac{E_{\alpha}\pi R^{2}sz^{-\alpha}}{\Gamma\left(1-\alpha\right)}\left[u\left(z,t\right)-u\left(z-\Delta z,t\right)\right] \tag{43}$$

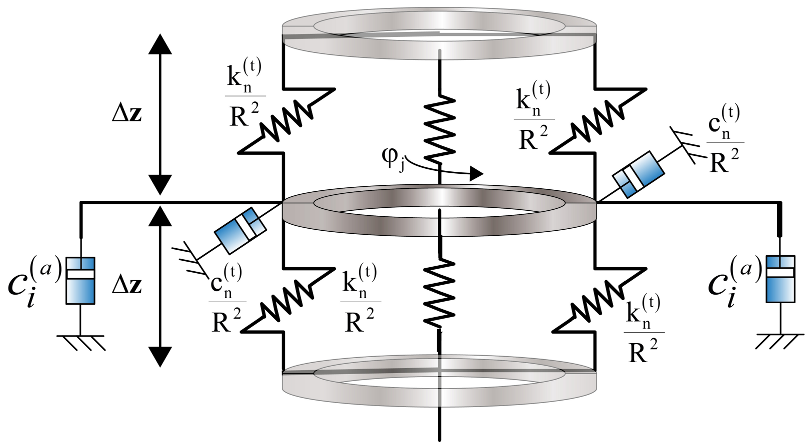


Figure 6: elements of the column with non-homogeneous viscosity

$$\frac{\eta_\alpha}{\Gamma(1+\alpha)} z^{-\alpha} 2\pi R^2 \Delta z \dot{\varphi}(z,t) = G_\alpha \pi R^4 (z+\Delta z)^{-\alpha} \left[\varphi(z+\Delta z,t) - \varphi(z,t)\right] +$$

$$+ G_\alpha \pi R^4 (z+\Delta z)^{-\alpha} \left[\varphi(z,t) - \varphi(z-\Delta z,t)\right] \quad (44)$$

that can be rewritten in differential form, letting $\Delta z \to 0$ as:

$$\frac{\eta_\alpha z^{-\alpha}}{\Gamma(1+\alpha)} \frac{\partial u(z,t)}{\partial t} = \frac{E_\alpha R s}{\Gamma(1-\alpha)} \frac{\partial}{\partial z} \left(z^{-\alpha} \frac{\partial u(z,t)}{\partial z} \right) \quad (45a)$$

$$\frac{\eta_\alpha z^{-\alpha}}{\Gamma(1+\alpha)} \frac{\partial \varphi(z,t)}{\partial t} = \frac{G_\alpha R}{\Gamma(1-\alpha)} \frac{\partial}{\partial z} \left(z^{-\alpha} \frac{\partial \varphi(z,t)}{\partial z} \right) \quad (45b)$$

Boundary conditions involving the differential fields $u(z,t)$ and $\varphi(z,t)$ in eqs.(45a,b) read, respectively.

$$\lim_{z\to\infty} u(z,t) = 0 \quad (46a)$$

$$\lim_{z\to 0} \frac{E_\alpha}{\Gamma(1-\alpha)} z^{-\alpha} \frac{\partial u}{\partial z} = F_0 \quad (46b)$$

$$\lim_{z\to\infty} \varphi(z,t) = 0 \quad (47a)$$

$$\lim_{z\to 0} \frac{G_\alpha}{\Gamma(1-\alpha)} z^{-\alpha} \frac{\partial \varphi}{\partial z} = M_0 \quad (47b)$$

The mathematical operators and the boundary conditions in eqs.(46a,b) are completely equivalent to those of a previous differential problem that has been solved resorting to a non- linear mapping followed by Laplace transform [28, 33]. Such a procedure yields a Bessel differential equation of second kind in terms of the anomalous Laplace parameters. Position of the boundary conditions and inverse Laplace transform provides solution in the form:

$$u_0(t) = u_0(z,t) = \lim_{z\to\infty} u(z,t) = \frac{t^{-\beta}}{k_\beta^{(L)}} F_0 = J_L(t)\,\overline{F}_0 \tag{48}$$

$$\varphi_0(t) = \varphi_0(z,t) = \lim_{z\to\infty} \varphi(z,t) = \frac{t^{-\beta}}{k_\beta^{(T)}} M_0 = J_T(t)\,\overline{M}_0 \tag{49}$$

with:

$$k_\beta^{(L)} = \frac{\Gamma(2\beta)\left(\tau_L^\beta\right)}{E_\alpha 2^{1-2\beta}\Gamma(\beta)\Gamma(1-\beta)} \tag{50}$$

$$k_\beta^{(T)} = \frac{\Gamma(2\beta)\left(\tau_L^\beta\right)}{G_\alpha 2^{1-2\beta}\Gamma(\beta)\Gamma(1-\beta)} \tag{51}$$

with $\beta = \frac{1+\alpha}{2}$ and the relaxation times:

$$\tau_L = \frac{\eta_\alpha}{E_\alpha}\frac{\Gamma(1-\alpha)}{\Gamma(1+\alpha)} \tag{52}$$

$$\tau_T = \frac{\eta_\alpha}{G_\alpha}\frac{\Gamma(1-\alpha)}{\Gamma(1+\alpha)} \tag{53}$$

Effect superpositions provides, resorting to the fundamental equations of linear viscoelasticity, the constitutive equations of the macroscopic variables as:

$$F_0(t) = k_\beta^{(L)}\left(D_{0+}^\beta u_0\right)(t) \tag{54}$$

$$M_0(t) = k_\beta^{(T)}\left(D_{0+}^\beta \varphi_0\right)(t) \tag{55}$$

Eqs.(54, 55) are the constitutive equation at the macro-scale and, recalling that $F_0 = \sigma_{33}A$ and $|\tau| = \sqrt{|t_{31}|^2 + |t_{32}|^2} = \frac{M_0}{2As}$ the constitutive equation of the material reads:

$$\sigma_{33} = C_\beta^{(L)}\left(D^\beta \varepsilon_{33}\right)(t) \tag{56}$$

$$|\tau| = C_\beta^{(T)} \left(D^\beta \, |\gamma| \right) (t) \tag{57}$$

with the coefficients $C_\beta^{(L)}$ and $C_\beta^{(T)}$ that read:

$$C_\beta^{(L)} = \frac{k_\beta^{(L)} \bar{l}}{A} \qquad\qquad C_\beta^{(T)} = \frac{k_\beta^{(T)}}{2As} \frac{R}{\bar{l}} \tag{58}$$

and $\bar{l}$ an internal length of the material.

4. Conclusions

The mathematical structure of the fractional-order isotropic hereditariness has been discussed in the paper. The study has been framed in the context of biomimetic ceramics used in cranioplasty neurosurgery (i.e. CustomBone® "prosthesis"). The creep and relaxation functions of isotropic linear hereditarinnes have been particularized for power-law decays yielding a multi-axial constitutive model in terms ofg fractional-order operators. Additionally a specific mechanical model has been introduced to describe the three-axial constitutive model expend in terms of fractional-order operator. In future studies experimental campaigns involving creep and relaxation of biomimetic ceramics will be reported to assess the validity of material isotropy. Additionally, the proposed hierarchy will be further extended to deal with non-linear hereditariness as those observed in creep and relaxations of tendons and ligaments.

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Power-Laws hereditariness of biomimetic ceramics for cranioplasty neurosurgery

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Abstract

We discuss the hereditary behavior of hydroxyapatite-based composites used for cranioplastic surgery in the context of material isotropy. We classify mixtures of collagen and hydroxiapatite composites as biomimetic ceramic composites with hereditary properties modeled by fractional-order calculus. We assune isotropy of the biomimetic ceramic is assumed and provide thermodynamic of restrictions for the material parameters. We exploit the proposed formulation of the fractional-order isotropic hereditariness further by means of a novel mechanical hierarchy corresponding exactly to the three-dimensional fractional-order constitutive model introduced.

Keywords: `Biomimetic materials,cranioplasty, fractional calculus, power-law hereditariness, isotropic hereditariness.`

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1. Introduction

Cranioplastic neurosurgery is widespread nowadays since it is necessary both in traumatic therapies or in the presence of specific oncologic pathology. Cranio-

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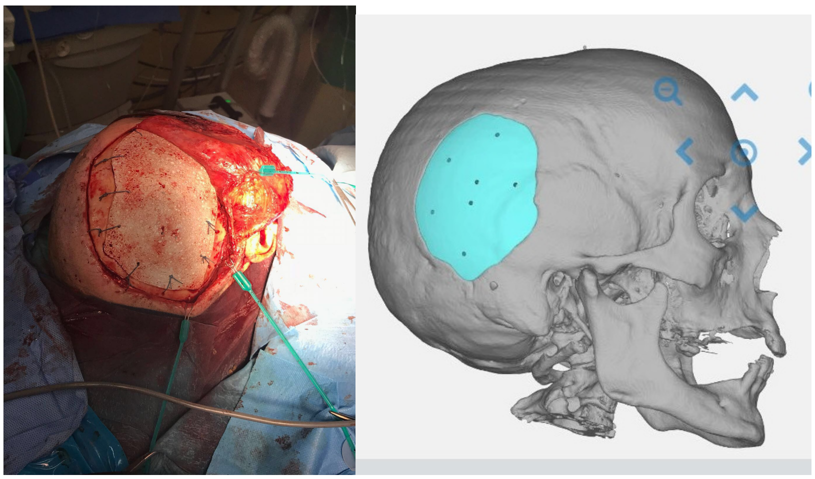
plasty is a surgical procedure that aims to re-establish the skull integrity following a previous craniotomy due to the occurrence of traumas, tumors and/or congenital malformations. In all cases cranioplasty can be considered as the conclusive action of a surgery initiated by the removal of a bone operculum, see fig.1.

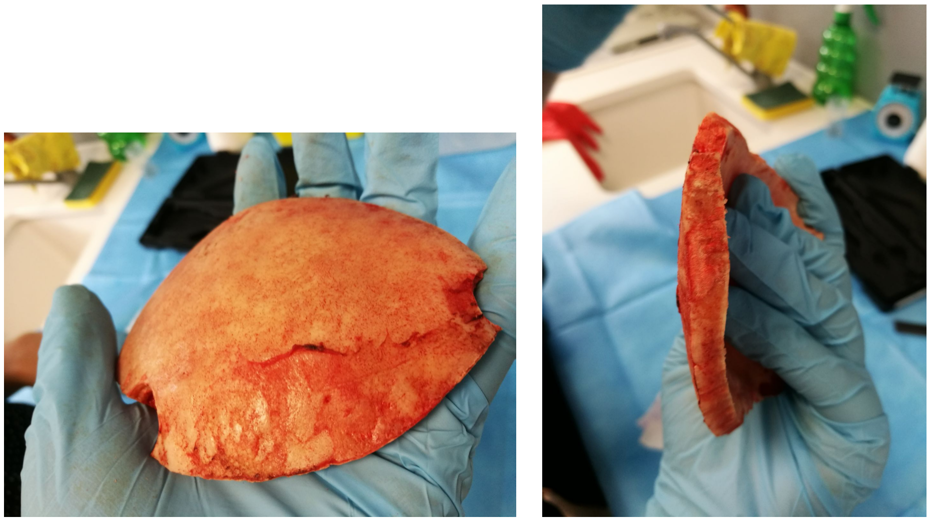


(a) (b)

Figure 1: (a) cranioplasty surgery, Policlinico Giaccone Palermo; (b) preclinical analysis

Ideally, cranioplasty procedures should provide restoration of the protective functions of the skull with maintenance of the original aesthetics and long-term mechanical performance [1]. The ideal material for cranioplasty should be chemically inert, biocompatible, biomechanically reliable, easily manufactured, individually shaped, safe, and able to promote osteoblast migration. Nowadays synthetic implants based on metallic (mainly titanium) or acrylic plates (mainly polymethylmeta-crylate or polyetheretherketone) are widely used in cranioplasty procedures. These are bioinert materials with good biocompatibility, resistance to infections, ease of sterilization, ability to be subjected to imaging diagnostics, and the capacity to undergo flexible design for adaptation to different clinical cases. They exhibit good mechanical strength, which offers adequate brain protection from external shocks. However, they present poor osteogenic and osteoconductive ability, thus resulting in a foreign body functioning as a shell expected to provide brain protection, but connected to the surrounding bone only by its perimeter contact surface. In order to overcome many limitations an Hydroxyapatite (HA)-based material has been widely considered for decades as the gold standard for bone scaffolds, as its composition is very close to that of bone mineral, thus exhibiting excellent biocompatibility, a low inflammatory reaction as well as good osteogenic ability and osteoconductivity. The hydrophilic character of HA favors cell attachment and tight adhesion of bone to the scaffold surface, which is a key target for the stability of the bone/implant interface. Therefore, HA scaffolds presenting wide, open and interconnected multiscale porosity can induce extensive bone ingrowth and penetration throughout the whole scaffold, partly thanks to the possibility of massive fluid perfusion, which triggers and assists neovascularization. Hence, cranial reconstruction using synthetic porous HA has recently become the subject of intense debate among surgeons, and it now represents a new concept in cranioplasty procedures. The custom-made concept was first applied to porous hydroxyapatite because of the need to overcome the fragility of the material itself. One of the advantages of HA-based prosthesis is customization.

Indeed, in the presence of cranioplasty, the morphology of the bone to be replaced with a synthetic prosthesis must match completely the original bone to accelerate the osteointegration of the prosthesis [2, 3, 4] in the surgical hole. In fig.(2 a-d) a human parietal bone and its synthetic prosthesis, see fig.(3 a-c) have been obtained from a university neurosurgery hospital in Palermo. The synthetic bone used for replacement is a CustomBone® (Finceramica Faenza), namely custom-made, porous hydroxyapatite scaffolds with total porosity in the range of 60 to 70 % and pore architecture based on macro-pores (> 100 micron) interconnected with micro-pores (5-10 micron). CustomBone® scaffolds were obtained by reproduction of the patients bone defect as modeled by 3D CT scan. They are made of a composite ceramics material obtained from chemical deposition of hydroxyiapatite with a small fraction of collagen type I (see fig.1a).



(a) (b)

Figure 2: (a) human parietal bone, Policlino Giaccone Palermo; (b) human parietal bone lateral section, Policlino Giaccone Palermo

The use of biomimetic ceramics to replace cortical as well as trabecular bone is a well- defined technique in bone surgery [5]. Indeed the mechanical features of the prosthesis in terms of elastic moduli and the strength of the biomimetic composite of integration are very similar. However, the use of ceramic materials to replace the bones of a human head may involve different behaviors in terms of energy dissipation. Indeed, biologic tissues show marked hereditariness due to the reptation of the collagen chains of the material as well as to the fiber recruitment in the tissues. Material hereditariness involves additional stresses that may be applied to the grafted ceramics prosthesis and may lead to fracture propagation during patient follow-up [6].

The hereditary properties of bone in uniaxial test are represented by creep $J(t)$ and relaxation $G(t)$ functions formulated in terms of power-law $J(t) \propto t^{\beta}$ and $G(t) \propto t^{-\beta}$ with $0 \leq \beta \leq 1$, yielding an accurate description of experimental data [5, 7, 8, 9, 10]. Power-laws hereditariness in conjuction with Boltzmann

(a) (b)

Figure 3: (a) CustomBone® prosthesis morphology; (b) CustomBone® lateral section of

superposition yields the constitutive behavior in terms of so-called fractional integrals and derivatives. Fractional calculus may be considered as a generalization of the classical differential calculus to real-order integration and differentiation $\left(i.e. df/dt \rightarrow d^{\beta} f/dt^{\beta}\right)$ with $\beta \in [0, 1]$ as reported in classical references [11, 12, 13, 14, 15]. In such a context, uniaxial hereditariness [16, 17, 18, 19] involving fractional order stress-strain relations has been reported since the beginning of the 20th century [20, 13] defining the so-called springpot element [21, 22].

In the presence of tensorial stress/strain state, as in the continuum mechanics description of biomimetic prosthesis, no generalities have been reported in the scientific literature to capture multiaxial hereditariness with fractional-order calculus, to the best of the authors' knowledge. Indeed, in several cases recently discussed in the scientific literature [23, 24], the use of power-laws without thermodynamic restrictions the parameters does not guarantee positive entropy rate for any strain/stress process involved by material.

In this paper, a 3D constitutive relation describing material hereditariness is discussed in the context of power-laws functional classes of the relaxation/creep functions. We show that, under the assumption of material isotropy, thermodynamical restrictions on the constitutive parameters allow us to formulate the constitutive behavior in terms of a Caputo fractional derivative that is formally analogous to the constitutive behavior in uniaxial state of stress/strain.

The paper is organized as follows. Section 2 provides generalities about fractional-order calculus and isotropic hereditariness; section 3 reports a mechanical hierarchy that corresponds exactly to the isotropic fractional-order hereditariness. Section 4 provides some conclusions about the proposed model of isotropic hereditariness and the influence on the mechanics of the biomimetic ceramics prosthesis.

2. Power-law hereditariness of isotropic biomimetic ceramics

In this section we outline the constitutive relations in the presence of power-law hereditariness, including the main arguments of power-law hereditariness under uniaxial stress/strain, generalization to the isotropic case, and thermodynamic restrictions on the material parameters.

2.1. Uniaxial power-law hereditariness: The fractional order constitutive equation

We describe the constitutive behavior of materials in long-standing mechanical tests is described by means of the well-known creep and relaxation functions, dubbed $J(t)$ and $G(t)$, respectively. The linear superposition applied to a generic stress/strain history, namely $\sigma(\tau)$ and $\varepsilon(\tau)$ with $\tau \leq t$, yields:

$$\sigma(t) = \int_0^t G(t-\tau)d\varepsilon(\tau) = \int_0^t G(t-\tau)\dot{\varepsilon}(\tau)d\tau \tag{1a}$$

$$\varepsilon(t) = \int_0^t J(t-\tau)d\sigma(\tau) = \int_0^t J(t-\tau)\dot{\sigma}(\tau)d\tau \tag{1b}$$

Eqs.(1a, b) are defined in terms of Boltzman superposition with $d\sigma = \dot{\sigma}dt$ and $d\varepsilon = \dot{\varepsilon}dt$ increments, where $[\dot{\cdot}] = \frac{d}{dt}$. Creep and relaxation functions characterize the material behavior and they must satisfy the conjugation relation $\hat{J}(s)\hat{G}(s) = 1/s^2$, where s indicates the Laplace parameter and $\hat{f}(s) = \mathcal{L}[f(t)]$ is the Laplace transform of the generic function $f(t)$. In the context of materials hereditariness, power-law representation of creep and relaxation functions, i.e. $J(t)$ and $G(t)$, was introduced at the beginning of the last century [20],

$$G(t) = \frac{C_\beta}{\Gamma(1-\beta)} t^{-\beta}, \tag{2a}$$

$$J(t) = \frac{1}{C_\beta \Gamma(\beta+1)} t^{\beta} \tag{2b}$$

where $\Gamma(\cdot)$ is the Euler-Gamma function, $\beta \in [0, 1]$ and $C_\beta > 0$, are material parameters, that may be estimated through a best-fitting procedure of experimental data [25, 26]. Straightforward manipulations show that the power-law functional class in eqs.(2a, b) satisfies the conjugation relation and it yields, upon substitution in eqs. (1a, b) the following constitutive relations:

$$\sigma(t) = \frac{C_\beta}{\Gamma(1-\beta)} \int_0^t (t-\tau)^{-\beta} \dot{\varepsilon}(\tau) d\tau = C_\beta \left(D^\beta_{0^+} \varepsilon \right)(t) \tag{3a}$$

$$\varepsilon(t) = \frac{1}{C_\beta \Gamma(\beta+1)} \int_0^t (t-\tau)^{\beta} \dot{\sigma}(\tau) d\tau = \frac{1}{C_\beta} \left(I^\beta_{0^+} \sigma \right)(t) \tag{3b}$$

in terms of the Caputo fractional derivative and Riemann-Liouville fractional integral, respectively.

Use of power-laws and, as a consequence, of fractional-order operators is usually connected, in a rheological context [27], to the introduction of the springpot element.

Springpot is a one-dimensional element defined in terms of two parameters, i.e. C_β and β, $0 \leq \beta < 1$ and $C_\beta > 0$ whose constitutive relation is reported in eqs.(3a,b). Such element with an intermediate behavior among elastic springs and viscous dashpot, is widely used nowadays to define several types of materials including as limiting cases, elastic ($\beta = 0$) and viscous elements ($\beta = 1$). More precisely, a simple spring corresponds to $\beta = 0$ and $\dfrac{d^\beta f}{dt^\beta} = \dfrac{d^0 f}{dt^0} = f$; whilst the case of $\beta = 1$ corresponds to a first order derivative, i.e. $\dfrac{d^\beta f}{dt^\beta} = \dfrac{df}{dt} = \dot{f}$, which is a Newtonian dashpot.

2.2. Constitutive relation for isotropic power-law hereditariness

The extension of the constitutive relation presented in section 2.1 and tensorial strain/stress state are discussed in this section by means of effect superposition.

Let us consider a 2nd-order stress tensor $\boldsymbol{\sigma}$ with component σ_{ij} represented in fig.(4) with the symmetries $\sigma_{ij} = \sigma_{ji}$ for $i \neq j$.

In the following we introduce the Voigt representation of the state variables of the material in terms of vector representation of stress and strains tensors as:

$$\boldsymbol{\sigma}^T(t) = [\sigma_{11}(t)\,\sigma_{22}(t)\,\sigma_{33}(t)\,\sigma_{32}(t)\,\sigma_{31}(t)\,\sigma_{12}(t)] \tag{4}$$

$$\boldsymbol{\varepsilon}^T(t) = [\varepsilon_{11}(t)\,\varepsilon_{22}(t)\,\varepsilon_{33}(t)\,2\varepsilon_{32}(t)\,2\varepsilon_{31}(t)\,2\varepsilon_{12}(t)] \tag{5}$$

where t is the current time and the mixed index stress and strain components, namely $\sigma_{ij}(t)$ and $\varepsilon_{ij}(t)$ with $i \neq j$ denote shear stress and strain, respectively. Let us assume that $\sigma_{ij}(t) = \delta_{ij}$ and let us consider a single normal stress $\sigma_{ii} = 1$ for (i = 1,2,3).

In such a context the evolution of the strain $\varepsilon_{ii}(t)$ along the stress direction $\sigma_{ii}(t)$ and in the orthogonal planes reads:

$$\varepsilon_{ii}(t) = J_L(t)\,\sigma_{ii} = J_L(t) \tag{6a}$$

$$\varepsilon_{kk}(t) = \varepsilon_{jj}(t) = -J_v(t)\,\sigma_{ii} \tag{6b}$$

with $i \neq j \neq k$ and $i,\, j,\, k = 1, 2, 3$. In eqs.(6a-b) $J_L(t)$ and $J_v(t)$ are the axial and the transverse creep functions with respect to the stress direction, respectively. Under the assumption of smooth load process $\sigma_{ij}(t)$ the presence of contemporaneous stress $\sigma_{ij}(t) = \sigma_{ij}(t)\delta_{ij}$, with $i = 1, 2, 3$, may be accounted for by the integral

$$\varepsilon_{ii}(t) = \int_0^t J_L(t-\tau)\,\dot{\sigma}_{ii}(\tau) - J_v(t-\tau)\,[\dot{\sigma}_{jj}(\tau) + \dot{\sigma}_{kk}(\tau)]\,d\tau \tag{7}$$

with $i \neq j \neq k$ and i,j,k=1,2,3, respectively.

In the context of material isotropy shear strains $2\varepsilon_{ij}(t)$, $(i \neq j)$, are not involved by the axial stress $\sigma_{ii}(t)$, but only by the shear stress as $\sigma_{ij}(t)$ with $i \neq j$. The evolution of the shear strain $2\varepsilon_{ij}(t)$ due to a generic shear stress history $\sigma_{ij}(t)$ may be obtained by superposition integrals by means of the shear creep function $J_T(\cdot)$ as:

$$2\varepsilon_{ij}\left(t\right) = \int_0^t J_T\left(t-\tau\right)\dot{\sigma}_{ij}\left(\tau\right)d\tau \tag{8}$$

with $i \neq j$ and $i,j = 1,2,3$. The constitutive equations reported in eqs.(7),(8) may be reported in Voigt notation as:

$$\boldsymbol{\varepsilon}\left(t\right) = \int_0^t \mathbf{J}\left(t-\tau\right)\dot{\boldsymbol{\sigma}}\left(\tau\right)d\tau \tag{9}$$

where $\mathbf{J}(t)$ is the creep functions matrix that is described as:

$$\mathbf{J}\left(t\right) = \begin{bmatrix} \mathbf{J}^{(A)}\left(t\right) & \mathbf{0} \\ \mathbf{0} & \mathbf{J}^{(T)}\left(t\right) \end{bmatrix} \tag{10}$$

where the elements of the axial creep matrix $\mathbf{J}^{(A)}(t)$ are:

$$J_{ij}^{(A)}\left(t\right) = J_L\left(t\right)\delta_{ij} - \left(1-\delta_{ij}\right)J_\upsilon\left(t\right) \tag{11}$$

with $i,j = 1,2,3$. The shear creep matrix $\mathbf{J}^{(T)}(t)$ is a diagonal matrix gathering the shear creep functions $J_T\left(t\right)$ as:

$$J_{ij}^{(T)}\left(t\right) = J_T\left(t\right)\delta_{ij} \tag{12}$$

The three creep functions $J_L(t)$,$J_\upsilon(t)$ and $J_T(t)$ are related by a linear relation that reads:

$$J_T(t) = 2J_L(t) - J_\upsilon(t) \tag{13}$$

that may be obtained, with straightforward manipulations, by introducing a shear stress state $\sigma_{ij}(t)$ that involves a shear strain state under isotropy assumption, namely $\gamma_{ij} = 2\varepsilon_{ij}\left(t\right)$, and as evaluating the elongation and the stress along the principal axes at angles of $\pi/4$.

Under the assumption of linear elasticity, the creep functions coincide with the material compliance, which reads $J_T = 1/G$, $J_L = 1/E$ and $J_\upsilon = \upsilon/E$. After substitution in Eq.(13), this yields:

$$\frac{1}{G} = 2\left(\frac{1}{E} + \frac{\upsilon}{E}\right) = \frac{2\left(1+\upsilon\right)}{E} \tag{14}$$

that is the well-known relation among elasticity moduli.

Knowledge of the creep function matrix $\mathbf{J}(t)$ in Eq.(10) allows for the definition of the relaxation matrix $\mathbf{G}(t)$ by means of the coniugation relation as:

$$\hat{\mathbf{G}}(s)\hat{\mathbf{J}}(s) = \frac{1}{s^2}\mathbf{I} \tag{15}$$

where $\mathbf{I}$ is the identity matrix and $\hat{\boldsymbol{G}}(s)$, $\hat{\boldsymbol{J}}(s)$ are the Laplace transforms of the relaxation $\mathbf{G}(t)$ and the creep functions $\mathbf{J}(t)$ matrices.

With straightforward manipulations of Eq.(15) and inverse Laplace transform, the relaxation matrix may be written as:

$$\mathbf{G}\left(t\right) = \begin{bmatrix} \mathbf{G}^{(A)}\left(t\right) & \mathbf{0} \\ \mathbf{0} & \mathbf{G}^{(T)}\left(t\right) \end{bmatrix} \tag{16}$$

where:

$$G_{ij}^{(A)}\left(t\right) = \mathcal{L}^{-1}\left[\frac{1}{s^2\left(\hat{J}_L + \hat{J}_v\right)\left(\hat{J}_L - 2\hat{J}_v\right)}\right]\left[\left(\hat{J}_L - \hat{J}_v\right)\delta_{ij} + \left(1-\delta_{ij}\right)\hat{J}_v\right] \tag{17a}$$

$$G_{ij}^{(T)}\left(t\right) = \mathcal{L}^{-1}\left[\frac{1}{s^2\left(\hat{J}_L + \hat{J}_v\right)}\right]\delta_{ij} \tag{17b}$$

Eqs.(17a),(17b) show that in the presence of material fading memory, the relaxation matrix $\mathbf{G}(t)$ is obtained as a combination of creep functions relative to uniaxial creep tests. Similar considerations may be also withdrawn from the observation that in uniaxial relaxation tests, the relaxation function $G_L(t)$ is obtained in lateral free conditions, that is the strain state involves $\varepsilon_{11} \neq \varepsilon_{22} \neq 0$ and $\varepsilon_{33} = 1$ and measuring only $\sigma_{33}(t) = G_L\left(t\right)$ relaxation with $\sigma_{11} = \sigma_{22} = 0$.

Knowledge of the relaxation matrix of the material $\mathbf{G(t)}$ allows to evaluate the stress vector as:

$$\sigma\left(t\right) = \int_0^t \mathbf{G}\left(t-\tau\right)\dot{\varepsilon}\left(\tau\right)d\tau \tag{18}$$

The longitudinal shear and transverse relaxation functions $G_T(t)$,$G_L(t)$ and

$G_\upsilon(t)$ are linearly related by an equation that is similar to the one involving creep functions in Eq.(13), reading:

$$G_T(t) = \frac{1}{2}\left(G_L(t) - G_\upsilon(t)\right). \tag{19}$$

The latter allows for the evaluation of the transverse relaxation $G_\upsilon(t)$, as:

$$G_\upsilon(t) = 2G_T(t) - G_\upsilon(t) \tag{20}$$

In the following section, we derive the thermodynamic restrictions among the material parameters used in power-law representation of isotropic material hereditariness.

2.3. Power-law isotropic hereditariness: Thermodynamic restrictions

Let us assume that relaxation functions in laterally restrained axial and torsion shear tests may be captured, respectively, by power-laws with different order ($\alpha \neq \beta$) as:

$$G_L(t) = G_L^{(\alpha)} t^{-\alpha} + \bar{G}_L; \quad G_T(t) = G_T^{(\beta)} t^{-\beta} + \bar{G}_T \tag{21a}$$

$$G_\upsilon(t) = 2\left(G_T{}^{(\beta)} t^{-\beta} + \bar{G}_T\right) - \left(G_L{}^{(\alpha)} t^{-\alpha} + \bar{G}_L\right) \tag{21b}$$

with Eq.(21b) obtained from the application of Eq.(16). The physical dimensions of the coefficients are $[C_L] = [C_T] = F/L^2$, $\left[C_L^{(\alpha)}\right] = \dfrac{F}{L^2 T^{-\alpha}}$, $\left[C_L^{(\alpha)}\right] = \dfrac{F}{L^2 T^{-\beta}}$.

The expressions of the relaxation functions in Eqs.(21a),(21b) yield the relaxation matrix of the material in Eq.(16), with elements in the block matrices $\mathbf{G}^{(\mathbf{A})}(\mathbf{t})$ and $\mathbf{G}^{(\mathbf{T})}(\mathbf{t})$ reading:

$$G_{ij}^{(A)} = G_L(t)\,\delta_{ij} + (1 - \delta_{ij})\,G_\upsilon(t) \tag{22a}$$

$$G_{ij}^{(T)}(t) = G_T(t)\,\delta_{ij} \tag{22b}$$

We see that the relaxation matrix involves elements decaying with different power-laws of order β and α ($\alpha, \beta \in [0, 1]$).

The coefficients and parameters involved in the power-law descriptions of the material relaxation, namely, $G_L(t)$, $G_v(t)$ and $G_T(t)$ are related by thermodynamical restrictions to ensure the requirement of positive entropy rate increment [28]. Indeed, a dissipative simple solid is defined only if the restrictions:

$$\mathbf{G}\left(0\right) \geq \mathbf{G}\left(\infty\right) \geq 0 \tag{23}$$

$$\dot{\mathbf{G}}\left(0\right) \geq 0 \tag{24}$$

are fulfilled by the relaxation matrix of the material as reported in basic references on material hereditariness [29, 30, 31, 32].

Eqs.(23,24) are always satisfied by assuming positive values of the coefficients $\bar{G}_L$, $\bar{G}_T$ and $G_L^{(\alpha)}$ and $G_T^{(\beta)}$, whereas Eq.(25) alone is satisfied as the eigenvalues of the first derivative of the matrix, namely, $\dot{\mathbf{G}}\left(0\right)$ are all negative. This requirement may be verified by introducing a one-parameter family of relaxation matrices defined on a real parameter δ as $\mathbf{G}_\delta(t) = \mathbf{G}(t+\delta)$, and by studying the behavior of $\dot{\mathbf{G}}_\delta(t)$ for the limiting case $\delta \to 0$.

The parameter-dependent family of matrices $\dot{\mathbf{G}}_\delta(t)$ is defined as:

$$\dot{\mathbf{G}}_\delta\left(t\right) = \begin{bmatrix} \dot{\mathbf{G}}_\delta^{(A)}\left(t+\delta\right) & \mathbf{0} \\ \mathbf{0} & \dot{\mathbf{G}}_\delta^{(T)}\left(t+\delta\right) \end{bmatrix} \tag{25}$$

where the elements read:

$$\dot{G}_\delta^{(A)}\left(t+\delta\right) = -G_L^{(\alpha)}\alpha(t+\delta)^{-\alpha-1} \tag{26a}$$

$$\dot{G}_\delta^{(T)}\left(t+\delta\right) = -G_T^{(\beta)}\beta(t+\delta)^{-\beta-1} \tag{26b}$$

$$\dot{G}_\delta^{(v)}\left(t+\delta\right) = -2G_T^{(\beta)}\beta(t+\delta)^{-\beta-1} + G_L^{(\alpha)}\alpha(t+\delta)^{-(\alpha+1)} \tag{26c}$$

Observe that the one-parameter family $\dot{\mathbf{G}}_\delta(t)$ tends to the limit:

$$\lim_{\delta\to 0} \dot{\mathbf{G}}_\delta\left(t\right) = \dot{\mathbf{G}}\left(t\right) \tag{27}$$

We can infer the behavior of $\dot{\mathbf{G}}(t)$ from that of $\mathbf{G}_\delta(t)$, and by letting $\delta \to 0$. In this regard, the requirement in Eq.(24) may be recast as:

$$-\dot{G}\left(0\right) = -\lim_{\delta\to 0} \dot{G}_\delta\left(t\right) \geq 0 \tag{28}$$

that is we evaluate the eigenvalues $\lambda_i(\delta)$ $(i = 1, 2, ...6)$ of the matrix $\dot{\mathbf{G}}_\delta(0)$ and with the additional constraints $-\lambda_i(\delta) \geq 0$ $(i = 1, 2, ...6)$ as $\delta \to 0$.

The evaluation of the eigenvalues $\lambda_i(\delta)$ gives:

$$-\lambda_1(\delta) = -\lambda_2(\delta) = -2\left(\dot{G}_L(\delta) - \dot{G}_T(\delta)\right) \geq 0 \tag{29a}$$

$$-\lambda_3(\delta) = -\lambda_4(\delta) = -\lambda_5(\delta) = -\dot{G}_T(\delta) \geq 0 \tag{29b}$$

$$-\lambda_6(\delta) = -4\dot{G}_T(\delta) + \dot{G}_L(\delta) \geq 0 \tag{29c}$$

Substitution of Eq.(26a),(26b) into Eq.(29b) shows that the inequality is fulfilled for $C_T^{(\beta)} \geq 0$ and $0 \leq \beta \leq 1$. The inequalities (29a),(29c) read, after substitution:

$$\alpha G_\alpha \delta^{-(\alpha+1)} - \beta G_\beta \delta^{-(\beta+1)} \geq 0 \tag{30a}$$

$$4\beta G_\beta \delta^{-(\beta+1)} - \alpha G_\alpha \delta^{-(\alpha+1)} \geq 0 \tag{30b}$$

that, after some straightforward manipulation, may be cast in a more suitable form, taking natural logarithms as:

$$\ln(A_{\alpha\beta}) \geq (\alpha - \beta)\ln\delta \tag{31a}$$

$$\ln\left[\frac{(A_{\alpha\beta})}{4}\right] \leq (\alpha - \beta)\ln\delta \tag{31b}$$

where $A_{\alpha\beta} = \alpha G_L^{(\alpha)} / \left(\beta G_T^{(\beta)}\right)$. Inequalities in eqs.(31a)(31b) must be fulfilled for any value of the parameter δ yielding that $\alpha = \beta$. Moreover, in this latter case the additional thermodynamical restriction holds true.

$$G_T^{(\beta)} \leq C_L^{(\beta)} \leq 3C_T^{(\beta)}. \tag{32}$$

In passing, we observe that the condition $\alpha = \beta$ holds true only for the two terms (or one term) description of the relaxation function in Eq.(22a). Indeed, as we assume that the relaxation functions $G_L(t)$ and $G_T(t)$ involve linear combinations of power-laws as:

$$G_L(t) = \sum_{j=1}^{n} G_L^{(\alpha_j)} t^{-\alpha_j}; \quad G_T(t) = \sum_{i=1}^{m} G_T^{(\beta_i)} t^{-\beta_i} \tag{33}$$

with n and m the number of power-laws involved. Under such circumstances, the thermodynamical arguments proposed in this study yield the same conditions among the order of the power-laws as:

$$\max_{j=1,N}(\alpha_j) = \max_{i=1,M}(\beta_j) \tag{34a}$$

$$\min_{j=1,N}(\alpha_j) = \min_{i=1,M}(\beta_j) \tag{34b}$$

Substitution of Eq.(22a),(22b) into the constitutive equations for the three-axial hereditariness yields a relation among the stress vector and the history of the strain vector $\boldsymbol{\varepsilon}(t)$ as:

$$\boldsymbol{\sigma}(t) = \mathbf{G}_\beta \int_0^t (t-\tau)^{-\beta} \dot{\boldsymbol{\varepsilon}}(\tau)\, d\tau + \bar{\mathbf{G}} = \mathbf{G}_\beta \left(D_{0^+}^{\beta} \boldsymbol{\varepsilon} \right)(t) + \bar{\mathbf{G}} \tag{35}$$

where we have embraced the Voigt representation of the relaxation tensor $\mathbf{G}(\mathbf{t})$ in matrix form and we have used the notation:

$$\mathbf{G}(t) = \mathbf{G}_\beta \frac{t^{-\beta}}{\Gamma(1-\beta)} + \bar{\mathbf{G}} \tag{36}$$

with the matrices:

$$\mathbf{G}_\beta(t) = \begin{bmatrix} G_\beta^{(L)} & G_\beta^{(v)} & G_\beta^{(v)} & 0 & 0 & 0 \\ G_\beta^{(v)} & G_\beta^{(L)} & G_\beta^{(v)} & 0 & 0 & 0 \\ G_\beta^{(v)} & G_\beta^{(v)} & G_\beta^{(L)} & 0 & 0 & 0 \\ 0 & 0 & 0 & G_\beta^{(T)} & 0 & 0 \\ 0 & 0 & 0 & 0 & G_\beta^{(T)} & 0 \\ 0 & 0 & 0 & 0 & 0 & G_\beta^{(T)} \end{bmatrix} \tag{37a}$$

$$\overline{\mathbf{G}} = \begin{bmatrix} \bar{G}_L & \bar{G}_v & \bar{G}_v & 0 & 0 & 0 \\ \bar{G}_v & \bar{G}_L & \bar{G}_v & 0 & 0 & 0 \\ \bar{G}_v & \bar{G}_v & \bar{G}_L & 0 & 0 & 0 \\ 0 & 0 & 0 & \bar{G}_T & 0 & 0 \\ 0 & 0 & 0 & 0 & \bar{G}_T & 0 \\ 0 & 0 & 0 & 0 & 0 & \bar{G}_T \end{bmatrix} \tag{37b}$$

The stress vector obtained as a functional of the strain vector $\boldsymbol{\varepsilon}(t)$ in Eq.(35) is the generalization of the constitutive equation reported in Eq.(3a) under the assumption of material isotropy.

In the next section the multiaxial fractional-order hereditariness will be further discussed by introducing a mechanical hierarchy that yields the constitutive model reported in Eq.(35)

3. Exact mechanical description of fractional-order isotropic hereditariness

The stress/strain tensor outlined in section (2) requires a multiaxial constitutive relation, as in Eq.(35), that under the assumption of $\bar{\mathbf{G}} = 0$ generalizes Eq.(3a).

The rheological element, namely the springpot, corresponding to Eq.(3a) can not, however, be defined also for the isotropic description in Section (2), namely for the presence of shear stress/strain. A mechanical model that may be involved in the presence of normal and shear stress to be used in experimental test is represented in Fig.5

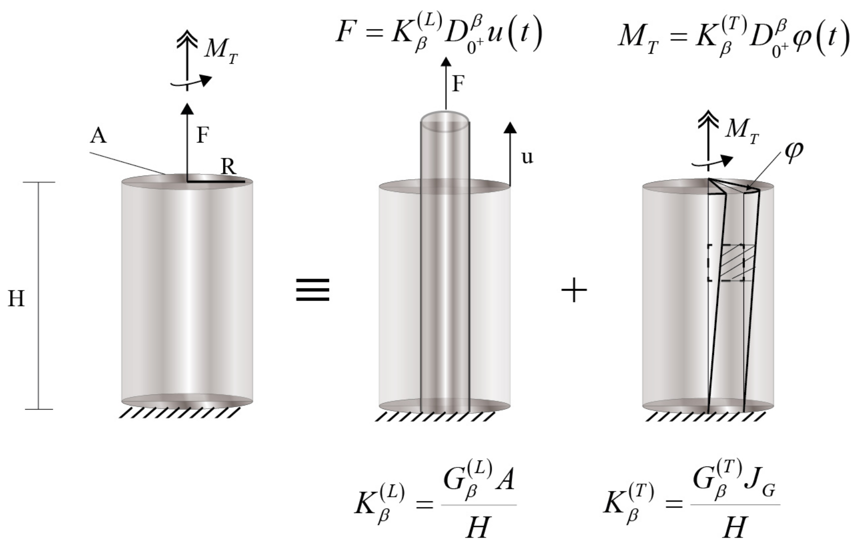


Figure 4: Rheologic elements

Under such conditions, the circular column of height H, cross section A and radius R under axial stress and shear stress related to the measured relative displacements $u(t)$ and twist angle $\varphi(t)$ provides these equilibrium equations:

$$
\begin{aligned}
F &= K_{\beta}^{(L)}(D_{0+}^{\beta} u)\,(t) \\
M_T &= K_{\beta}^{(T)}(D_{0+}^{\beta} \varphi)\,(t)
\end{aligned}
\tag{38}
$$

where $J_G = \pi R^4/4$ is the polar moment of inertia of the circular cross-section represented in Fig.5. The constitutive equation(38) involve for limiting cases: i) a linear elastic spring ($\beta = 0$); and ii) a linear viscous element ($\beta = 1$), respectively.

In the following, we introduce a hierarchic mechanical model to capture the axial and shear hereditariness assuming power-law description of the creep and relaxation functions for axial and shear stress/strain, respectively [33, 17, 34, 16]. The obtained mechanical hierarchy corresponds exactly to an axial and shear springpots with the same order of time evolution/decay.

To this aim let us introduce an elastic column of unbounded length with circular cross section of radius R. The elastic features of the column are non-costant along the column axis and vary with the coordinate as:

$$
E\,(z) = \frac{E_\alpha}{\Gamma\,(1-\alpha)} z^{-\alpha}; \quad G\,(z) = \frac{G_\alpha}{\Gamma\,(1-\alpha)} z^{-\alpha} \qquad -1 \le \alpha \le 1 \tag{39}
$$

The column is externally restrained by a set of torsional and axial viscous dashpots fig.(5) with non-homogeneous viscosity $\eta(z)$ as:

$$
\eta\,(z) = \frac{\eta_\alpha}{\Gamma\,(1+\alpha)} z^{-\alpha} \qquad -1 \le \alpha \le 1 \tag{40}
$$

Axial and torsional equilibrium along the column axis reads:

$$
\frac{\eta_\alpha}{\Gamma\,(1+\alpha)} z^{-\alpha} 2\pi R \Delta z \dot{u}\,(z,t) = \frac{E_\alpha \pi R^2 s (z+\Delta z)^{-\alpha}}{\Gamma\,(1-\alpha)} \left[u\,(z+\Delta z,t) - u\,(z,t)\right] +
$$
$$
+ \frac{E_\alpha \pi R^2 s z^{-\alpha}}{\Gamma\,(1-\alpha)} \left[u\,(z,t) - u\,(z-\Delta z,t)\right] \tag{41}
$$

$$
\frac{\eta_\alpha}{\Gamma\,(1+\alpha)} z^{-\alpha} 2\pi R^2 \Delta z \dot{\varphi}\,(z,t) = G_\alpha \pi R^4 (z+\Delta z)^{-\alpha} \left[\varphi\,(z+\Delta z,t) - \varphi\,(z,t)\right] +
$$
$$
+ G_\alpha \pi R^4 (z+\Delta z)^{-\alpha} \left[\varphi\,(z,t) - \varphi\,(z-\Delta z,t)\right] \tag{42}
$$

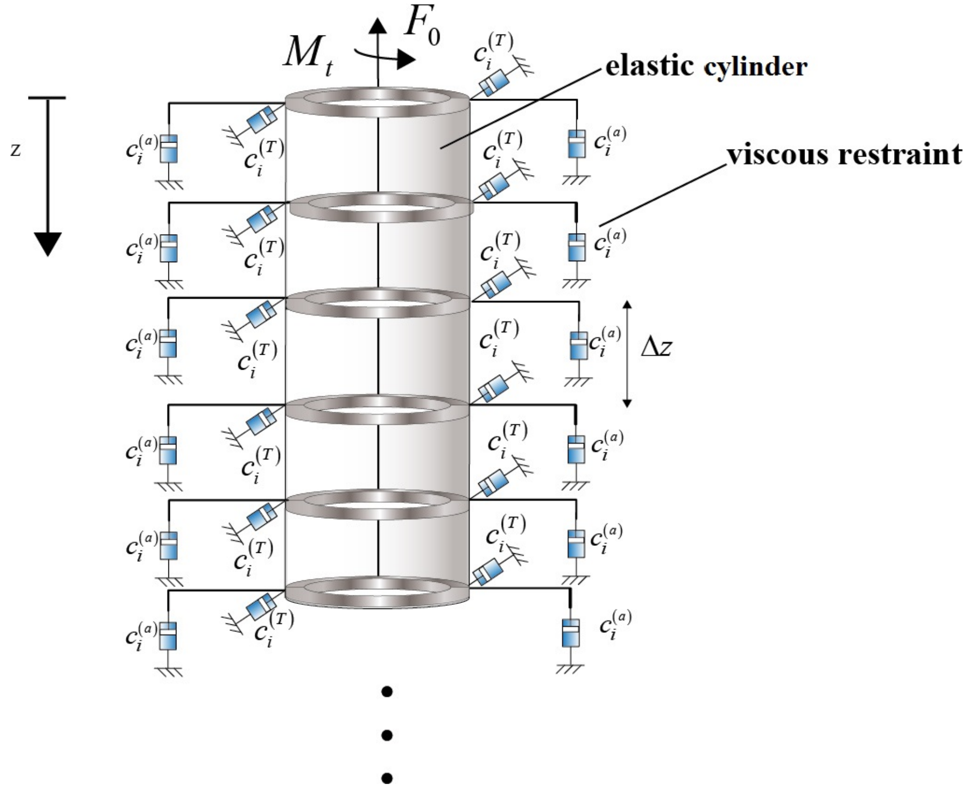


Figure 5: column with non-homogeneous viscosity

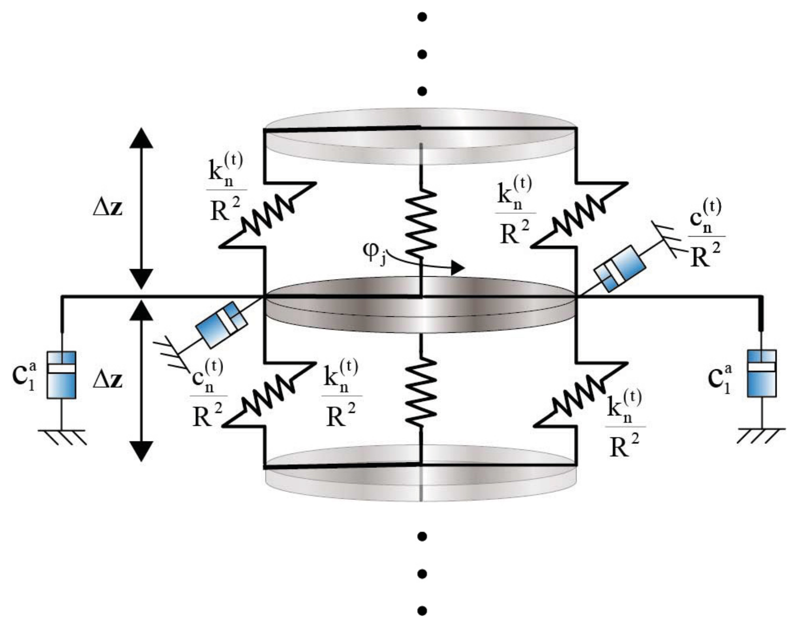


Figure 6: elements of the column with non-homogeneous viscosity

that, can be rewritten in differential form, by letting $\Delta z \to 0$ as:

$$\frac{\eta_\alpha z^{-\alpha}}{\Gamma(1+\alpha)} \frac{\partial u(z,t)}{\partial t} = \frac{E_\alpha R s}{\Gamma(1-\alpha)} \frac{\partial}{\partial z}\left(z^{-\alpha}\frac{\partial u(z,t)}{\partial z}\right) \tag{43a}$$

$$\frac{\eta_\alpha z^{-\alpha}}{\Gamma(1+\alpha)} \frac{\partial \varphi(z,t)}{\partial t} = \frac{G_\alpha R}{\Gamma(1-\alpha)} \frac{\partial}{\partial z}\left(z^{-\alpha}\frac{\partial \varphi(z,t)}{\partial z}\right) \tag{43b}$$

The boundary conditions involving the differential fields $u(z,t)$ and $\varphi(z,t)$ in Eqs.(43a),(43b) read, respectively.

$$\lim_{z\to\infty} u(z,t) = 0 \tag{44a}$$

$$\lim_{z\to 0} \frac{E_\alpha}{\Gamma(1-\alpha)} z^{-\alpha}\frac{\partial u}{\partial z} = F_0 \tag{44b}$$

$$\lim_{z\to\infty} \varphi(z,t) = 0 \tag{45a}$$

$$\lim_{z\to 0} \frac{G_\alpha}{\Gamma(1-\alpha)} z^{-\alpha}\frac{\partial \varphi}{\partial z} = M_0 \tag{45b}$$

Mathematical operators and boundary conditions in Eqs.(46a,b) are completely equivalent to those of a previous differential problem that has been solved by resorting to a non- linear mapping followed by Laplace transforms [17, 35]. Such a procedure yields a Bessel differential equation of second kind in terms of the anomalous Laplace parameters. The position of the boundary conditions and inverse Laplace transform provides the solution in the form:

$$u_0\left(t\right) = u_0\left(z,t\right) = \lim_{z\to\infty} u\left(z,t\right) = \frac{t^{-\beta}}{k_\beta^{(L)}} F_0 = J_L\left(t\right) F_0 \tag{46}$$

$$\varphi_0\left(t\right) = \varphi_0\left(z,t\right) = \lim_{z\to\infty} \varphi\left(z,t\right) = \frac{t^{-\beta}}{k_\beta^{(T)}} M_0 = J_T\left(t\right)_0 \tag{47}$$

with:

$$k_\beta^{(L)} = \frac{\Gamma\left(2\beta\right)\left(\tau_L^\beta\right)}{E_\alpha 2^{1-2\beta}\Gamma\left(\beta\right)\Gamma\left(1-\beta\right)} \tag{48}$$

$$k_\beta^{(T)} = \frac{\Gamma\left(2\beta\right)\left(\tau_L^\beta\right)}{G_\alpha 2^{1-2\beta}\Gamma\left(\beta\right)\Gamma\left(1-\beta\right)} \tag{49}$$

with $\beta = \frac{1+\alpha}{2}$ and the relaxation times:

$$\tau_L = \frac{\eta_\alpha}{E_\alpha}\frac{\Gamma\left(1-\alpha\right)}{\Gamma\left(1+\alpha\right)} \tag{50}$$

$$\tau_T = \frac{\eta_\alpha}{G_\alpha}\frac{\Gamma\left(1-\alpha\right)}{\Gamma\left(1+\alpha\right)} \tag{51}$$

The Superposition principle provides, by resorting to the fundamental equations of linear viscoelasticity, the constitutive equations of the macroscopic variables, as:

$$F_0\left(t\right) = k_\beta^{(L)}\left(D_{0^+}^\beta u_0\right)\left(t\right) \tag{52}$$

$$M_T\left(t\right) = k_\beta^{(T)}\left(D_{0^+}^\beta \varphi_0\right)\left(t\right) \tag{53}$$

Eqs.(52),(53) are the constitutive equation at the macro-scale and, by recalling that $F_0 = \sigma_{33}A$ and $|\tau| = \sqrt{|t_{31}|^2 + |t_{32}|^2} = \frac{M_0}{2As}$, the constitutive equations of the material read:

$$\sigma_{33} = G_\beta^{(L)}\left(D^\beta \varepsilon_{33}\right)\left(t\right) \tag{54}$$

$$|\tau| = G_\beta^{(T)} \left(D^\beta |\gamma|\right)(t) \tag{55}$$

where the coefficients $G_\beta^{(L)}$ and $G_\beta^{(T)}$ read:

$$G_\beta^{(L)} = \frac{\bar{k}_\beta^{(L)} \bar{l}}{A} \qquad G_\beta^{(T)} = \frac{\bar{k}_\beta^{(T)}}{2As} \frac{R}{\bar{l}} \tag{56}$$

and where$\bar{l}$ is an internal length of the material. Eqs.(54),(55) are the multiaxial constitutive relations of the isotropic material and, henceforth, correspond to the hierarchy introduced by the fractional-order isotropy.

4. Conclusions

The mathematical structure of the fractional-order isotropic hereditariness has been discussed in this paper. The study has been framed in the context of biomimetic ceramics used in cranioplasty neurosurgery (i.e. CustomBone® ”prosthesis”). The creep and relaxation functions of isotropic linear hereditarinnes have been particularized for power-law decays, yielding a multi-axial constitutive model in terms of fractional-order operators. Additionally, a specific mechanical model has been introduced, which corresponds to the fractional-order isotropic hereditariness. In future studies experimental campaigns involving creep and relaxation of biomimetic ceramics will be reported to assess the validity of material isotropy. Additionally, the proposed hierarchy will be further extended to deal with non-linear hereditariness as those observed in creep and relaxations of tendons and ligaments.

Acknowledgments:

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Response to Reviewer Comments

Manuscript number: NLM2018123

Manuscript title: Power-Laws hereditariness of biomimetic ceramics for cranioplasty neurosurgery

The authors wish to thank the editors and reviewers for their time in effort in reviewing our manuscript. We hope the changes listed have made the manuscript suitable for publication and we look forward to your response.

1 Reviewer 1's Comments

In this paper the authors provide a study of the hereditary behavior of hydroxyapatite-based materials which are among the composites used for cranioplastic surgery. The study is per se interesting and insightful. Hence, I believe it deserves to be published once some major issues have been addressed.

Reviewer 1 Comment 1

First of all, from a linguistic perspective, the manuscript requires a certain degree of revision as it presents several typos and syntactically odd sentences such as Additionally after the introduction of the mathematical model has been introduced to generalize the uniaxial mechanical hierarchy previous introduced to deal with 1D material hereditaniness. Besides, many articles and connectors appear to be missing throughout the paper, so I invite the authors to read carefully the manuscript and improve the exposition.

Response

We thank the reviewer for this comment. In the manuscript, syntax changes were made such as: Additionally, the same relations of the 1D case for the material hereditariness has been introduced in the mathematical model to generalize the uniaxial mechanical hierarchy.

Reviewer 1 Comment 2

From a terminological point of view, the authors refer to well-known operators in a very unusual way, which is completely inconsistent with the typical notation used by the fractional calculus community. For example, the authors use the term fractional-order calculus instead of fractional calculus and in Par. 105 they denote by Riemann integral and Leibniz derivative of non-integer orders two operators which are known in the community as the Riemann-Liouville fractional integral and the Caputo fractional derivative. This may generate confusion, therefore I ask the authors to comply with the standard notation. The quality of the figures displayed in the paper is extremely bad. It seems like if they where cut out from a different (low resolution) file and then pasted in the manuscript. Thus, the authors should fix this issue.

Response

We thank the reviewer for this comment. We changed the formal definition as suggested.

- The three-axial isotropic behavior is framed in the context of fractional calculus and same details about thermodynamical restrictions of memory functions used in the formulation of the three-axial isotropic constitutive equations.
- The introduction of power-laws in the description of creep and relaxations yields that the constitutive behavior of the material is expressed in terms of the so-called fractional calculus.
- That represent the generalization of the well-known Riemann-Liouville fractional integraland Caputo fractional derivative.

We we have increased the quality of the figures, in particular fig. 4, figure in pag. 16 and 17, fig.5, fig.6

Reviewer 1 Comment 3

While discussing the mathematical framework of fractional viscoelasticity the authors missed the following relevant reference: Mainardi, F., & Spada, G. (2011). Creep, relaxation and viscosity properties for basic fractional models in rheology. The European Physical Journal Special Topics, 193(1), 133-160. Thus, I invite the authors to quote this paper in Section 2 and add it to the list of references.

Response

We thank the reviewer for this comment. We introduce this relevant reference in line 282 of the References list.

Reviewer 1 Comment 4

Finally, in the conclusion the authors quote explicitly the CustomBone prosthesis. However, what I find really confusing is connected with the fact that the arguments presented in the paper work for several different materials beside the one used for the aforementioned prosthesis. Without some in-depth experimental studies of this specific system the first statement in the conclusion (beside requiring some linguistic improvements) might fall empty in meaning or sound more like a product placement rather than a scientific remark. I understand that the CustomBone prosthesis is one of the motivations of the study, but it should be referred to as such.

Response

We thank the reviewer for this comment. we clarified the point added a comment in the conclusion. We wrote:

The mathematical structure of the fractional-order isotropic hereditariness has been discussed in paper in the paper. The study has been framed in the context of biomimetic ceramics used cranioplasty neurosurgery (i.e.CustomBone® ”prosthesis”)

2 Reviewer 2's Comments

The authors study the time-dependent behavior of biomimetic ceramic materials used in patient-specific prosthesis. The morphology of the analyzed prosthesis, the Custombone, allow the three axial material behavior to be represented by means of an isotropic fractional-order model. The 3D constitutive behavior in creep and relaxation for power-laws best-fitting is discussed providing thermodynamical restrictions on model parameters. Further, a mechanical hierarchy that corresponds, exactly, to the used power-laws in creep and relaxation is discussed in the paper.

Reviewer 2 Comment 1

The use of power-laws with only one term in relaxation and creep functions for the longitudinal and shear modulus appears to limit the application of power-laws. Are there any other thermodynamical restrictions on model parameters, as linear combinations of power-laws is used to capture creep/relaxation?

Response

We thank the reviewer for this comment. We added a comment (from line 166 of the original manuscript), in order to clarify this point. We wrote:

In passing we observe that the condition $\alpha = \beta$ holds true only for the two terms (or one terms) description of the relaxation function in eq.(25 a).

Indeed, as assume that the relaxation functions $G_L(t)$ and $G_T(t)$ involve linear combinations of power-laws:

$$G_L(t) = \sum_{j=1}^{N} C_L^{(j)} t^{-\alpha_j}; G_T(t) = \sum_{i=1}^{M} C_T^{(i)} t^{-\beta_i} \tag{1}$$

with N and M the number of power-laws involved. The thermodynamical arguments proposed this study yields that:

$$\max_{j=1,N} (\alpha_j) = \max_{i=1,M} (\beta_j) \tag{2a}$$

$$\min_{j=1,N} (\alpha_j) = \min_{i=1,M} (\beta_j) \tag{2b}$$

Reviewer 2 Comment 2

The authors may update the reference list including recent applications of fractional-order calculus in materials engineering as well as bio-medical engineering:

- Ionescu C., Lopes A., Copota D., Machado J.A.T., Bates J.H.T., 2017, The role of fractional calculus in modeling biological phenomena: A review, Communications in Nonlinear Science and Numerical Simulation.
- Magin R.L., 2010, Fractional calculus models of complex dynamics in biological tissues, Computers & Mathematics with Applications.
- Sun H.G., Zhang Y., Baleanu D., Chen W., Chen Y.Q., 2018, A new collection of real world applications of fractional calculus in science and engineering, Communications in Nonlinear Science and Numerical Simulation.
- Caputo M., Cametti C., 2017, Fractional derivatives in the diffusion process in heterogeneous systems: The case of transdermal patches, Mathematical Biosciences.
- Alotta G., Di Paola M., Failla G., Pinnola F.P., 2018, On the dynamics of non-local fractional viscoelastic beams under stochastic agencies, Composites Part B/Engineering.
- Failla, G., 2017, Stationary response of beams and frames with fractional dampers through exact frequency response functions, Journal of Engineering Mechanics.

Response

We thank the reviewer for this comment. We have updated the reference list of the original manuscript.

3 Reviewer 3's Comments

"It looks good for publication in the current form. One comment is that they could justify why a linear elastic response is sufficient for this system, and do they have any ideas for the future on how to approach nonlinear elasticity"

Reviewer 3 Comment 1

Since the special issue has a special focus on nonlinear aspects, we kindly ask that elements of nonlinearity should be highlighted for future developments of the theory. Please address the points raised in your rebuttal and in your revision. Please show your changes using color.

Response

We thank the reviewer for this comment. We added a comment in the conclusion, in order to clarify this point. We wrote:

In future studies experimental campaigns involving creep and relaxation of biomimetic ceramics will be reported to assess the validity of material isotropy. Additionally, the proposed hierarchy will be further extended to deal with non-linear hereditariness as those observed in creep and relaxations of tendons and ligaments.

Paper Highlights

- Isotropic fractional-order hereditariness has been introduced for biomimetic ceramics
- Thermodynamic restrictions on the power-law exponents in axial and shear relaxation/creep functions have been provided
- A novel mechanical hierarchy corresponding contemporary to axial/shear creep and relaxation functions have been reported in the paper.

Power-Laws hereditariness of biomimetic ceramics for cranioplasty neurosurgery

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Abstract

We discuss the hereditary behavior of hydroxyapatite-based composites used for cranioplastic surgery in the context of material isotropy. We classify mixtures of collagen and hydroxiapatite composites as biomimetic ceramic composites with hereditary properties modeled by fractional-order calculus. We assune isotropy of the biomimetic ceramic is assumed and provide thermodynamic of restrictions for the material parameters. We exploit the proposed formulation of the fractional-order isotropic hereditariness further by means of a novel mechanical hierarchy corresponding exactly to the three-dimensional fractional-order constitutive model introduced.

Keywords: `Biomimetic materials,cranioplasty, fractional calculus, power-law hereditariness, isotropic hereditariness.`

2010 MSC: 00-01, 99-00

1. Introduction

Cranioplastic neurosurgery is widespread nowadays since it is necessary both in traumatic therapies or in the presence of specific oncologic pathology. Cranio-

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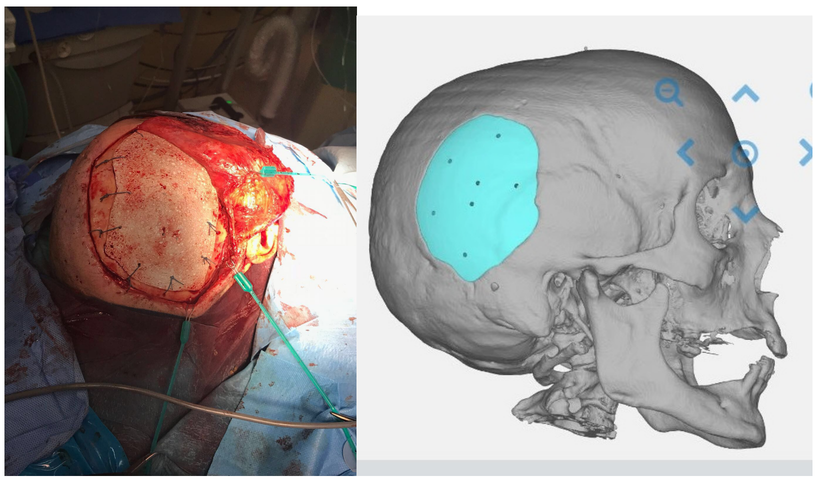
plasty is a surgical procedure that aims to re-establish the skull integrity following a previous craniotomy due to the occurrence of traumas, tumors and/or congenital malformations. In all cases cranioplasty can be considered as the conclusive action of a surgery initiated by the removal of a bone operculum see fig.1.



(a) (b)

Figure 1: cranioplasty surgery, Policlinico Giaccone Palermo

Ideally, cranioplasty procedures should provide restoration of the protective functions of the skull with maintenance of the original aesthetics and long-term mechanical performance [1]. The ideal material for cranioplasty should be chemically inert, biocompatible, biomechanically reliable, easily manufactured, individually shaped, safe, and able to promote osteoblast migration. Nowadays synthetic implants based on metallic (mainly titanium) or acrylic plates (mainly polymethylmeta-crylate or polyetheretherketone) are widely used in cranioplasty procedures. These are bioinert materials with good biocompatibility, resistance to infections, ease of sterilization, ability to be subjected to imaging diagnostics, and the capacity to undergo flexible design for adaptation to different clinical cases. They exhibit good mechanical strength, which offers adequate brain protection from external shocks. However, they present poor osteogenic and osteoconductive ability, thus resulting in a foreign body functioning as a shell expected to provide brain protection, but connected to the surrounding bone only by its perimeter contact surface. In order to overcome many limitations an Hydroxyapatite (HA)-based material has been widely considered for decades as the gold standard for bone scaffolds, as its composition is very close to that of bone mineral, thus exhibiting excellent biocompatibility, a low inflammatory reaction as well as good osteogenic ability and osteoconductivity. The hydrophilic character of HA favors cell attachment and tight adhesion of bone to the scaffold surface, which is a key target for the stability of the bone/implant interface. Therefore, HA scaffolds presenting wide, open and interconnected multiscale porosity can induce extensive bone ingrowth and penetration throughout the whole scaffold, partly thanks to the possibility of massive fluid perfusion, which triggers and assists neovascularization. Hence, cranial reconstruction using synthetic porous HA has recently become the subject of intense debate among surgeons, and it now represents a new concept in cranioplasty procedures. The custom-made concept was first applied to porous hydroxyapatite because of the need to overcome the fragility of the material itself. One of the advantages of HA-based prosthesis there is the important issue of customization.

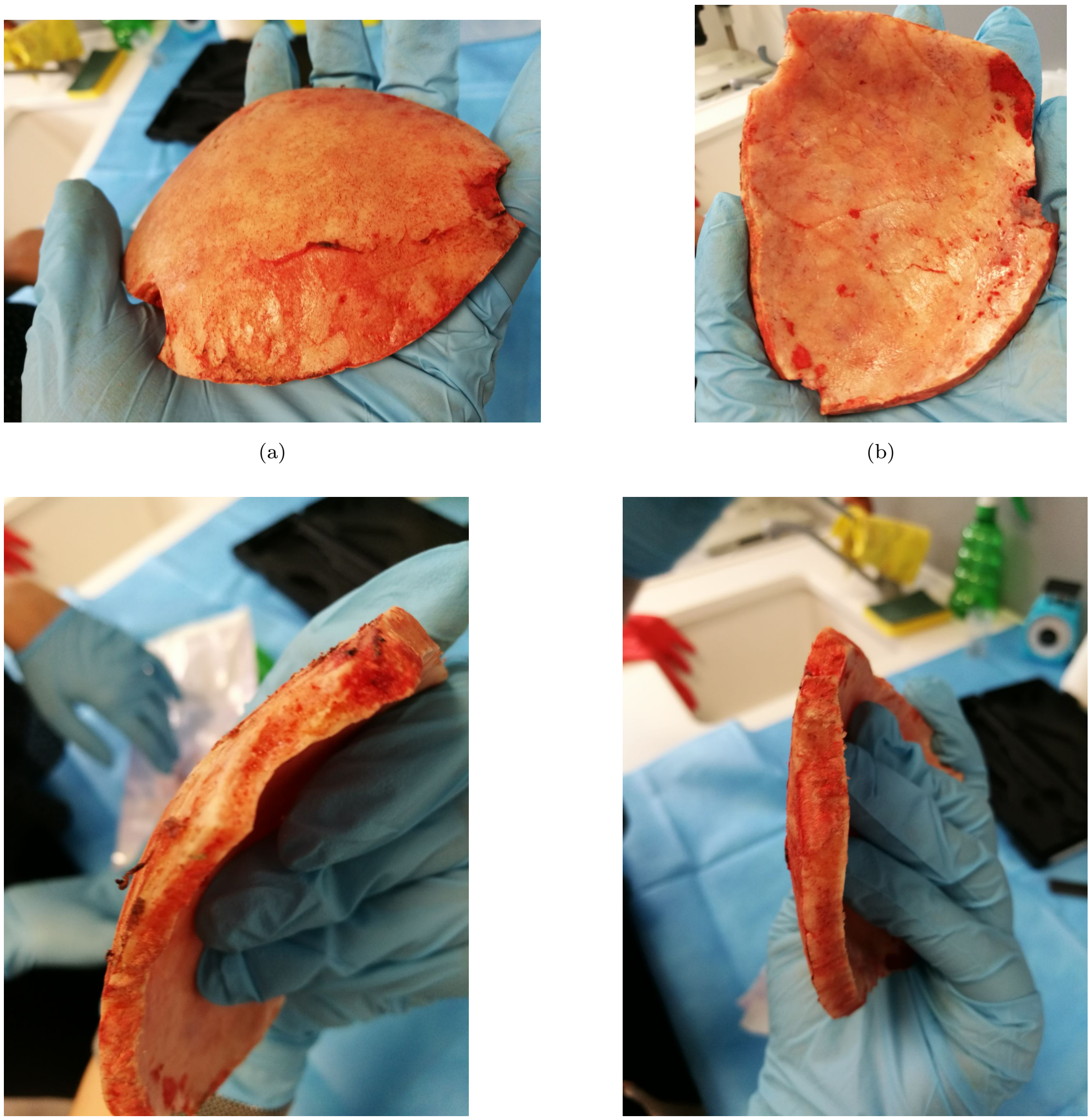
Indeed, in the presence of cranioplasty, the morphology of the bone to be replaced with a synthetic prosthesis must match completely the original bone to accelerate the osteointegration of the prosthesis [2, 3, 4] in the surgical hole. In fig.(2 a-d) a human parietal bone and its synthetic prosthesis fig.(3 a-c) have been obtained from a university neurosurgery hospital in Palermo. The synthetic bone used for replacement is a CustomBone® (Finceramica Faenza), namely custom-made, porous hydroxyapatite scaffolds with total porosity in the range of 60 to 70 % and pore architecture based on macro-pores (> 100 micron) interconnected with micro-pores (5-10 micron). CustomBone® scaffolds were obtained by reproduction of the patient's bone defect as modeled by 3D CT scan. They are made of a composite ceramics material obtained from chemical deposition of hydroxyiapatite with a small fraction of collagen type I (see fig.1a).

The use of biomimetic ceramics to replace cortical as well as trabecular bone is well- defined technique in bone surgery [5]. Indeed the mechanical features of the prosthesis in terms of elastic moduli and the strength of the biomimetic composite of integration are very similar. However, the use of ceramic materials to replace the bones of a human head may involve different behaviors in terms of energy dissipation. Indeed, biologic tissues show marked hereditariness due to the reptation of the collagen chains of the material as well as to the fiber recruitment in the tissues. Material hereditariness involves additional stresses that may be applied to the grafted ceramics prosthesis and may lead to fracture propagation during patient follow-up [6].

The hereditary properties of bone in uniaxial test are represented by creep $J(t)$ and relaxation $G(t)$ functions formulated in terms of power-law $J(t) \propto t^{\beta}$ and $G(t) \propto t^{-\beta}$ with $0 \leq \beta \leq 1$, yielding accurate description of experimental data [5, 7, 8, 9, 10]. Power-laws hereditariness in conjuction with Boltzmann superposition yields the constitutive behavior in terms of so-called fractional integrals and derivatives. Fractional calculus may be considered as a generalization of the classical differential calculus to real-order integration and differentiation $\left(i.e.df/dt \rightarrow d^{\beta}f/dt^{\beta} with \beta \in [0,1]\right)$ as reported in classical references [11, 12, 13, 14, 15, 16, 17, 18]. In such a context, uniaxial hereditariness [19, 20, 21, 22] involving fractional order stress-strain relations has been reported since the beginning of the 20th century [23, 16] defining the so-called springpot element [24, 25].

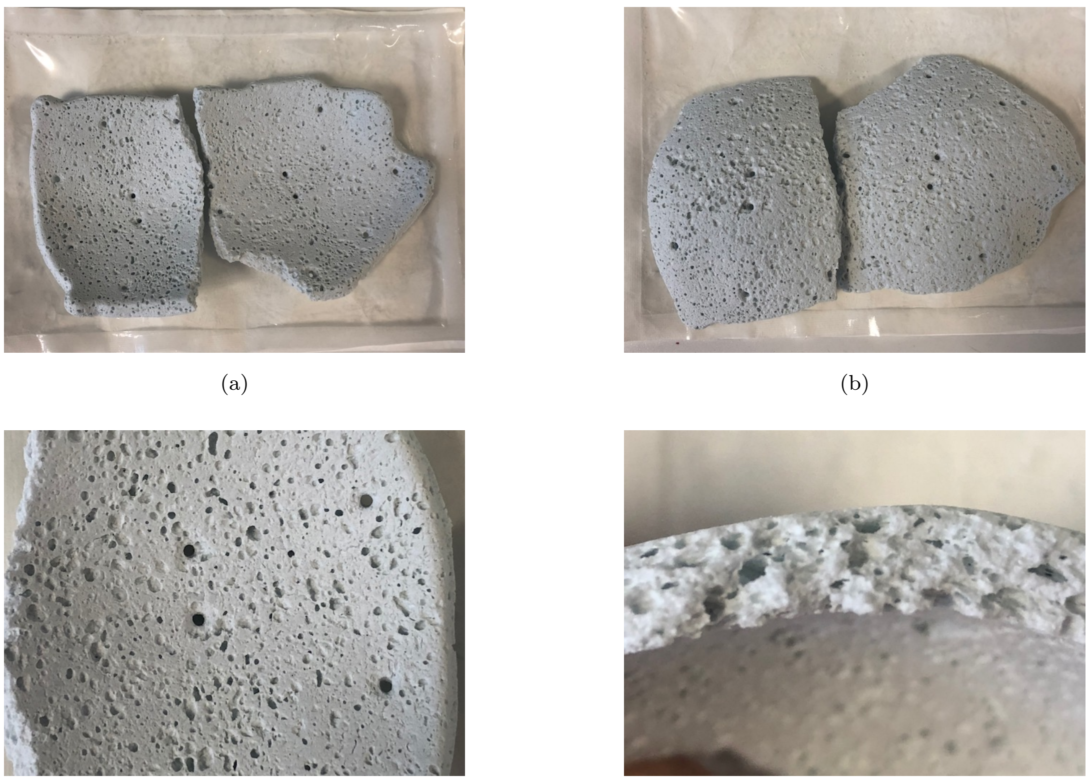
In the presence of tensorial stress/strain state, as in the continuum mechanics description of biomimetic prosthesis, no generalities have been reported in the scientific literature to capture multiaxial hereditariness with fractional-order calculus, to the best of the authors' knowledge. Indeed, in several case, recently discussed in the scientific literature [26, 27, 28], the use of power-laws without thermodynamic restrictions the parameters does not guarantee positive entropy rate for any strain/stress process involved by material.

In this paper, a 3D constitutive relation describing material hereditariness is



(a) (b) (c) (d)

Figure 2: human parietal bone, Policlino Giaccone Palermo



(a) (b) (c) (d)

Figure 3: CustomBone® prosthesis morphology

discussed in the context of power-laws functional classes of the relaxation/creep functions. We show that, under the assumption of material isotropy, thermodynamical restrictions on the constitutive parameters allow to formulate the constitutive behavior in terms of a Caputo fractional derivative that is formally analogous to the constitutive behavior in uniaxial state of stress/strain.

The paper is organized as follow: sec.2 provides generalities about fractional-order calculus and isotropic hereditariness; sec.3 reports a mechanical hierarchy that corresponds exactly to the isotropic fractional-order hereditariness. Sec.4 provides some conclusions about the proposed model of isotropic hereditariness and the influence on the mechanics of the biomimetic ceramics prosthesis.

2. Power-law hereditariness of isotropic biomimetic ceramics

In this section we outline the constitutive relations in the presence of power-law hereditariness, including the main arguments of power-law hereditariness under uniaxial stress/strain; generalization to the isotropic case and thermodynamic restrictions on the material parameters.

2.1. Uniaxial power-law hereditariness: The fractional order constitutive equation

We describe the constitutive behavior of materials in long-standing mechanical tests is described by means of the well-known creep and relaxation functions, dubbed $J(t)$ and $G(t)$, respectively. The linear superposition applied to a generic stress/strain history, namely $\sigma(\tau)$ and $\varepsilon(\tau)$ with $\tau \leq t$, yields:

$$\sigma(t) = \int_0^t G(t-\tau)d\varepsilon(\tau) = \int_0^t G(t-\tau)\dot{\varepsilon}(\tau)d\tau \tag{1a}$$

$$\varepsilon(t) = \int_0^t J(t-\tau)d\sigma(\tau) = \int_0^t J(t-\tau)\dot{\sigma}(\tau)d\tau \tag{1b}$$

Eqs.(1a, b) are defined in terms of Boltzman superposition with $d\sigma = \dot{\sigma}dt$ and $d\varepsilon = \dot{\varepsilon}dt$ increments, where $[\dot{\cdot}] = \frac{d}{dt}$. Creep and relaxation functions characterize the material behavior and they must satisfy the conjugation relation $\hat{J}(s)\hat{G}(s) = 1/s^2$, where s indicates the Laplace parameter and $\hat{f}(s) = \mathcal{L}[f(t)]$ is the Laplace transform of the generic function $f(t)$. In the context of materials hereditariness, power-law representation of creep and relaxation functions, i.e. $J(t)$ and $G(t)$, was introduced at the beginning of the last century [23],

$$G(t) = \frac{C_\beta}{\Gamma(1-\beta)} t^{-\beta}, \tag{2a}$$

$$J(t) = \frac{1}{C_\beta \Gamma(\beta+1)} t^{\beta} \tag{2b}$$

where $\Gamma(\cdot)$ is the Euler-Gamma function, $\beta \in [0,1]$ and $C_\beta > 0$, are material parameters, that may be estimated through a best-fitting procedure of experimental data [29, 30]. Straightforward manipulations show that the power-law functional class in eqs.(2a, b) satisfies the conjugation relation and it yields, upon substitution in eqs. (1a, b) the following constitutive relations:

$$\sigma(t) = \frac{C_\beta}{\Gamma(1-\beta)} \int_0^t (t-\tau)^{-\beta} \dot{\varepsilon}(\tau) d\tau = C_\beta \left(D_{0^+}^{\beta} \varepsilon \right)(t) \tag{3a}$$

$$\varepsilon(t) = \frac{1}{C_\beta \Gamma(\beta+1)} \int_0^t (t-\tau)^{\beta} \dot{\sigma}(\tau) d\tau = \frac{1}{C_\beta} \left(I_{0^+}^{\beta} \sigma \right)(t) \tag{3b}$$

in terms of the Caputo fractional derivative and Riemann-Liouville fractional integral, respectively.

Use of power-laws and, as a consequence, of fractional-order operators is usually connected, in a rheological context [28], to the introduction of the springpot element.

Springpot is a one-dimensional element defined in terms of two parameters, i.e. C_β and β, $0 \leq \beta < 1$ and $C_\beta > 0$ whose constitutive relation is reported in eqs.(3a,b). Such element with an intermediate behavior among elastic springs and viscous dashpot, is widely used nowadays to define several types of materials including as limiting cases, elastic ($\beta = 0$) and viscous elements ($\beta = 1$). More precisely, a simple spring corresponds to $\beta = 0$ and $\frac{d^\beta f}{dt^\beta} = \frac{d^0 f}{dt^0} = f$; whilst, case of $\beta = 1$ corresponds to a first order derivative, i.e. $\frac{d^\beta f}{dt^\beta} = \frac{df}{dt} = \dot{f}$, which is a Newtonian dashpot.

2.2. Constitutive relation for isotropic power-law hereditariness

The extension of the constitutive relation presented in sec.2.1 and tensorial strain/stress state is discussed in this section by means of effect superposition.

Let us consider a 2nd-order stress tensor $\boldsymbol{\sigma}$ with component σ_{ij} represented in fig.(4) with the symmetries $\sigma_{ij} = \sigma_{ji}$ for $i \neq j$.

In the following we introduce the Voigt representation of the state variables of the material in terms of vector representation of stress and strains tensors as:

$$\boldsymbol{\sigma}^T(t) = [\sigma_{11}(t)\,\sigma_{22}(t)\,\sigma_{33}(t)\,\sigma_{32}(t)\,\sigma_{31}(t)\,\sigma_{12}(t)] \tag{4}$$

$$\boldsymbol{\varepsilon}^T(t) = [\varepsilon_{11}(t)\,\varepsilon_{22}(t)\,\varepsilon_{33}(t)\,2\varepsilon_{32}(t)\,2\varepsilon_{31}(t)\,2\varepsilon_{12}(t)] \tag{5}$$

where t is the current time and the mixed index stress and strain components, namely $\sigma_{ij}(t)$ and $\varepsilon_{ij}(t)$ with $i \neq j$ denote shear stress and strain, respectively. Let us assume that $\sigma_{ij}(t) = \delta_{ij}$ and let us consider a single normal stress $\sigma_{ii} = 1$ for (i = 1,2,3) reported in fig.4 a,b,c):

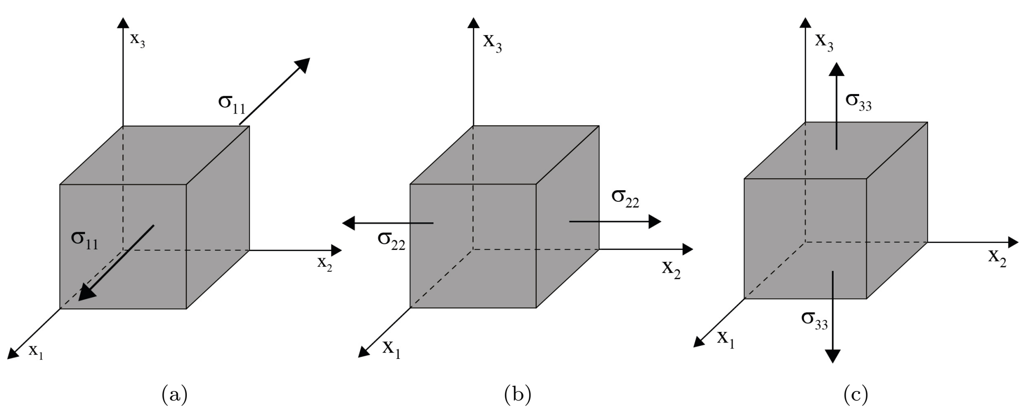


Figure 4: elementary representative cube: (a)only σ_{11}, (b)only σ_{22}, (c) only σ_{33}

In such a context the evolution of the strain $\varepsilon_{ii}(t)$ along the stress direction

$\sigma_{ii}\,(t)$ and in the orthogonal planes reads:

$$\varepsilon_{ii}\,(t) = J_L\,(t)\,\sigma_{ii} = J_L\,(t) \tag{6a}$$

$$\varepsilon_{kk}\,(t) = \varepsilon_{jj}\,(t) = -J_v\,(t)\,\sigma_{ii} \tag{6b}$$

with $i \neq j \neq k$ and $i,\, j,\, k = 1, 2, 3$.

In eqs.(6a-b) $J_L\,(t)$ and $J_v\,(t)$ are the axial and the transverse creep functions with respect to the stress direction, respectively. Under the assumption of smooth load process $\sigma_{ij}(t)$ the presence of contemporaneous stress $\sigma_{ij}(t) = \sigma_{ij}(t)\delta_{ij}$, with $i = 1, 2, 3$, may be accounted for by the integral

$$\varepsilon_{ii}\,(t) = \int_0^t J_L\,(t-\tau)\,\dot{\sigma}_{ii}\,(\tau) - J_v\,(t-\tau)\,[\dot{\sigma}_{jj}\,(\tau) + \dot{\sigma}_{kk}\,(\tau)]\,d\tau \tag{7}$$

with $i \neq j \neq k$ and i,j,k=1,2,3, respectively.

In the context of material isotropy shear strains $2\varepsilon_{ij}(t)$, $(i \neq j)$, are not involved by the axial stress $\sigma_{ii}\,(t)$, but only by the shear stress as $\sigma_{ij}(t)$ with $i \neq j$. The evolution of the shear strain $2\varepsilon_{ij}(t)$ due to a generic shear stress history $\sigma_{ij}(t)$ may be obtained by superposition integrals by means of the shear creep function $J_T(\cdot)$ as:

$$2\varepsilon_{ij}\,(t) = \int_0^t J_T\,(t-\tau)\dot{\sigma}_{ij}\,(\tau)\,d\tau \tag{8}$$

with $i \neq j$ and $i, j = 1, 2, 3$. The constitutive equations reported in eqs.(7),(8) may be reported in Voigt notation as:

$$\boldsymbol{\varepsilon}\,(t) = \int_0^t \mathbf{J}\,(t-\tau)\dot{\boldsymbol{\sigma}}\,(\tau)\,d\tau \tag{9}$$

where $\mathbf{J}(t)$ is the creep functions matrix that is described as:

$$\mathbf{J}\,(t) = \begin{bmatrix} \mathbf{J}^{(A)}\,(t) & \mathbf{0} \\ \mathbf{0} & \mathbf{J}^{(T)}\,(t) \end{bmatrix} \tag{10}$$

where the elements of the axial creep matrix $\mathbf{J}^{(A)}(t)$ are:

$$J_{ij}^{(A)}\,(t) = J_L\,(t)\,\delta_{ij} - (1-\delta_{ij})\,J_v\,(t) \tag{11}$$

with $i,j=1,2,3$. The shear creep matrix $\mathbf{J}^{(T)}(t)$ is a diagonal matrix gathering the shear creep functions $J_T\,(t)$ as:

$$J_{ij}^{(T)}\,(t)=J_T\,(t)\,\delta_{ij} \tag{12}$$

The three creep functions $J_L(t)$,$J_\upsilon(t)$ and $J_T(t)$ are related by a linear relation that reads:

$$J_T(t)=2J_L(t)-J_\upsilon(t) \tag{13}$$

that may be obtained, with straightforward manipulations, by introducing a shear stress state $\sigma_{ij}(t)$ that involves a shear strain state under isotropy assumption, namely $\gamma_{ij}=2\varepsilon_{ij}\,(t)$, and as evaluating the elongation and the stress along the principal axes at angles of $\pi/4$.

Under the assumption of linear elasticity, the creep functions coincide with the material compliance, that reads $J_T=1/G$, $J_L=1/E$ and $J_\upsilon=\upsilon/E$. The substitution in eq.(13), this yields:

$$\frac{1}{G}=2\left(\frac{1}{E}+\frac{\upsilon}{E}\right)=\frac{2\,(1+\upsilon)}{E} \tag{14}$$

that is the well-known relation among elasticity moduli.

Knowledge of the creep function matrix $\mathbf{J}(t)$ in eq.(10) allows for the definition of the relaxation matrix $\mathbf{G}(t)$ by means of the coniugation relation as:

$$\hat{\mathbf{G}}(s)\hat{\mathbf{J}}(s)=\frac{1}{s^2}\mathbf{I} \tag{15}$$

where $\mathbf{I}$ is the identity matrix and $\hat{\boldsymbol{G}}(s)$, $\hat{\boldsymbol{J}}(s)$ are the Laplace transforms of the relaxation $\mathbf{G}(t)$ and the creep functions $\mathbf{J}(t)$ matrices.

Straightforward manipulations of eq.(15) and inverse Laplace transform the relaxation matrix may be written as:

$$\mathbf{G}\,(t)=\begin{bmatrix}\mathbf{G}^{(A)}\,(t) & \mathbf{0}\\ \mathbf{0} & \mathbf{G}^{(T)}\,(t)\end{bmatrix} \tag{16}$$

where:

$$G_{ij}^{(A)}(t) = \mathcal{L}^{-1}\left[\frac{1}{s^2\left(\hat{J}_L+\hat{J}_v\right)\left(\hat{J}_L-2\hat{J}_v\right)}\right]\left[\left(\hat{J}_L-\hat{J}_v\right)\delta_{ij}+\left(1-\delta_{ij}\right)\hat{J}_v\right] \tag{17a}$$

$$G_{ij}^{(T)}(t) = \mathcal{L}^{-1}\left[\frac{1}{s^2\left(\hat{J}_L+\hat{J}_v\right)}\right]\delta_{ij} \tag{17b}$$

Eqs.(17a),(17b) show that in the presence of material fading memory, the relaxation matrix $\mathbf{G}(t)$ is obtained as a combination of creep functions relative to uniaxial creep tests. Similar considerations may be also withdrawn from the observation that in uniaxial relaxation tests, the relaxation function $G_L(t)$ is obtained in lateral free conditions, that is the strain state involves $\varepsilon_{11} \neq \varepsilon_{22} \neq 0$ and $\varepsilon_{33} = 1$ and measuring only $\sigma_{33}(t) = G_L(t)$ relaxation with $\sigma_{11} = \sigma_{22} = 0$.

Knowledge of the relaxation matrix of the material $\mathbf{G(t)}$ allows to evaluate the stress vector as:

$$\sigma(t) = \int_0^t \mathbf{G}(t-\tau)\dot{\varepsilon}(\tau)\,d\tau \tag{18}$$

The longitudinal shear and transverse relaxation functions $G_T(t)$,$G_L(t)$ and $G_v(t)$ are linearly related by an equation that is similar to the one involving creep functions in eq.(13), reading:

$$G_T(t) = \frac{1}{2}\left(G_L(t) - G_v(t)\right) \tag{19}$$

the latter allows for the evaluation of the transverse relaxation $G_v(t)$, as:

$$G_v(t) = 2G_T(t) - G_v(t) \tag{20}$$

In the following section, we derive the thermodynamic restrictions among the material parameters used in power-law representation of isotropic material hereditariness.

2.3. Power-law isotropic hereditariness: Thermodynamic restrictions

Let us assume that relaxation functions in laterally restrained axial and torsion shear tests may be captured, respectively, by power-laws with different order ($\alpha \neq \beta$) as:

$$G_L(t) = G_L^{(\alpha)} t^{-\alpha} + \bar{G}_L; \quad G_T(t) = G_T^{(\beta)} t^{-\beta} + \bar{G}_T \tag{21a}$$

$$G_v(t) = 2\left(G_T{}^{(\beta)} t^{-\beta} + \bar{G}_T\right) - \left(G_L{}^{(\alpha)} t^{-\alpha} + \bar{G}_L\right) \tag{21b}$$

with eq.(21b) obtained from the application of eq.(16). Physical dimensions of the coefficients are $[C_L] = [C_T] = F/L^2$, $\left[C_L^{(\alpha)}\right] = \dfrac{F}{L^2 T^{-\alpha}}$, $\left[C_L^{(\alpha)}\right] = \dfrac{F}{L^2 T^{-\beta}}$.

The expressions of the relaxation functions in eqs.(21a),(21b) yield the relaxation matrix of the material in eq.(16), with elements in the block matrices $\mathbf{G}^{(\mathbf{A})}(\mathbf{t})$ and $\mathbf{G}^{(\mathbf{T})}(\mathbf{t})$ reading:

$$G_{ij}^{(A)} = G_L(t)\,\delta_{ij} + (1 - \delta_{ij})\,G_v(t) \tag{22a}$$

$$G_{ij}^{(T)}(t) = G_T(t)\,\delta_{ij} \tag{22b}$$

We see that the relaxation matrix involves elements decaying with different power-laws of order β and α ($\alpha, \beta \in [0, 1]$). Start here the functional classes in eqs. (21a), (21b) are replaced in eqs.(22a), (22b).

The Coefficients and parameters involved in the power-law descriptions of the material relaxation, namely, $G_L(t)$, $G_v(t)$ and $G_T(t)$ are related by thermodynamical restrictions to ensure the requirement of positive entropy rate increment [31]. Indeed, a dissipative simple solid is defined only if the restrictions:

$$\mathbf{G}(0) \geq \mathbf{G}(\infty) \geq 0 \tag{23}$$

$$\dot{\mathbf{G}}(0) \geq 0 \tag{24}$$

are fulfilled by the relaxation matrix of the material as reported in basic references on material hereditariness [32, 33, 34, 35].

Eqs.(23,24) are always satisfied by assuming positive values of the coefficients $\bar{G}_L$, $\bar{G}_T$ and $G_L^{(\alpha)}$ and $G_T^{(\beta)}$, whereas eq.(25) alone is satisfied as the eigenvalues of the first derivative of the matrix, namely, $\dot{\mathbf{G}}\,(0)$ are all negative. This requirement may be verified by introducing a one-parameter family of relaxation matrices defined on a real parameter δ as $\mathbf{G}_\delta(t) = \mathbf{G}(t+\delta)$, and by studying the behavior of $\dot{\mathbf{G}}_\delta(t)$ for the limiting case $\delta \to 0$.

The parameter-dependent family of matrices $\dot{\mathbf{G}}_\delta(t)$ is defined as:

$$\dot{\mathbf{G}}_\delta\,(t) = \begin{bmatrix} \dot{\mathbf{G}}_\delta^{(A)}\,(t+\delta) & \mathbf{0} \\ \mathbf{0} & \dot{\mathbf{G}}_\delta^{(T)}\,(t+\delta) \end{bmatrix} \tag{25}$$

where the elements read:

$$\dot{G}_\delta^{(A)}\,(t+\delta) = -G_L^{(\alpha)}\alpha(t+\delta)^{-\alpha-1} \tag{26a}$$

$$\dot{G}_\delta^{(T)}\,(t+\delta) = -G_T^{(\beta)}\beta(t+\delta)^{-\beta-1} \tag{26b}$$

$$\dot{G}_\delta^{(\upsilon)}\,(t+\delta) = -2G_T^{(\beta)}\beta(t+\delta)^{-\beta-1} + G_L^{(\alpha)}\alpha(t+\delta)^{-(\alpha+1)} \tag{26c}$$

Observe that the one-parameter family $\dot{\mathbf{G}}_\delta(t)$ tends to the limit:

$$\lim_{\delta\to 0} \dot{\mathbf{G}}_\delta\,(t) = \dot{\mathbf{G}}\,(t) \tag{27}$$

We can infer the behavior of $\dot{\mathbf{G}}(t)$ from that of $\mathbf{G}_\delta(t)$, and by letting $\delta \to 0$. In this regard, the requirement in eq.(24) may be recast as:

$$-\dot{G}\,(0) = -\lim_{\delta\to 0} \dot{G}_\delta\,(t) \geq 0 \tag{28}$$

that is we evaluate the eigenvalues $\lambda_i(\delta)$ $(i = 1, 2, ...6)$ of the matrix $\dot{\mathbf{G}}_\delta(0)$ and with the additional constraints $-\lambda_i(\delta) \geq 0$ $(i = 1, 2, ...6)$ as $\delta \to 0$.

The evaluation of the eigenvalues $\lambda_i(\delta)$ gives:

$$-\lambda_1\,(\delta) = -\lambda_2\,(\delta) = -2\left(\dot{G}_L\,(\delta) - \dot{G}_T\,(\delta)\right) \geq 0 \tag{29a}$$

$$-\lambda_3\,(\delta) = -\lambda_4\,(\delta) = -\lambda_5\,(\delta) = -\dot{G}_T\,(\delta) \geq 0 \tag{29b}$$

$$-\lambda_6\,(\delta) = -4\dot{G}_T\,(\delta) + \dot{G}_L\,(\delta) \geq 0 \tag{29c}$$

Substitution of eq.(26a),(26b) into eq.(29b) shows that the inequality is fulfilled for $C_T^{(\beta)} \geq 0$ and $0 \leq \beta \leq 1$. The inequalities (29a),(29c) read, after substitution:

$$\alpha G_\alpha \delta^{-(\alpha+1)} - \beta G_\beta \delta^{-(\beta+1)} \geq 0 \tag{30a}$$

$$4\beta G_\beta \delta^{-(\beta+1)} - \alpha G_\alpha \delta^{-(\alpha+1)} \geq 0 \tag{30b}$$

that, after some straightforward manipulation, may be cast in a more suitable form, taking natural logarithms as:

$$\ln\left(A_{\alpha\beta}\right) \geq (\alpha - \beta)\ln\delta \tag{31a}$$

$$\ln\left[\frac{(A_{\alpha\beta})}{4}\right] \leq (\alpha - \beta)\ln\delta \tag{31b}$$

where $A_{\alpha\beta} = \alpha G_L^{(\alpha)} / \left(\beta G_T^{(\beta)}\right)$. Inequalities in eqs.(31a)(31b) must be fulfilled for any value of the parameter δ yielding that $\alpha = \beta$. Moreover, in this latter case the additional thermodynamical restriction holds true.

$$G_T^{(\beta)} \leq C_L^{(\beta)} \leq 3C_T^{(\beta)} \tag{32}$$

In passing, we observe that the condition $\alpha = \beta$ holds true only for the two terms (or one term) description of the relaxation function in eq.(22a). Indeed, as we assume that the relaxation functions $G_L(t)$ and $G_T(t)$ involve linear combinations of power-laws as:

$$G_L\left(t\right) = \sum_{j=1}^{n} G_L^{(\alpha_j)} t^{-\alpha_j}; G_T\left(t\right) = \sum_{i=1}^{m} G_T^{(\beta_i)} t^{-\beta_i} \tag{33}$$

with n and m the number of power-laws involved. Under such circumstances, the thermodynamical arguments proposed in this study yield the same conditions among the order of the power-laws as:

$$\max_{j=1,N}\left(\alpha_j\right) = \max_{i=1,M}\left(\beta_j\right) \tag{34a}$$

$$\min_{j=1,N}\left(\alpha_j\right) = \min_{i=1,M}\left(\beta_j\right) \tag{34b}$$

Substitution of eq.(22a),(22b) into the constitutive equations for the three-axial hereditariness yields a relation among the stress vector and the history of the strain vector $\boldsymbol{\varepsilon}(t)$ as:

$$\boldsymbol{\sigma}(t) = \mathbf{G}_\beta \int_0^t (t-\tau)^{-\beta} \dot{\boldsymbol{\varepsilon}}(\tau)\, d\tau + \bar{\mathbf{G}} = \mathbf{G}_\beta \left(D_{0^+}^{\beta} \boldsymbol{\varepsilon} \right)(t) + \bar{\mathbf{G}} \tag{35}$$

where we have embraced the Voigt representation of the relaxation tensor $\mathbf{G}(\mathbf{t})$ in matrix form and we have used the notation:

$$\mathbf{G}(t) = \mathbf{G}_\beta \frac{t^{-\beta}}{\Gamma(1-\beta)} + \bar{\mathbf{G}} \tag{36}$$

with the matrices:

$$\mathbf{G}_\beta(t) = \begin{bmatrix} G_\beta^{(L)} & G_\beta^{(v)} & G_\beta^{(v)} & 0 & 0 & 0 \\ G_\beta^{(v)} & G_\beta^{(L)} & G_\beta^{(v)} & 0 & 0 & 0 \\ G_\beta^{(v)} & G_\beta^{(v)} & G_\beta^{(L)} & 0 & 0 & 0 \\ 0 & 0 & 0 & G_\beta^{(T)} & 0 & 0 \\ 0 & 0 & 0 & 0 & G_\beta^{(T)} & 0 \\ 0 & 0 & 0 & 0 & 0 & G_\beta^{(T)} \end{bmatrix} \tag{37a}$$

$$\overline{\mathbf{G}} = \begin{bmatrix} \bar{G}_L & \bar{G}_v & \bar{G}_v & 0 & 0 & 0 \\ \bar{G}_v & \bar{G}_L & \bar{G}_v & 0 & 0 & 0 \\ \bar{G}_v & \bar{G}_v & \bar{G}_L & 0 & 0 & 0 \\ 0 & 0 & 0 & \bar{G}_T & 0 & 0 \\ 0 & 0 & 0 & 0 & \bar{G}_T & 0 \\ 0 & 0 & 0 & 0 & 0 & \bar{G}_T \end{bmatrix} \tag{37b}$$

The stress vector obtained as a functional of the strain vector $\boldsymbol{\varepsilon}(t)$ in eq.(35) is the generalization of the constitutive equation reported in eq.(3a) under the assumption of material isotropy.

In the next section the multiaxial fractional-order hereditariness will be further discussed by introducing a mechanical hierarchy that yields the constitutive model reported in eq.(35)

3. Exact mechanical description of fractional-order isotropic hereditariness

The stress/strain tensor outlined in sec.(2) requires a multiaxial constitutive relation, as in eq.(35), that under the assumption of $\bar{\mathbf{G}} = 0$ generalizes eq.(3a).

The rheological element, namely the springpot, corresponding to eq.(3a) can not, however, be defined also for the isotropic description in sec.(2), namely for the presence of shear stress/strain. A mechanical model that may be involved in the presence of normal and shear stress to be used in experimental test is represented in fig.5

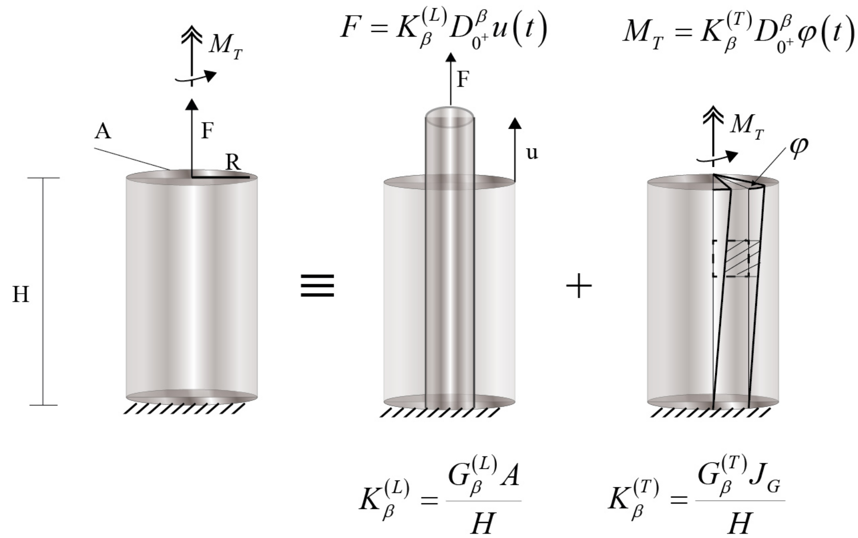


Figure 5: Rheologic elements

Under such conditions, the circular column of height H, cross section A and radius R under axial stress and shear stress related to the measured relative displacements $u(t)$ and twist angle $\varphi(t)$ provides these equilibrium equations:

$$\begin{aligned} F &= K_\beta^{(L)} \left(D_{0+}^\beta u\right)(t) \\ M_T &= K_\beta^{(T)} \left(D_{0+}^\beta \varphi\right)(t) \end{aligned} \tag{38}$$

where $A = \pi R^2$ and $J_G = \pi R^4/4$ are the cross section and the polar moment of inertia of the circular cross-section represented in fig.5. The constitutive equations in eq.(38) involve for limiting cases: i) a linear elastic spring ($\beta = 0$); and ii) a linear viscous element ($\beta = 1$), respectively.

In the following, we introduce a hierarchic mechanical model to capture the axial and shear hereditariness assuming power-law description of the creep and relaxation functions for axial and shear stress/strain, respectively [36, 20, 37, 19]. The obtained mechanical hierarchy corresponds exactly to an axial and shear springpots with the same order of time evolution/decay.

To this aim let us introduce an elastic column of unbounded length with circular cross section of radius R. The elastic features of the column are non-costant along the column axis and vary with the coordinate as:

$$E(z) = \frac{E_\alpha}{\Gamma(1-\alpha)} z^{-\alpha}; \quad G(z) = \frac{G_\alpha}{\Gamma(1-\alpha)} z^{-\alpha} \quad -1 \leq \alpha \leq 1 \tag{39}$$

The column is externally restrained by a set of torsional and axial viscous dash-pots fig.(5) with non-homogeneous viscosity $\eta(z)$ as:

$$\eta(z) = \frac{\eta_\alpha}{\Gamma(1+\alpha)} z^{-\alpha} \quad -1 \leq \alpha \leq 1 \tag{40}$$

Axial and torsional equilibrium along the column axis reads:

$$\frac{\eta_\alpha}{\Gamma(1+\alpha)} z^{-\alpha} 2\pi R \Delta z \dot{u}(z,t) = \frac{E_\alpha \pi R^2 s (z+\Delta z)^{-\alpha}}{\Gamma(1-\alpha)} \left[u(z+\Delta z,t) - u(z,t)\right] + \\ + \frac{E_\alpha \pi R^2 s z^{-\alpha}}{\Gamma(1-\alpha)} \left[u(z,t) - u(z-\Delta z,t)\right] \tag{41}$$

$$\frac{\eta_\alpha}{\Gamma(1+\alpha)} z^{-\alpha} 2\pi R^2 \Delta z \dot{\varphi}(z,t) = G_\alpha \pi R^4 (z+\Delta z)^{-\alpha} \left[\varphi(z+\Delta z,t) - \varphi(z,t)\right] + \\ + G_\alpha \pi R^4 (z+\Delta z)^{-\alpha} \left[\varphi(z,t) - \varphi(z-\Delta z,t)\right] \tag{42}$$

that, can be rewritten in differential form, by letting $\Delta z \to 0$ as:

$$\frac{\eta_\alpha z^{-\alpha}}{\Gamma(1+\alpha)} \frac{\partial u(z,t)}{\partial t} = \frac{E_\alpha R s}{\Gamma(1-\alpha)} \frac{\partial}{\partial z} \left(z^{-\alpha} \frac{\partial u(z,t)}{\partial z} \right) \tag{43a}$$

$$\frac{\eta_\alpha z^{-\alpha}}{\Gamma(1+\alpha)} \frac{\partial \varphi(z,t)}{\partial t} = \frac{G_\alpha R}{\Gamma(1-\alpha)} \frac{\partial}{\partial z} \left(z^{-\alpha} \frac{\partial \varphi(z,t)}{\partial z} \right) \tag{43b}$$

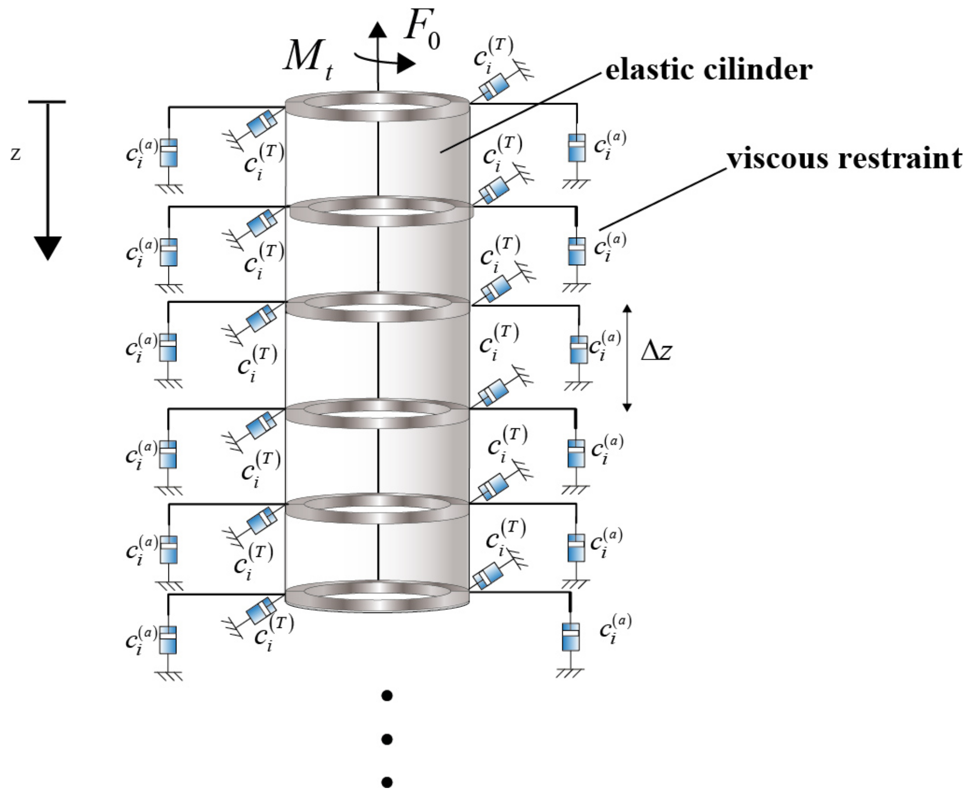


Figure 6: column with non-homogeneous viscosity

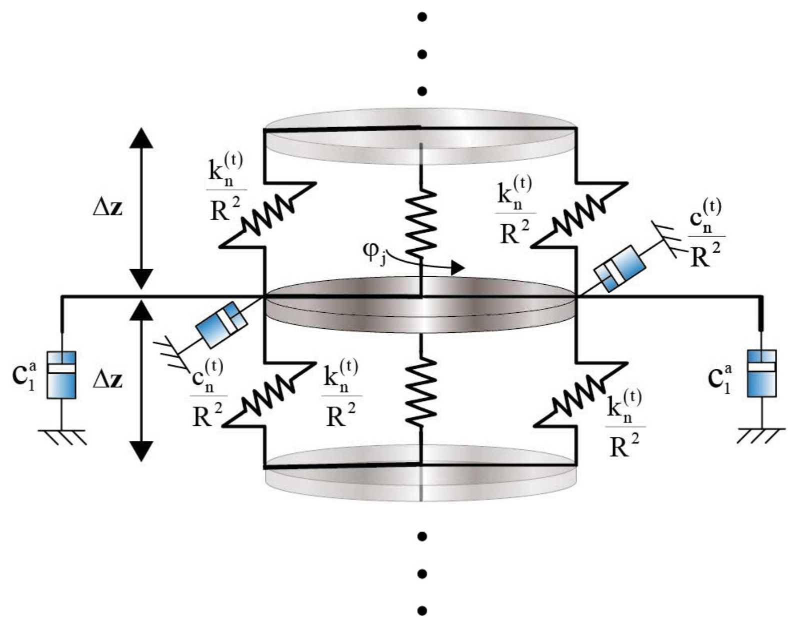


Figure 7: elements of the column with non-homogeneous viscosity

Boundary conditions involving the differential fields $u(z,t)$ and $\varphi(z,t)$ in eqs.(43a),(43b) read, respectively.

$$\lim_{z\to\infty} u\left(z,t\right)=0 \tag{44a}$$

$$\lim_{z\to 0}\frac{E_\alpha}{\Gamma\left(1-\alpha\right)}z^{-\alpha}\frac{\partial u}{\partial z}=F_0 \tag{44b}$$

$$\lim_{z\to\infty} \varphi\left(z,t\right)=0 \tag{45a}$$

$$\lim_{z\to 0}\frac{G_\alpha}{\Gamma\left(1-\alpha\right)}z^{-\alpha}\frac{\partial \varphi}{\partial z}=M_0 \tag{45b}$$

Mathematical operators and boundary conditions in eqs.(46a,b) are completely equivalent to those of a previous differential problem that has been solved by resorting to a non- linear mapping followed by Laplace transform [20, 38]. Such a procedure yields a Bessel differential equation of second kind in terms of the anomalous Laplace parameters. Position of the boundary conditions and inverse Laplace transform provides solution in the form:

$$u_0(t) = u_0(z,t) = \lim_{z\to\infty} u(z,t) = \frac{t^{-\beta}}{k_\beta^{(L)}} F_0 = J_L(t) F_0 \tag{46}$$

$$\varphi_0(t) = \varphi_0(z,t) = \lim_{z\to\infty} \varphi(z,t) = \frac{t^{-\beta}}{k_\beta^{(T)}} M_0 = J_T(t)_0 \tag{47}$$

with:

$$k_\beta^{(L)} = \frac{\Gamma(2\beta)\left(\tau_L^\beta\right)}{E_\alpha 2^{1-2\beta}\Gamma(\beta)\Gamma(1-\beta)} \tag{48}$$

$$k_\beta^{(T)} = \frac{\Gamma(2\beta)\left(\tau_L^\beta\right)}{G_\alpha 2^{1-2\beta}\Gamma(\beta)\Gamma(1-\beta)} \tag{49}$$

with $\beta = \frac{1+\alpha}{2}$ and the relaxation times:

$$\tau_L = \frac{\eta_\alpha}{E_\alpha}\frac{\Gamma(1-\alpha)}{\Gamma(1+\alpha)} \tag{50}$$

$$\tau_T = \frac{\eta_\alpha}{G_\alpha}\frac{\Gamma(1-\alpha)}{\Gamma(1+\alpha)} \tag{51}$$

Superpositions principle provides, by resorting to the fundamental equations of linear viscoelasticity, the constitutive equations of the macroscopic variables, as:

$$F_0(t) = k_\beta^{(L)}\left(D_{0^+}^\beta u_0\right)(t) \tag{52}$$

$$M_T(t) = k_\beta^{(T)}\left(D_{0^+}^\beta \varphi_0\right)(t) \tag{53}$$

Eqs.(52),(53) are the constitutive equation at the macro-scale and, by recalling that $F_0 = \sigma_{33}A$ and $|\tau| = \sqrt{|t_{31}|^2 + |t_{32}|^2} = \frac{M_0}{2As}$, the constitutive equations of the material read:

$$\sigma_{33} = G_\beta^{(L)}\left(D^\beta \varepsilon_{33}\right)(t) \tag{54}$$

$$|\tau| = G_\beta^{(T)}\left(D^\beta |\gamma|\right)(t) \tag{55}$$

where the coefficients $G_\beta^{(L)}$ and $G_\beta^{(T)}$ read:

$$G_\beta^{(L)} = \frac{\bar{k}_\beta^{(L)}\bar{l}}{A} \qquad G_\beta^{(T)} = \frac{\bar{k}_\beta^{(T)}}{2As}\frac{R}{\bar{l}} \tag{56}$$

and where$\bar{l}$ is an internal length of the material. Eqs.(54),(55) are the multiaxial constitutive relations of the isotropic material and, henceforth, correspond to the hierarchy introduced to the fractional-order isotropy.

4. Conclusions

The mathematical structure of the fractional-order isotropic hereditariness has been discussed in this paper. The study has been framed in the context of biomimetic ceramics used in cranioplasty neurosurgery (i.e. CustomBone® "prosthesis"). The creep and relaxation functions of isotropic linear hereditarinnes have been particularized for power-law decays, yielding a multi-axial constitutive model in terms of fractional-order operators. Additionally, a specific mechanical model has been introduced, that correspond to the fractional-order isotropic hereditariness. In future studies experimental campaigns involving creep and relaxation of biomimetic ceramics will be reported to assess the validity of material isotropy. Additionally, the proposed hierarchy will be further extended to deal with non-linear hereditariness as those observed in creep and relaxations of tendons and ligaments.

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