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FRACTIONAL THERMOVISCOELASTIC MATERIALS

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Abstract. *This paper aims to investigate the effects of the change of temperature in the fractional constitutive law of viscoelastic material. It is shown that the well known William Landell and Ferry (WLF) (or the Harrenius) principle is not valid for real fractional viscoelastic materials unless the order of fractional operators is indipendent on the temperature. With the aid of some experimental date obtained by performing Creep test at different temperature the order of the fractional operators is not constant and then they may be considered thermoreologically simple. This modification of the WLF is here proposed in order to fully investigate the effects of the temperature in the fractional constitutive law.*

1 INTRODUCTION

Creep and Relaxation function of real materials are well fitted by power law [1]. As a consequence , in the linear hypothesis, the Boltzmann superposition principle holds and the convolution integrals are the so called fractional operators [3,4,5]. Experimental evidence shows that the parameters are very sensitive to the change of temperature. As this effect will be taken into account, the various parameters will depend on the temperature and then implicitly on the time if the temperature is time dependent is it happens in many engineering applications. Many papers have been devoted to this subject [7,8,10,13,14]. Thermoviscoelastic theory has been also investigated in many papers Lord and Shulman Dhaliwal and Sherif [15,16].
How stated before the parameters strictly depend on the temperature and in literature many studies have been devoted to this subject. In particular the so-called time temperature superposition is used. Such a principle states that as the temperature increase (or decrease) as the temporal scale is properly modified the Creep function will remain the same at the various temperature. This principle strictly holds only for *thermoreologically simple* materials. This principle however does not holds true for real materials. This is due to the fact that Creep and/or relaxation function are Power-law [1] rather than exponential like in the classical models of viscoelasticity (Kelvin-Voigt, Maxwell, Zener, Burger, ...). This produce two effects: first of all the constitutive laws are ruled by fractional operators (Riemann-Liouville

and Caputo's fractional derivative) ruther than classical derivatives and integrals; the second effect is that the parameters of the fractional viscoelastic model are strongly influenced by the temperature. It follows that in the case in which the temperature depends on the time (day-night, summer-winter) also the parameters depend on the time and the Creep and/or relaxation function will depend not only on the stress (or the strain) but also on the temperature that in turn depend on the time. In this paper it is shown that the TTS for the fractional model is still applicable only if the order of fractional derivative (or integral) is indipendent on the temperature. That is the fractional model thermoreologically simple only if the order of the fractional operator does not depends on the temperature. Two real materials have been taken into account the Polypropylene and Ethylene Vinyl Acetate (EVA). Creep test are such a materials performed at different temperatures show that both are not thermoreologically simple because the order of fractional operators are not constants. In order to work with such a material a modified William Landell and Ferry law is proposed that is not only the temporal scale is modified but also the shift on the order of fractional operators is examined.

2 PRELIMINARIES IN FRACTIONAL VISCOELASTICITY

Nutting [1], on the basis of a wide experimental campaign observed that real materials like rubber, polymers steal ..., states that the for an assigned stress history $\sigma(t)=\sigma_0 U(t)$ ($U(t)$ being the unit step function) the corrisponding strain is given in the form:

$$\varepsilon(t)=at^{\alpha}\sigma_0^{\mu};(a,\mu\in \mathrm{R}^{+});0\leq\alpha\leq 1 \tag{1}$$

with a, α and μ characteristic parameters depending on the material at hands. In particular if $\sigma_0=1$ eq.(1) becomes the so called *Creep function* in the following denoted $\mathsf{C}(\mathrm{t})$. The conseguence of eq.(1) is that as we assume that σ_0 is small, then the creep function assumes the form:

$$\mathsf{C}(\mathrm{t})=\frac{1}{\mathrm{C}_{\alpha}\Gamma(1+\alpha)}\mathrm{t}^{\alpha};\ 0\leq\alpha\leq 1 \tag{2}$$

where $\Gamma(\cdot)$ is the Euler Gamma function and C_{α} is an anomalous coefficient (Pas^{α}). As soon as the Creep function is assumed in the form expressed in eq.(2) the Boltzmann superposition principle, for an assigned stress hystory $\sigma(t)$, is given as:

$$\varepsilon(t)=\frac{1}{C_{\alpha}\Gamma(1+\alpha)}\int_0^t\dot{\sigma}(\tau)(t-\tau)^{\alpha}d\tau \tag{3}$$

Such an equation is valid if the system is quiescent at $t=0$ ($\varepsilon(0)=0$) and $\sigma(0)=0$. If $\sigma(0)\neq 0$ we have to add at the r.h.s of eq.(3) the term $\mathsf{C}(\mathrm{t})\sigma(0)$. Integration by part eq.(3) leads to:

$$\varepsilon(t)=C_{\alpha}^{-1}(I_{0^+}^{\alpha}\sigma)(t) \tag{4}$$

where the symbol $(I^{\alpha}_{0^+}\sigma)(t)$ means *Riemann– Liouville fractional integral,* that is:

$$(I^{\alpha}_{0^+} f)(t) = \frac{1}{\Gamma(\alpha)}\int_0^t (\tau - t)^{\alpha-1} f(\tau)d\tau \tag{5}$$

Let us now denote as $\mathsf{R}(t)$ the *relaxation function* that is the stress hystory to an imposed strain history of the kind $\varepsilon(t) = U(t)$. Since the following general law of the viscoelasticity holds true in Laplace domain (Flugge)

$$\hat{\mathsf{C}}(\mathrm{s})\hat{\mathsf{R}}(\mathrm{s})=1/\mathrm{s}^2 \tag{6}$$

where $\hat{\mathsf{C}}(\mathrm{s})$ and $\hat{\mathsf{R}}(\mathrm{s})$ are the Laplace transform of $\mathsf{C}(\mathrm{t})$ and $\mathsf{R}(\mathrm{t})$, respectively and s is the (complex) parameter of the Laplace operator. From eq.(6), after some algebra, it is recognised that $\mathsf{R}(\mathrm{t})$, corresponding to the creep function in eq.(2) is given as:

$$\mathsf{R}(t) = \frac{C_\alpha}{\Gamma(1-\alpha)} t^{-\alpha} \tag{7}$$

It follows that the Boltzmann superposition principle gives the stress history corresponding to a general strain history $\varepsilon(t)$ as:

$$\sigma(t) = \frac{C_\alpha}{\Gamma(1-\alpha)}\int_0^t \dot{\varepsilon}(\tau)(t-\tau)^{-\alpha} d\tau \tag{8}$$

or in equivalent form:

$$\sigma(t) = C_\alpha ({}_C D^{\alpha}_{0^+}\varepsilon)(t) \tag{9}$$

where the symbol $({}_C D^{\alpha}_{0^+}\varepsilon)(t)$ is the so called *Caputo's fractional derivative* defined as:

$$({}_C D^{\alpha}_{0^+} f)(t) = \frac{1}{\Gamma(1-\alpha)}\int_0^t (\tau - t)^{-\alpha} \dot{f}(\tau)d\tau;\ 0 \le \alpha \le 1 \tag{10}$$

The constitutive law expressed in eq.(9) is valid provided $\varepsilon(0) = 0$. In this case the Caputo's fractional derivative coalesces with the Riemann– Liouville fractional derivative.

Eq.(4) and (9) represent the constitutive laws of fractional viscoelasticity. At this stage some remarks are necessary: i) As $\alpha = 0$ the elastic law is recovered and for $\alpha = 1$ the Newton-Petrof law is recovered, it follows that the intermediate value $0 < \alpha < 1$ gives an intermediate behaviour of the two extreme (idealized) cases; ii) for a quiescent system (at $t = 0$) the operators (5) and (10) are inverse each another; iii) operators (5) and (10) are linear ones and for such operators all the rules of classical derivative and integral hold true (linearity, integration by parts, Leibnitz rule, ...), moreover in Fourier and in Laplace domain they exactly behaves like for the classical derivative and integrals; iv) these operators are valid for $\alpha \in \mathrm{R}^+$ or even for $\alpha \in \mathrm{C}$ (with $Re(\alpha) > 0$).

In the next section the effects of the temperature on the fractional constitutive law is presented.

3 FRACTIONAL ORDER THERMOVISCOELASTICITY

In this section we assume that the body is hysotropic, and the component of stress σ and the corresponding strain are composed by only one component as it happens for the purely tangential stress or purely volumetric component. Such an example for the purely tangential stress τ eq.(4) is rewritten in the form $\gamma(t) = G_\alpha^{-1}(I_{0^+}^{\alpha}\tau)(t)$, γ being the tangential strain corresponding to the tangential stress τ; or for the volumetric stress σ_v it corresponds to $\varepsilon_v = 3k_\alpha^{-1}(I_{0^+}^{\alpha}\sigma_v)(t)$ and C_α has been assumed as G_α^{-1} and $3k_\alpha^{-1}$, because they are the shear and the Bulk's modulus (anomalous) for these two cases. In the following we shall assume the general law in eq.(4) and in eq.(9). Now it has been bserved that as the temperature varies, the parameters α and C_α varie that is

$$\alpha = \alpha(T) \ ; \ C_\alpha = C_\alpha(T) \tag{11}$$

The variation of these coefficients is general very large. Such an example for the water at $T \leq 0^o C$ the liquid phase disappear (glass) and $0;\ \alpha$, while at $T > 0^o C$ the solid phase disappears and $\alpha;\ 1$ (pure Newtonian fluid).

These variations may be used in order to reduce times of experimental results. Such an example for a resin at a temperature of $25^o C$ we need of 5 hours to determine the experimental Creep or Relaxation function, but if we performe the same experiment at $35^o C$ we may use a reduced time of order 20 minuts. Another reason to investigate in this field is that after many materials commonly used in structural elements like pultruded beams in structural engineering or bitumens in the railways engineering suffer of cyclic change of temperature (day-night and/or summer-winter) and in these circumstances the evaluation of the response to given loads in terms of deformation depend on these changes it follows that we have to take into account for these variation, in the equation of motion. Usually empirical models have been proposed in literature in order to take into account the effects of the change of temperature the so-called shift factor that is valid for the so-called *thermoreologically simple* materials is applied.This principle is formally proposed in the following form:

$$\mathsf{R}(t,T) = \mathsf{R}(t_0,T_0) \tag{12}$$

That reads: the relaxation function at a given time t at the temperature T equals to the relaxation function at time t_0 corresponding to a reference temperature T_0. The general form of t_0 is:

$$t_0 = \frac{t_0}{a_T} \tag{13}$$

where a_T is the so-called *shift factor*. Different expression of the shift factor have been proposed in literature. The most popular expression is the William-Landel and Ferry (WLF), that is:

$$\log a_T = -\frac{C_1^0(T-T_0)}{C_2^0+(T-T_0)} \tag{14}$$

where C_1^0 and C_2^0 are coefficients obtanied by best fitting on experimental data. The WLF expression is valid for $T > T_g$, being T_g is the glass transition temperature. For $T < T_g$ the other model is that proposed by Arrhenius, that is:

$$\log a_T = -\frac{\Delta H_a}{R}\left(\frac{1}{T}-\frac{1}{T_0}\right) \tag{15}$$

where R is the universal constant of gasses and ΔH_a is the activation energy.
For thermoreologically simple materials the Time Temperature Superposition (TTS) holds that basically consists in assuming that the time scale of observation at the given temperature, if properly changed, produce identical result of that obtained at a reference experiment by changing the temporal scale according to the eq.(13). Now the question is: what are the conditions for which a fractional viscoelastic material is thermoreologically simple?
In order to answer to this fundamental question let us assume that the material is thermorologically simple then, according to eq.(12) we may write:

$$\frac{C_\alpha(T)}{\Gamma(1-\alpha(T))}t^{-\alpha(T)} = \frac{C_\alpha(T_0)}{\Gamma(1-\alpha(T_0))}t^{-\alpha(T_0)} \tag{16}$$

From this equation it is evident that a material is not thermoreologically simple unless $\alpha(T)=\alpha(T_0)$. Unfortunately this is not the case for many real materials, such an example from the experimental data in the laboratory of Palermo [8] performed on the propilene for a given load ($55MPa$) and for different temperature ($10^o C, 23^o C, 37^o C, 51^o C$ and $65^o C$) the Creep functions of the material at different temperature are represented in fig.1

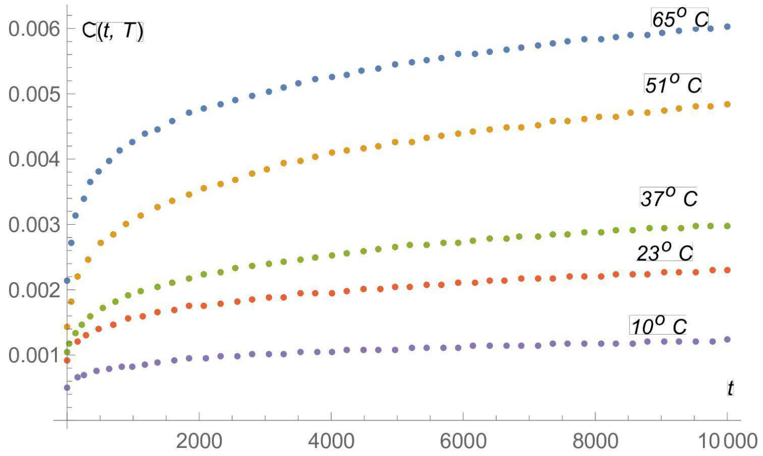

Fig.1 Creep function at different temperatures (experiments)

The best fitting determine the parameters C_α and α for the different temperatures and the result are depicted in fig(2) and (3), respectively

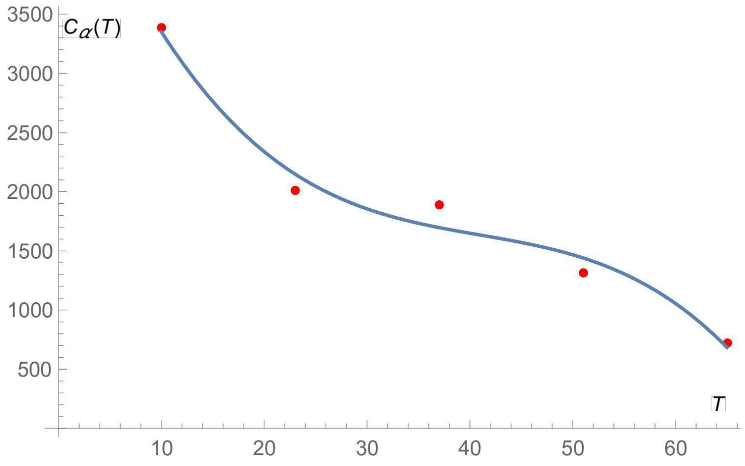

Fig.2 Best fitting of $C_\alpha(T)$

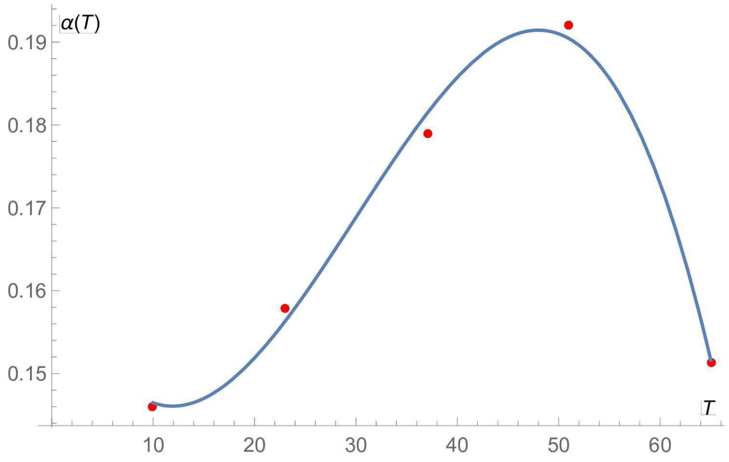

Fig.3 Best fitting of $\alpha(T)$

From figs.(2) and (3) we may states that $C_\alpha(T)$ is well suited by the law:

$$C_\alpha(T) = C_0 + C_1 T + C_2 T^2 + C_3 T^3 \tag{17}$$

where:

$$C_0 = 0.99344, C_1 = -0.0438913, C_2 = 0.00100136, C_3 = -8.15417 * 10^{-6}$$

$$\alpha(T) = A_0 + A_1 T + AT^2 + A_3 T^3 \;\; a > 0, b > 0 \tag{18}$$

where:

$$A_0 = 0.164219, A_1 = -0.00332012, A_2 = 0.000173832, A_3 = -1.93437 * 10^{-6}$$

and equation (18) reveals that since $\alpha(T)$ is not a constant the polyvinile is not thermoreologically simple.

Analogous result is found for the Ethylen Vinil acetate (EVA) commonly used for a solar cells, glass cover and so-on. A recent study [10] give the result of α and C_α based by the best fitting performed at different temperature.

These results are reported in Table 1

$T[^oC]$	α	$C_\alpha\,[Pa * s^\alpha]$
-35	0.22600	814.7
-28	0.16810	182.7
-18	0.10150	52.63
0	0.05566	23.55
20	0.04227	11.04
40	0.07417	4.668
49	0.08634	4.116
60	0.06542	1.544
80	0.05117	1.049
100	0.04179	0.9276
119	0.03610	0.7965
139	0.03311	0.8228

Table 1 result by best fitting based on experimental campaign at different temperatures (result reported in [10])

From this table is apparent that $\alpha(T)$ may not be considered constant with respect to the temperature, so also EVA may not be considered thermoreologically simple materials.

In the next section a Modified TTS (MTTS) is proposed to define the behavior of materials not thermoreologically simple.

4 MODIFIED TIME TEMPERATURE SUPERPOSITION PRINCIPLE

We introduce two shift factors one for the time a_T and the other one for the variation of $\alpha(T)$ that will labeled as b_T. Eq.(12) for a thermoreologically simple material read as:

$$\mathrm{R}(t,T,\alpha)=\mathrm{R}(a_T * t_0, T_0, \alpha) \tag{19}$$

for the materials for which α is not constant we propose:

$$\mathrm{R}(t,T,\alpha)=\mathrm{R}(a_T * t_0, T_0, b_T * \alpha_0) \tag{20}$$

It is obvious that this modification reproduce the TTS for the case in which α is indipendent on the temperature ($b_T = 1$).

Such an example for the EVA the coefficient b_T is plotted in fig.4. The best fitting is performed by assuming $T_0 = -18^o C$, $\alpha_0 = 0{,}10150$ and a third order polynomial of the form $b_T = b_0 + b_1 T + b_2 T^2 + b_3 T^3$ match quite well all the experimental data (dotted line) with the value $b_0 = 0.493646$, $b_1 = -0.0147932$, $b_2 = 0.000764126$, $b_3 = -6.37002 * 10^{-6}$.

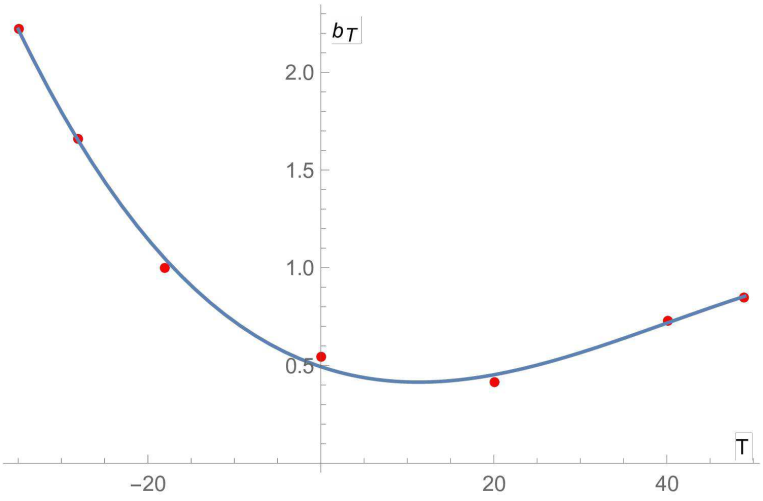

Fig.4 b_T for different value of the temperature; continuos line best fitting

Once b_T is obtained the value of the shift factor for the temperature may be easily found. In fig.(5) the $\log a_T$ is plotted versus T for the EVA. In dotted line are reported the $\log a_T$ and in solid line. The result of the best fitting with a polynomial of third order $\log a_T = a_0 + a_1 T + a_2 T^2 + a_3 T^3$. For the EVA the coefficient are $a_0 = 7.40345$, $a_1 = 0.435922$, $a_2 = -0.00150735$, $a_3 = -0.000109548$.

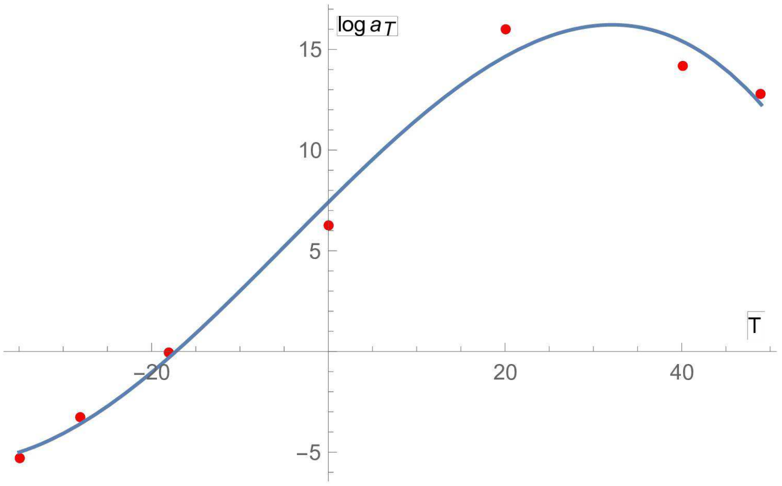

Fig.5 $\log a_T$ for different value of the temperature;

5 CONCLUSION

In this paper the effect of the temperature on the parameters of the fractional constitutive law (order $\alpha : 0 \le \alpha \le 1$) has been investigated. It has been shown that for the fractional model the time-temperature shift method may not be applied unless the exponent α of the power law in the Creep function is independent on the temperature. It is also shown that from the experimental data obtained for polypropylene and on the EVA ath different temperatures the order of derivative is not constant. Then a modification of the WLF is proposed by shifting both the time and the order α.

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