Evaluation of the temperature effect on the fractional linear viscoelastic model for an epoxy resin.

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Abstract. The paper deals with the evolution of the parameters of a fractional model for different values of temperature. An experimental campaign has been performed on epoxy resin at different levels of temperature. It is shown that epoxy resin is very sensitive to the temperature.

Keywords: fractional calculus, linear viscoelasticity, temperature, creep test, epoxy resin.
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INTRODUCTION

Linear viscoelasticity has traditionally been modeled by different proper arrangements of springs and dashpots. The most well-know mechanical models are Kelvin-Voigt and Maxwell. Kelvin-Voigt model consists of a spring in parallel with a dashpot and Maxwell model is a spring in series with a dashpot. Other classical models as standard linear solid, the Zener model, the Burger's, etc. are obtained by means of more complex and sophisticated arrangements of springs and dashpots. Classical viscoelastic models are governed by constitutive laws in which integer order derivatives and/or integrals appear. That implies that, for all classical models the strain history under a constant imposed stress history (the *creep function*) and also the response in terms of stress history under a constant imposed strain history (the *relaxation function*) are exponential laws.

The first who showed the experimental inconsistency of classical models to describe the viscoelasticity was Nutting. In fact, in his work [1], he observed that the stress relaxation of several materials have to be modeled as power-law time function with order $\beta : 0 \leqslant \beta \leqslant 1$, and this experimental evidence has been confirmed by other authors for different materials [3, 4]. In order to satisfy the Nutting's experience, Gemant in the 1936 [2] proposed the fractional-order derivative in the stress-strain relation to represent properly the stiffness and damping properties of viscoelastic media. These fractional operators are nothing else that the simple generalization of the classical ones for non-integer differential/integral order. Later, Scott-Blair & Caffyn [5] suggested the use of fractional calculus to provide a mathematical interpretation of the Nutting's experimental results.

Viscoelasticity is characterized by the time dependence. But it also has been shown, theoretically and experimentally, that the temperature effect change drastically the properties of viscoelastic materials. Temperature and time effect on viscoelasticity have been classically studied thought the *Time Temperature Superposition Principle* [6, 7].

In this paper, the effect of the temperature is studied through the coefficients that characterize the fractional viscoelastic model. On the basis of an experimental campaign for an epoxy resin, the variation of the relevant parameters of the proposed model is reported for different values of temperature.

FRACTIONAL LINEAR VISCOELASTIC MODEL

Linear viscoelasticity is based upon the knowledge of the creep and the relaxation function. The creep function, denoted by $J(t)$, represents the time response in terms of deformation due to a constant imposed stress-history $\sigma(t) = U(t)$, ($U(t) = 0 \; \forall \; t < 0$, $U(t) = 1$ elsewhere). The relaxation function, denoted by $G(t)$, is the time response in terms

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of stress due to a constant imposed strain-history $\varepsilon(t) = U(t)$. The Boltzmann superposition principle states that the stress history resulting from an arbitrary strain history $\varepsilon(t)$ is given as

$$\sigma(t) = \varepsilon_0 G(t) + \int_0^t G(t-\tau)\dot{\varepsilon}(\tau)d\tau, \tag{1}$$

where the kernel is the relaxation function $G(t)$ and ε_0 is $\varepsilon(t)_{t=0}$.

Viceverse, the strain history an assigned stress history $\sigma(t)$ is

$$\varepsilon(t) = \sigma_0 J(t) + \int_0^t J(t-\tau)\dot{\sigma}(\tau)d\tau, \tag{2}$$

where the kernel is the creep function $J(t)$ and σ_0 is $\sigma(t)_{t=0}$.

The fundamental relationship in Laplace domain of J(t) and G(t) may be easily derived from Eq.s (1) and (2) and results

$$\hat{G}(s)\hat{J}(s) = s^{-2}, \tag{3}$$

being $\hat{G}(s)$ and $\hat{J}(s)$ the Laplace transforms of $G(t)$ and $J(t)$, respectively.

The creep (or the relaxation) function may be determined through experimental data. On the bases of the experiments that Nutting [1] conducted on several materials, the creep (or the relaxation) function is well fitted by a power-law, that is

$$J(t) = \frac{t^\beta}{E_\beta\,\Gamma(1+\beta)}, \tag{4}$$

where E_β and $\beta \in \mathbb{R}^+ : 0 \leqslant \beta \leqslant 1$ are coefficients obtained from the best fitting of experimental data, and $\Gamma(\cdot)$ denotes the Euler gamma function. From the relationship in Eq. (3) and the creep function defined in Eq. (4), the relaxation function may be immediately obtained, and is given as

$$G(t) = \frac{E_\beta\, t^{-\beta}}{\Gamma(1-\beta)}. \tag{5}$$

By using the creep and relaxation functions in Eq.s (4) and (5) as kernels of the two convolution integrals in Eq.s (1) and (2), for a quiescent system at $t = 0$ the stress history $\sigma(t)$ and the strain history are given as

$$\sigma(t) = \frac{E_\beta}{\Gamma(1-\beta)}\int_0^t (t-\tau)^{-\beta}\dot{\varepsilon}(\tau)d\tau \qquad \varepsilon(t) = \frac{1}{E_\beta\Gamma(1+\beta)}\int_0^t (t-\tau)^{\beta}\dot{\sigma}(\tau)d\tau. \tag{6}$$

By definition

$$\frac{1}{\Gamma(1-\beta)}\int_0^t (t-\tau)^{-\beta}\frac{d\varepsilon(\tau)}{d\tau}d\tau = \left({}^{\mathrm{C}}D_{0^+}^{\beta}\varepsilon\right)(t), \tag{7}$$

that is the so called *Caputo's fractional derivative* of order β of the strain history. An integration by parts of $\varepsilon(t)$ in Eq. (6) leads to

$$\varepsilon(t) = \frac{1}{E_\beta\Gamma(\beta)}\int_0^t (t-\tau)^{\beta-1}\sigma(\tau)d\tau, \tag{8}$$

where $E_\beta\varepsilon(t)$ represents the *Riemann-Liouville fractional integral* of the stress history, denoted by $\left(I_{0^+}^{\beta}\sigma\right)(t)$.

So the constitutive laws of linear fractional viscoelasticity are written as

$$\sigma(t) = E_\beta\left({}^{\mathrm{C}}D_{0^+}^{\beta}\varepsilon\right)(t) \qquad \varepsilon(t) = \frac{1}{E_\beta}\left(I_{0^+}^{\beta}\sigma\right)(t). \tag{9}$$

Observe that for the two limit values of β ($\beta = 0$ and $\beta = 1$), since $\left({}^{\mathrm{C}}D_{0^+}^{0}\varepsilon\right)(t) = \varepsilon(t)$ and $\left({}^{\mathrm{C}}D_{0^+}^{1}\varepsilon\right)(t) = \dot{\varepsilon}(t)$ the elastic and the viscous behaviors are recovered, respectively.

The problem arises in the evaluation of the coefficient E_β for different values of temperature. As in fact, E_β has anomalous dimensions $[ML^{-1}T^{\beta-2}]$. Then direct comparison for different values of temperature may not be done since E_β depends on β. In order to overcome this issue, the creep function $J(t)$ may be rewritten in the form

$$J(t) = \frac{1}{E_\beta^*}\left(\frac{t}{\tau_\varepsilon}\right)^\beta, \tag{10}$$

where E_β^* is a coefficient with Young modulus dimensions $[ML^{-1}T^{-2}]$ and τ_ε is the fractional creep characteristic time. The fractional creep characteristic time, τ_ε, has been calculated through the proposed methodology in [8]. For a periodic stress input, τ_ε results

$$\tau_\varepsilon = q(\beta)\cot\left(\frac{\pi\beta}{2}\right)\sqrt[\beta]{\Gamma(1+\beta)}, \tag{11}$$

where $q(\beta) = \lambda_\varepsilon(0)\,\lambda_\varepsilon(\beta+1)/\lambda_\varepsilon(2)\,\lambda_\varepsilon(\beta)$ accounts for the distribution of the power due to each harmonic component present on the periodic function and its dimension is time [T]. $\lambda_\varepsilon(\rho)$ are the spectral moments of order ρ of the power associated to each harmonic component that is,

$$\lambda_\varepsilon(\rho) = \sum_{j=1}^{n} \varepsilon_j^2 (j\omega)^\rho, \quad \rho = 0, 2, \beta, \beta+1. \tag{12}$$

The constant imposed stress-history ($\sigma(t) = U(t)$) input of which the creep function is response, may be sought as periodic function (as a sum of harmonic functions), see Figure 1. For the case under study it is assumed a periodic stress function, and for this reason the characteristic creep time may be calculated as Eq (11).

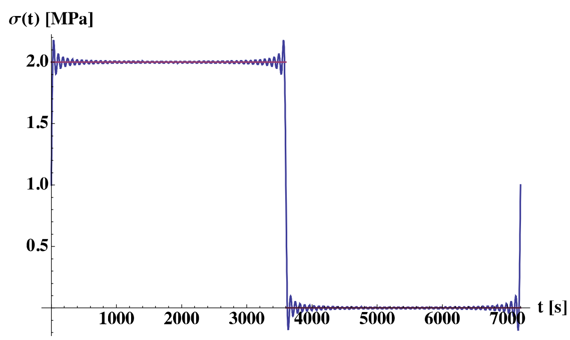

Figure 1. Stress function input in periodic form.

In the next section, the proposed model is used to evaluate the effect of the temperature in the creep function.

MODELLING THE EFFECT OF THE TEMPERATURE IN THE CREEP TEST

The fractional linear viscoelastic model may be used for every viscoelastic material. In this paper, the creep behavior of an epoxy resin is studied at different temperatures. The correspondent mechanical model for the material under study is the *fractional Maxwell model*, see Figure 2, that consists on a spring in series with a *springpot*. The springpot represents the fractional linear viscoelastic model described in the previous section.

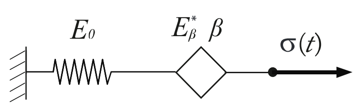

Figure 2. Fractional Maxwell model for SX8.

The constitutive law that characterizes the behavior of the fractional Maxwell model is

$$\varepsilon(t) = \frac{1}{E_0}\sigma(t) + \frac{1}{E_\beta^*}\left(I_{0^+}^{\beta}\sigma\right)(t), \tag{13}$$

where E_0 is the stiffness at $t = 0$. So, the creep function for the fractional Maxwell model is defined as

$$J(t) = \frac{1}{E_0} + \frac{1}{E_\beta^*}\left(\frac{t}{\tau_\varepsilon}\right)^\beta. \tag{14}$$

An epoxy resin (SX8 EVO) is under study, this thermosetting polymer has been cured for 24h at ambient temperature under atmospheric pressure. The creep tests have been performed in a DMA (Dynamic Mechanical Analyzer) machine, equipped with a load cell of 150N and capable of measuring displacement up to 6000 μm. The imposed stress history was $\sigma(t) = \sigma_0 U(t)$, where $\sigma_0 = 2$ MPa. The nominal dimensions of the specimens were 50mm x 3.5mm x 2.7mm.

The coefficients of the creep function have been obtained by a best fitting procedure from the experimental results of the creep test. The best fitting has been done with the program Mathematica 9. The results of the best fitting are depicted in the Figure 3, where in blue is plotted the experimental data and in red the best-fitting.

In the table , the values are obtained for the coefficients through the best fitting of the experimental results.

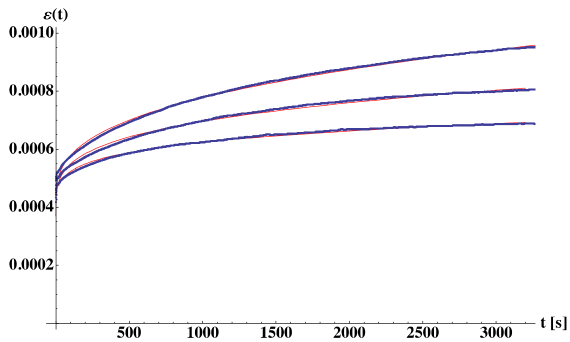

Figure 3. Best fitting results.

T [°C]	E_0 [MPa]	E^*_β [MPa]	β	τ_ε [s]
25	3009	4445	0.182	243.6
27	2649	4698	0.255	177.2
32.5	2434	5180	0.339	151.3

Table 1. Best fitting results at different temperatures.

CONCLUSIONS

In this paper, fractional linear viscoelastic model has been used to model the creep behavior of a polymer at different temperatures, concretely an epoxy resin. The present model and methodology may be used for modeling all viscoelastic materials. It has to be noticed that, from the obtained coefficients at the different temperatures it is possible to characterize the dynamic behavior of the material.

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References

[1] Nutting, P. G. (1921) *A new general law of deformation*, J. of The Franklin Institute, 191, pp. 679-685.
[2] Gemant, A. (1936) *A method of analyzing experimental results obtained by elasto-viscous bodies*, J. of Physics, 7, pp. 311-317.
[3] Di Paola, M., Pirrotta, A. & Valenza, A. (2011) *Visco-elastic behavior through fractional calculus: an easier method for best fitting experimental results*, Mechanics of materials, 43, pp. 799-806.
[4] Di Paola, M., Fiore, V., Pinnola, F. P. & Valenza, A. (2014) *On the influence of the initial ramp for a correct definition of the parameters of fractional viscoelastic materials*, Mechanics of Materials, 69, pp. 63-70.
[5] Scott Blair, G. W. & Caffyn, J. E. (1949) *An application of the theory of quasi-properties to the treatment of anomalous strain-stress relations*, The Philosophical Magazine 40 (300), pp. 80-94.
[6] Williams, M.L., Landel, R.F. & Ferry, J.D. (1955) *The temperature dependence of relaxation mechanisms in amorphous polymers and other glass-forming liquids*, J. of American Chemical Society, 77, pp. 3701-3707.
[7] Gross, B., (1968) *Mathematical Structure of the Theories of Viscoelasticity*, Hermann et Cie., Paris.
[8] Colinas-Armijo, N., Di Paola, M., & Pinnola, F. P. (2016) *Fractional characteristic times and dissipated energy in fractional linear viscoelasticity*, Communications in Nonlinear Science and Numerical Simulation, doi:10.1016/j.cnsns.2016.01.003.