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A DISTRIBUTED ORDER VISCOELASTIC MODEL FOR SMALL DEFORMATIONS

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Abstract. *In this work we discuss the connection between classical, fractional and distributed order viscoelastic Maxwell models, presenting the basic theory supporting these constitutive equations, and establishing some background on the admissibility of the distributed order Maxwell model. We derive the storage and loss modulus functions for the distributed order viscoelastic model and perform a fitting to experimental data. The fitting results are compared with the Maxwell and Fractional Maxwell models.*

1 INTRODUCTION

Viscoelastic fluids are abundant in nature, and also play an important role in our daily lives. Examples include paints that present better or worse adherence to walls; food and plastic products; saliva and DNA, etc. These materials show a combination of a viscous and a elastic behaviour, thus often resulting in a counter-intuitive behaviour. It is therefore imperative to better understand, and model these materials, so that improvements in the processing of such materials can be achieved.

1.1 The Boltzmann Approach

In 1876 Boltzmann performed a reprinting of his 1874 paper *Zur Theorie der elastischen Nachwirkungen* [1, 2], where he suggested the following: consider the function $\gamma(t)$ (the strain or deformation), as a cause, that will promote some effect (the stress, $\sigma(t)$) on the viscoelastic material. With, $G(t)$ being a property of the material, relating the cause and effect.

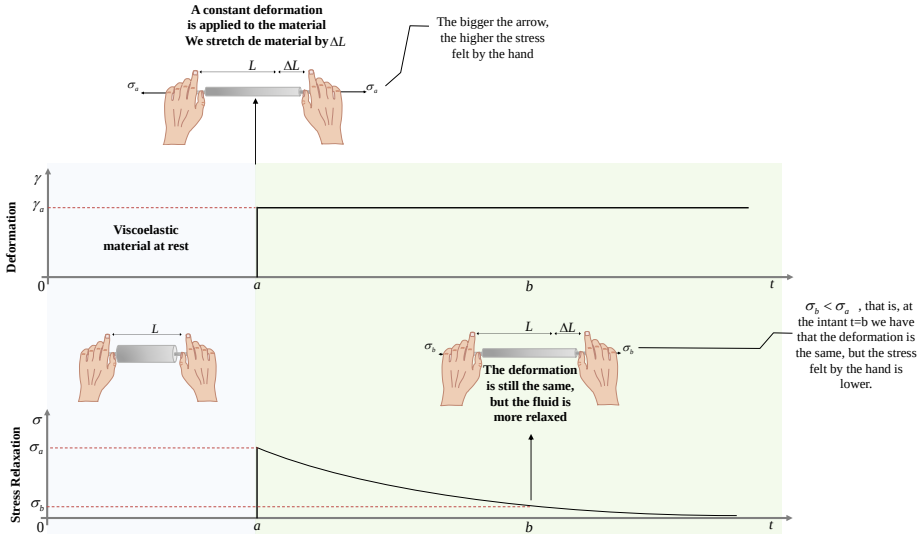

Figure 1: Two hands hold a viscoelastic material. At $t = a$ we stretch the material by ΔL (this deformation is kept constant along time). The viscous forces allow the material to adapt to these deformation, and the stress felt in hour hands starts to decrease, until its 0. Note that we reach a zero stress but the the deformation is still constant, that is, the material does not recover its initial configuration (this would only happen for a pure elastic material).

In order to understand his idea lets look at the relaxation experiment shown in figure (1). Two hands hold a viscoelastic material from $t = 0$ till $t = a$. The material is at rest, and therefore the hands do not *feel* any stress (we assume the weight of the material is negligible). At $t = a$ we stretch the material by ΔL (see the top graph for the strain $\gamma(t)$) and we *feel* a stress σ_a (this deformation is kept constant along time, with a value

of γ_a). The stress appears due to the elastic forces that want to recover the original position at rest. As times goes by, the viscous forces allow the material to adapt to these deformations, and the stress felt in hour hands starts to decrease. For example, at $t = b$ we have that $\sigma_b < \sigma_a$ (see in the bottom graph the decay of the stress along time).

Taking into account the fact that a variation in the deformation (cause) at time t' will produce a corresponding effect at some later time, t (part of the effect is almost instantaneous, but the influence of the cause propagates through time, thus influencing the future), it is plausible to assume $G(t)$ as a function of the time delay between cause and effect. Then, it can be said that

$$\sigma(t) = G(t - t')\,\gamma(t') \tag{1}$$

with t' a past time, and $G(t - t')$ representing a fading memory. G depends on the elapsed time $t - t'$ between the remembered past and the present. Note that the strain is a function of the past t'.

Now, assume that small variations of the cause (γ) are performed, leading to small variations of the effect (σ), and that this variations are so small that G does not depend on γ (this is called linear viscoelasticity). This can be easily understood with the experiment shown in figure (1), and further explored in figure (2), where two different constant strains are applied and the respective stresses are monitored at $t = t_1$ and $t = t_2$ (assume that $t' = 0$). Since the strain (γ) is constant we have that

$$G(t) = \frac{\sigma(t)}{\gamma}. \tag{2}$$

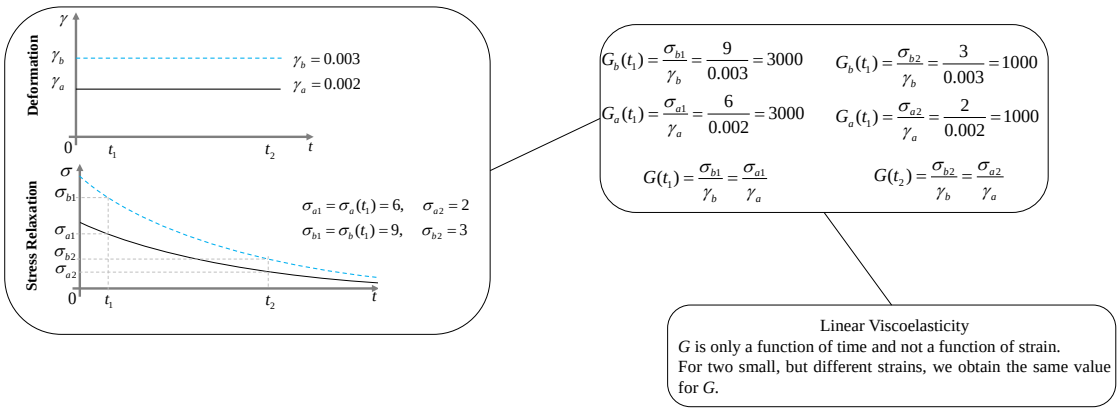

Figure 2: Two different constant deformations/strains are applied, γ_a and γ_b. The stress is measured at instant t_1 and t_2, for both the relaxations. The relaxation modulus is measured using equation (2). It is observed that the relaxation modulus only varies with time and not with γ (linear viscoelasticity).

Two different constant deformations/strains are applied, γ_a and γ_b. Depending on the deformation applied, we obtain a certain relaxation of the stress. The stress is measured

at instants t_1 and t_2, for both the relaxations. The relaxation modulus is measured using equation (2). It is observed that the relaxation modulus only varies with time and not with γ. This is called linear viscoelasticity, and can only be obtained for *small* deformations. In this work we will study a viscoelastic model that is only valid in this linear regime.

The Boltzmann principle states that the stress caused by a deformation, can be decomposed into independent contributions of very small deformations, and therefore, we can integrate all the stress history up to the moment of interest, t. Note that the sum of all this deformation is still assumed to be in the linear regime. This can be easily verified in the following figure (3). We have imposed a certain strain (input) or deformation (that is growing with time), and we want to know the stress obtained along time (output).

Firstly, assume that the variable input can be represented by a series of step inputs each of which begins at a different instant as shown below.

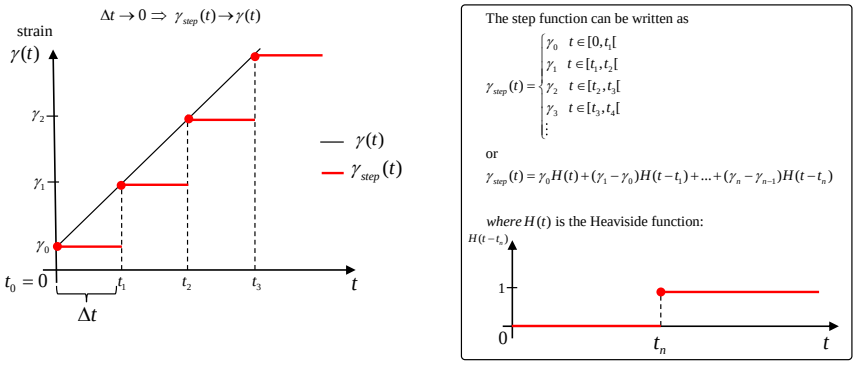

Figure 3: The strain approximated by a series of step inputs.

We see that as the time step becomes smaller the step function becomes a good approximation to the real strain or deformation. In reality we can approximate the strain by the step function to any degree of accuracy desired. What is important to note is that the strain can be written as (with $H(t)$ the Heaviside function defined in figure (3)):

$$\gamma(t) = \gamma_0 H(t-t_0) + (\gamma_1 - \gamma_0) H(t-t_1) + (\gamma_2 - \gamma_1) H(t-t_2) + \ldots + (\gamma_n - \gamma_{n-1}) H(t-t_n) \quad (3)$$

Secondly, consider the stress and the strain have the additive property (Boltzmann Principle - in the linear viscoelastic regime, the stress (strain) responses to successive strain (stress) stimuli are additive.). As shown in figure (4), the different strain inputs shown in figure (3) can be seen as independent from each other, and the overall strain is the sum of these independent strains ($\Delta\gamma_0$, $\Delta\gamma_1, \ldots$). Note that in the first step we have an input of $\Delta\gamma_0$ that continues along time, in the second and independent instant we have an input of $\Delta\gamma_1$ that also continues along time, and so on. When comparing figure (3)

and figure (4) we only obtain a strain input of γ_1 if we sum $\Delta\gamma_0$ and $\Delta\gamma_1$. This process continues as long as the deformation occurs (assuming that everything stays in the linear regime).

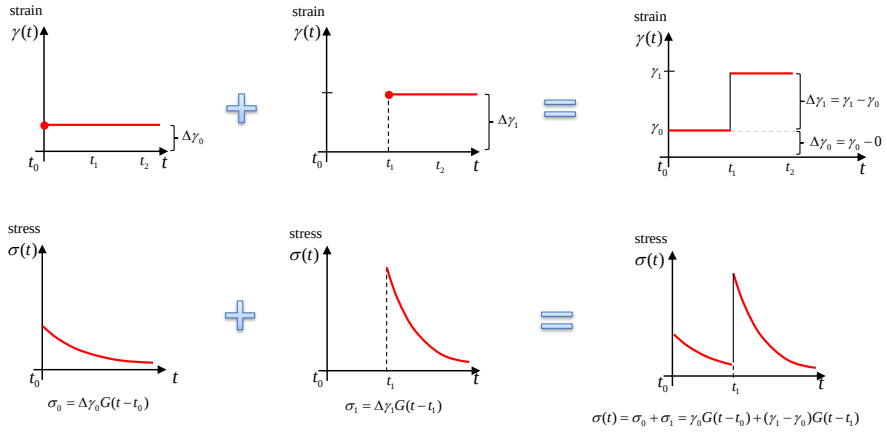

Figure 4: The Boltzmann principle illustrated.

The stress obtained is then the sum of the individual and independent stresses caused by the the strains $\Delta\gamma_0$ and $\Delta\gamma_1$. A more refined set of input strains would lead to a more continuous variation of the stress along time, and thus allowing one to obtain the stress as a continuous function of time.

To guarantee that the future stress outputs won't influence the past outputs, the Heaviside function will by multiplied by each stress output, resulting in the following formula:

$$\sigma(t) = \gamma_0 G(t-t_0)H(t-t_0) + (\gamma_1-\gamma_0)G(t-t_1)H(t-t_1) + \ldots + (\gamma_n-\gamma_{n-1})G(t-t_n)H(t-t_n) \tag{4}$$

The new $G(t-t_n)H(t-t_n)$ term with the Heaviside function, forces it to be 0 for $t < t_n$. Assuming $t_0 = 0$ we have $\sigma(t) = \gamma_0 G(t)H(t) + \sum_{i=1}^{n}(\gamma_i - \gamma_{i-1})G(t-t_i)H(t-t_i)$. Multiplying and dividing by Δt, we can write the previous equation as,

$$\sigma(t) = \gamma_0 G(t)H(t) + \sum_{i=1}^{n} \frac{(\gamma_i - \gamma_{i-1})}{\Delta t} G(t-t_i)H(t-t_i)\Delta t. \tag{5}$$

Let $f(t,i) = \frac{\gamma_i - \gamma_{i-1}}{\Delta t} G(t-t_i)H(t-t_i)$, by taking the limit $\Delta \to 0$ and $n \to \infty$ we obtain a Riemann sum,

$$\sigma(t) = \gamma_0 G(t)H(t) + \lim_{\Delta t \to 0, n\to\infty} \sum_{i=1}^{n} f(t,i)\Delta t. \tag{6}$$

and $\frac{\gamma_i - \gamma_{i-1}}{\Delta t} \to \gamma'(t') = \frac{d\gamma}{dt'}$ with $t' \in [t_{i-1}, t_i]$.

If the sum exists, then the stress can be written as $\sigma(t) = \gamma_0 G(t)H(t) + \int_{0^+}^{t} G(t-t') \frac{d\gamma}{dt'} dt'$ or,

$$\sigma(t) = \int_0^t G(t-t') \frac{d\gamma}{dt'} dt' \tag{7}$$

where the integral includes the jump discontinuity in strain at the origin.

It should be remarked that the Boltzmann Principle lead to a huge discussion among the scientific community [3]. With most scientist being against or criticising this principle. The principle results were *presented* experimentally in 1876 by F. Kohlrauaeh using the torsion of a rubber filament [4].

Keeping in mind the relaxation experiment (figure (1)), and the fact that (see equation (1)) $G(t-t') = \frac{\sigma(t)}{\gamma}$ (with γ a constant along the relaxation experiment), we expect G to a have an exponential decay, $G(t) = G_0 e^{\frac{-t}{\lambda}}$, leading to the following relationship between stress and strain (known as the integral version of the Maxwell model),

$$\sigma(t) = \int_0^t G_0 e^{-\frac{t-t'}{\lambda}} \frac{d\gamma}{dt'} dt' \tag{8}$$

where G_0 and λ are two fitting parameters to be obtained from experimental results.

The differential model can be easily obtained by differentiating in time both sides of equation (8), leading to:

$$\sigma(t) + \frac{\eta}{G_0} \frac{d\sigma(t)}{dt} = \eta \frac{d\gamma(t)}{dt} \tag{9}$$

where $\eta = \lambda G_0$.

This differential equation is known as Maxwell model, because, it was presented by James Clark Maxwell, without much explanation and while studying gases (in 1867). Boltzmann allowed this model to become popular.

1.2 The *spring and damper* analogy - year 1903

J.H. Poynting and J.J. Thomson [6] introduced in 1903 the *spring and damper (or dashpot)* analogy. They introduced the Maxwell model by using a spring and a dashpot and the representing the Hookean and Newtonian models, respectively (for a more detailed explanation on these models please see [7, 8, 9, 10]). For ease of understanding we can say that the stress felt by stretching a spring is proportional to the deformation, γ_e (Hooke's law) and the stress felt by stretching a dashpot is proportional to the velocity of that deformation, $\frac{d\gamma_f}{dt}$ (a smooth stretch leads to a low stress and and a rapid stretch leads to a high stress. This technology is used to prevent doors from slamming). This mechanical analogue is illustrated in figure (5) (a).

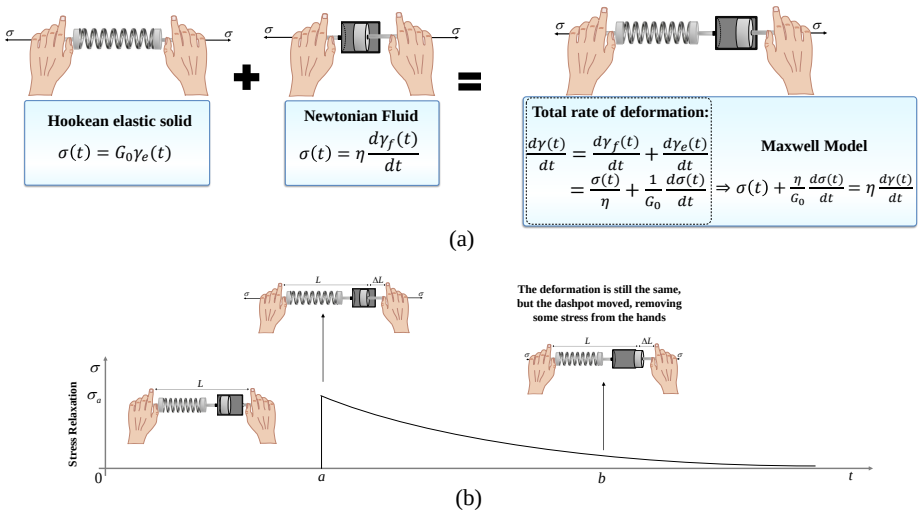

Figure 5: (a) Mechanical analogue of the Maxwell model. Assuming the total rate of deformation is given by the sum of the viscous and elastic contributions, then the Maxwell model is obtained; (b) Relaxation test viewed as a spring and a dashpot being deformed.

Figure (5) (b) shows what mechanically would happen if we use a spring and a dashpot in series. After the constant stretch, the dashpot starts to absorb the stress from the spring, and the total stress felt by the hands starts to decrease (it goes to 0). Several models were proposed in the literature using springs and dashpots in series, in parallel, and in different numbers and formats, and also using molecular theory [11, 12, 13].

1.3 The Relaxation and Memory Problem

The Maxwell model is not able to deal with all sort of viscoelastic materials. Even if we think of just one type of viscoelastic material, there may be regions in the same material that present different relaxations from each other.

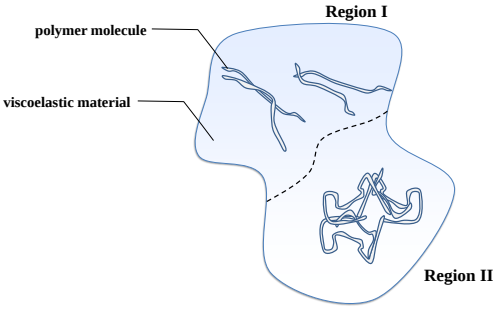

Figure 6: Different states of relaxation in the same material.

That is, the big molecules may be more stretched or more relaxed in the different

portions of material (see figure (6)).

Some solutions were proposed in the literature to deal with this problem.

- Prony series: the different regions of the material are modelled by using more than one Maxwell model (or other viscoelastic model), each one adding up a contribution to the total stress observed in that material (see figure (7)).

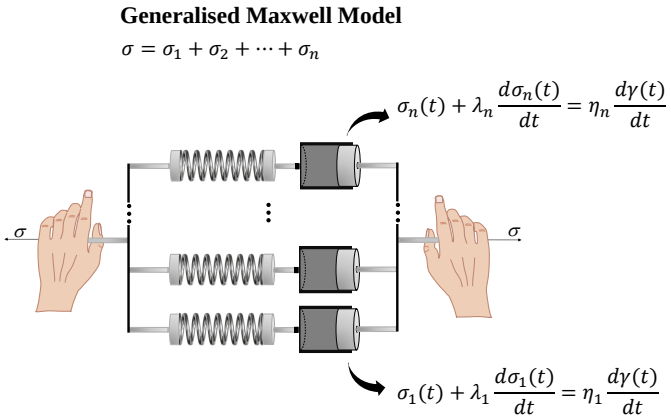

Figure 7: The generalised Maxwell Model.

- Different relaxation functions: The Maxwell-Debye relaxation (exponential decay) is observed in several complex viscoelastic fluids, but there are other materials showing different types of fading memory, such as an algebraic decay, $G(t) = St^{-\alpha}$, with $0 < \alpha < 1$ and S a scalar measure of the strength [14].

  If we re-write the relaxation modulus in the form $G(t-t') = \frac{\mathbb{V}}{\Gamma(1-\alpha)}(t-t')^{-\alpha}$, then, equation (8) can be written as,

$$\boldsymbol{\sigma}(t) = \frac{1}{\Gamma(1-\alpha)} \int_0^t \mathbb{V}(t-t')^{-\alpha} \frac{d\boldsymbol{\gamma}}{dt'} dt'. \tag{10}$$

  The fractional derivative in the Caputo sense ($0 < \alpha < 1$) is given by [15]:

$$ {}_0^C D_t^\alpha f(t) = \frac{1}{\Gamma(1-\alpha)} \int_0^t (t-t')^{-\alpha} \frac{df}{dt'} dt'. \tag{11}$$

  The constitutive equation for a material exhibiting relaxation $St^{-\alpha}$ can therefore be re-written as $\boldsymbol{\sigma}(t) = \mathbb{V}\, {}_0^C D_t^\alpha \boldsymbol{\gamma}(t)$. Using the compact notation ${}_0^C D_t^\alpha \equiv \frac{d^\alpha}{dt^\alpha}$, we have, $\boldsymbol{\sigma}(t) = \mathbb{V}\frac{d^\alpha \boldsymbol{\gamma}(t)}{dt^\alpha}$ where $\mathbb{V}$ is a constant for a fixed α, with physical dimensions $Pa.s^\alpha$.

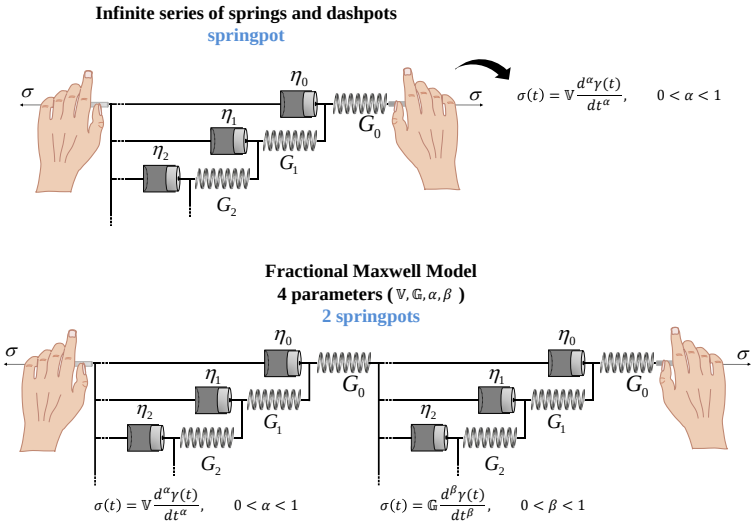

Figure 8: The springpot and the Fractional Maxwell Model.

$\mathbb{V}$ is a generalised modulus or a *quasi-property* [16, 17, 18, 19, 8]. This is illustrated in figure (8) by a springpot (a combination of springs and dashpots).

Two springpots arranged in series lead to the Fractional Maxwell Model,

$$\boldsymbol{\sigma}(t) + \frac{\mathbb{V}}{\mathbb{G}} \frac{d^{\alpha-\beta}\boldsymbol{\sigma}(t)}{dt^{\alpha-\beta}} = \mathbb{V}\frac{d^{\alpha}\boldsymbol{\gamma}(t)}{dt^{\alpha}}, \tag{12}$$

where it has been assumed (without loss of generality) that $0 < \beta \leq \alpha < 1$.

This four parameter linear viscoelastic model is able to describe a much wider range of complex fluid behaviour when compared to the classical Maxwell model (obtained in the limit $\alpha = 1, \beta = 0$).

- Use of integral models in combination with appropriate memory and damping functions. This subject falls outside of the scope of this work. See [8, 11, 16, 20] for more details.

2 THE DISTRIBUTED ORDER MODEL

The models presented before allow a good description of the behaviour of viscoelastic materials. We now present a more generalised model with improved fitting capabilities. This model is based on the distributed order fractional derivative.

2.1 Basic Definitions

Definition 2.1. *Distributed Order Fractional Derivative*
The Caputo Distributed Order Fractional Derivative (${}_0^C\mathbb{D}_t$) *of a general function* f *is given by:*

$$ {}_0^C\mathbb{D}_t f(t) = \int_0^1 c(\alpha) {}_0^C D_t^\alpha f(t) \; d\alpha = \int_0^1 c(\alpha) \frac{1}{\Gamma(1-\alpha)} \int_0^t (t-t')^{-\alpha} \frac{df}{dt'} dt' \; d\alpha \tag{13} $$

where the function $c(\alpha)$ is acting as weight for the order of differentiation is such that ([21], [22]) $c(\alpha) \geq 0$ and $\int_0^1 c(\alpha) \; d\alpha = C > 0$.

The function $c(\alpha)$ is used to represent mathematically the presence of multiple memory formalisms. If $c(\alpha) = \delta(\alpha - \beta)$, where $\delta()$ is the delta Dirac function, then (13) reduces to the Caputo derivative ${}_0^C D_t^\beta f(t)$. Note that the dimensions of $c(\alpha)$ are $[time]^\alpha / [length]^2$.

2.2 A Distributed Order Viscoelastic Model (DOVM)

As seen before, there are different types of fading memory, such as an algebraic decay, $G(t) = St^{-\alpha}$. Now assume that a material shows a combination of algebraic decays, such as,

$$ G(t) = S_1 t^{-\alpha_1} + S_2 t^{-\alpha_2} + ... + S_n t^{-\alpha_n}, \tag{14} $$

with $0 < \alpha_i < 1$, $i = 1, ..., n$.

Now assume that $n \to \infty$, covering the open set $(0, 1)$. Then we can write a continuous version of the previous finite combination of algebraic decays, which is given by:

$$ G(t) = \int_0^1 S(\alpha) t^{-\alpha} \; d\alpha. \tag{15} $$

The relaxation modulus can then be written in the form:

$$ G(t - t') = \int_0^1 \frac{c(\alpha)}{\Gamma(1-\alpha)} (t-t')^{-\alpha} \; d\alpha. \tag{16} $$

Inserting equation (16) into (7), we obtain:

$$ \sigma(t) = \int_0^t \int_0^1 \frac{c(\alpha)}{\Gamma(1-\alpha)} (t-t')^{-\alpha} d\alpha \frac{d\gamma}{dt'} dt' \tag{17} $$

This equation can be rewritten as (changing the integration order):

$$ \sigma(t) = \int_0^1 \int_0^t \frac{c(\alpha)}{\Gamma(1-\alpha)} (t-t')^{-\alpha} \frac{d\gamma}{dt'} dt' d\alpha = \int_0^1 \frac{c(\alpha)}{\Gamma(1-\alpha)} \int_0^t (t-t')^{-\alpha} \frac{d\gamma}{dt'} dt' d\alpha \tag{18} $$

In a more compact form, we have that (using definition (2.1)):

$$ \sigma(t) = {}_0^C \mathbb{D}_t \gamma(t). \tag{19} $$

We just proved the following lemma:

Lemma 2.1. *If the relaxation modulus is given by* $G(t-t') = \int_0^1 \frac{c(\alpha)}{\Gamma(1-\alpha)}(t-t')^{-\alpha}\, d\alpha$, *then the hereditary integral proposed by Boltzmann* ($\sigma(t) = \int_0^t G(t-t')\frac{d\gamma}{dt'}dt'$), *is given by:*

$$\sigma(t) =_0^C \mathbb{D}_t\gamma(t). \tag{20}$$

Definition 2.2. *The Distributed Order Fractional Viscoelastic Model (DOVM) is given by*

$$\sigma(t) =_0^C \mathbb{D}_t\gamma(t). \tag{21}$$

were ${}_0^C\mathbb{D}_t$ *is the distributed order derivative of the Caputo type.*

For another approaches on the distributed order derivative please see [23, 24, 25, 26, 27]. These papers also discuss the thermodynamics admissibility of distributed order equations (imposing restrictions on the model parameters).

3 Properties of the Distributed Order Viscoleastic Model

We now present some properties of the model.

3.1 The Relaxation Modulus

In the FMM model, we have a relaxation modulus of the form $St^{-\alpha}$. The success of this relaxation relies on the fact that it is more general, and, it leads to non-integer order derivatives that incorporate an extra parameter, α, allowing in this way to better represent the different rates of deformation and relaxation in a material under stress. This means that if a have a part of the material evolving at a rate of 0.9 and other part of the material evolving at a rate of 0.4, perhaps a first order derivative is not the more appropriate derivative to model this phenomenon.

The relaxation modulus in the DOVM is given by by equation (16), meaning that we can cover the all range of α values, and, attribute weights according the different contributions of the α values. We have a sum of weighted fractional derivatives. Now the question is: how to choose a proper weight function $c(\alpha)$?

The answer to this question is not easy. A possible solution would be:

- first, find the simple fractional order derivatives (${}_0^C D_t^\alpha$) that better represent the phenomenon being modelled (by comparing the model results with the experimental data). $S(\alpha)$ can be seen as a generic function that interpolates $(\alpha_i, S_i), i = 1, ..., n$ (in practice we only have a finite number of S_i);

- Second, derive a weight function $c(\alpha)$ that attributes more weight to the order derivatives found in the first step;

- Repeat steps 1 and 2 until a *good* degree of fitting is obtained with the DOVM.

If the $c(\alpha)$ function is already given, the fitting should be performed using an optimization algorithm.

We now present some weight functions and the evolution in time of the relaxation modulus (figure (9)).

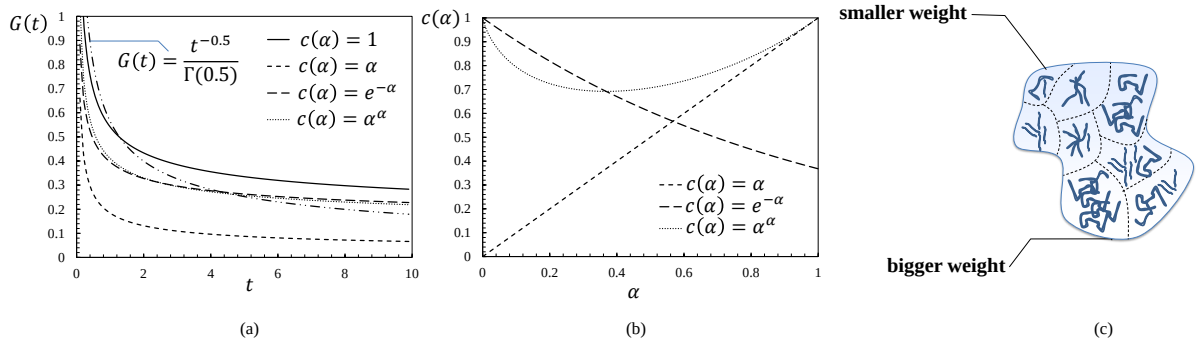

Figure 9: (a) Relaxation modulus; (b) Weight functions; (c) Portion of material divided into different regions of relaxation. More weight should be given to the dominant regions.

In figure (9) (a) is presented the relaxation modulus using the different weight functions presented in figure (9) (b). For example, using $c(\alpha) = \alpha^\alpha$, we are attributing more weight to the low and high order derivatives. Figure (9) (c) shows a portion of material divided into different regions of relaxation. More weight should be given to the dominant regions.

It should be remarked that the chosen $c(\alpha)$ functions should be consistent with the principles of thermodynamics and result in a non-negative internal work and a non-negative rate of energy dissipation [23, 24].

3.2 The Storage Modulus and Loss Modulus

The Storage Modulus and Loss Modulus allows one to study the way our model behaves when an oscillatory deformation is imposed. We are able to see how much is recovered (Hook's law) and how much is lost (Newtonian law) when the deformation is applied. the storage and loss are represented by G' and G'', respectively. Figure (10) illustrates a real experiment where the loss and storage can be measured experimentally and a relationship between the experimental results and the Maxwell model can be established. The γ_0 represents the amplitude of the oscillation (it should be small because we aim the linear viscoelastic regime) and ω is the frequency (how fast we rotate the upper plate). By using different values of ω we can see how the material behaves. If G' is bigger than G'', it means that the fluid shows a more elastic behaviour, If G' is smaller than G'' the fluid shows a more viscous behaviour. Note that G' and G'' depend on how fast we rotate de upper plate, that is, they depend on ω. Different behaviours will be observed depending on the value of ω.

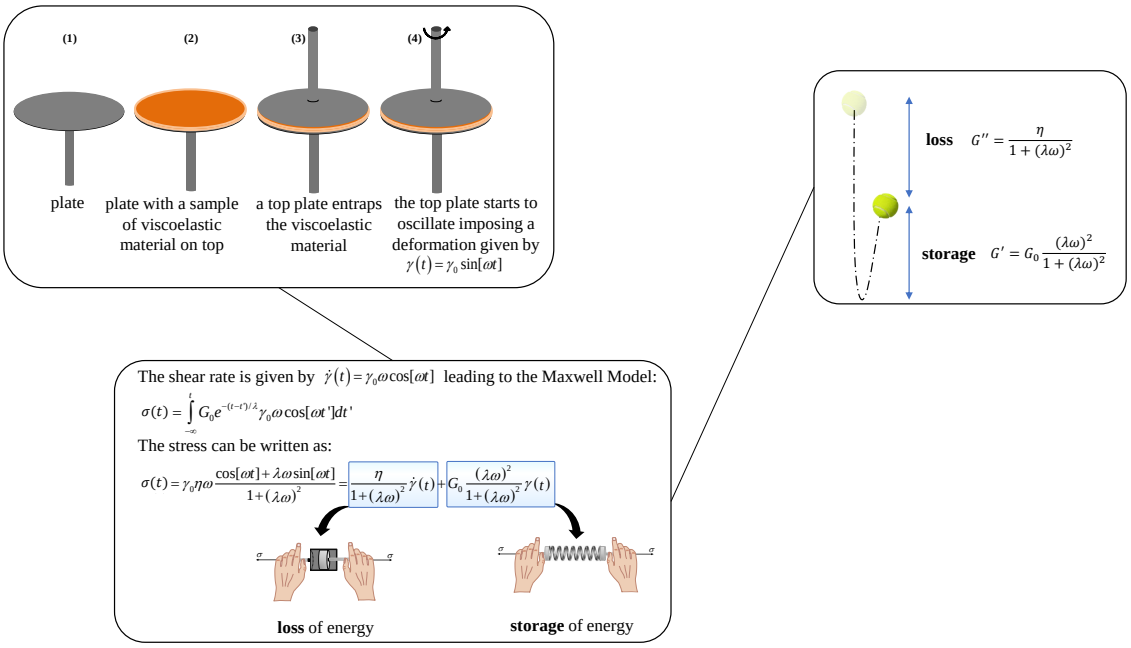

Figure 10: The Storage Modulus (G') and Loss Modulus (G''). The stress is written as a sum of two components. One component proportional the rate of deformation (as in Newtonian fluids) and the other component proportional to the deformation (as in Hook's law). The dashpot loses energy, while the spring stores energy.

The G' and G'' can be obtained in a different way. Using the Laplace Transform, $\mathcal{L}$, and the convolution theorem, we have that, for the Maxwell model (assuming that $\sigma(t)=0$ for $t<0$)

$$\mathcal{L}\{\sigma(t)\}=\bar{\sigma}(s)=\mathcal{L}\left\{\int_0^t G_0e^{-\frac{t-t'}{\lambda}}\frac{d\gamma}{dt'}dt'\right\}=\int_0^\infty G_0e^{-\frac{t}{\lambda}}e^{-st}dt\int_0^\infty\frac{d\gamma(t)}{dt}e^{-st}dt=\frac{sG_0\lambda\bar{\gamma}(s)}{1+s\lambda}\quad(22)$$

That is, in the transform space, the Laplace transform of $\sigma(t)$ and $\gamma(t)$ ($\bar{\sigma}(s)$ and $\bar{\gamma}(s)$) are proportional, as in a linear elastic material (and therefore the symbol G^* to represent this *modulus*),

$$\frac{\bar{\sigma}(s)}{\bar{\gamma}(s)}=\frac{sG_0\lambda}{1+s\lambda}=G^*(s).\quad(23)$$

Now assume that $s=i\omega$ ($i=\sqrt{-1}$), we have that,

$$G^*(i\omega)=\frac{G_0(\lambda\omega)^2}{1+(\lambda\omega)^2}+i\frac{\eta\omega}{1+(\lambda\omega)^2}=G'+iG'',\quad(24)$$

as in figure (10).

Lemma 3.1. *The storage and loss modulus for the DOVM are given by:*

$$G' = \int_0^1 c(\alpha)\omega^\alpha \cos\left(\frac{\pi}{2}\alpha\right)\ d\alpha \tag{25}$$

$$G'' = \int_0^1 c(\alpha)\omega^\alpha \sin\left(\frac{\pi}{2}\alpha\right)\ d\alpha \tag{26}$$

Proof. Using the same procedure for the DOVM as in the Maxwell model, we have that,

$$\bar{\sigma}(s) = \int_0^1 c(\alpha)\mathcal{L}\{{}_0^C D_t^\alpha \gamma(t)\}\ d\alpha = \bar{\gamma}(s)\int_0^1 c(\alpha)s^\alpha\ d\alpha \tag{27}$$

Using the fact that $s = i\omega$ and $(i\omega)^\alpha = \omega^\alpha \cos\left(\frac{\pi}{2}\alpha\right) + i\omega^\alpha \sin\left(\frac{\pi}{2}\alpha\right)$, we have:

$$G^*(i\omega) = \int_0^1 c(\alpha)\omega^\alpha \cos\left(\frac{\pi}{2}\alpha\right)\ d\alpha + i\int_0^1 c(\alpha)\omega^\alpha \sin\left(\frac{\pi}{2}\alpha\right)\ d\alpha = G' + iG'' \tag{28}$$

□

3.2.1 Storage and Loss Functions for the DOVM

For some particular cases of $c(\alpha)$ it is possible to obtain a closed form solution for G' and G''. A few of these particular cases are presented next.

- $c(\alpha) = C$
  $G' = \frac{2C(\pi\omega - 2\log(\omega))}{4\log^2(\omega)+\pi^2}$, $\quad G'' = \frac{2C(2\omega\log(\omega)+\pi)}{4\log^2(\omega)+\pi^2}$
- $c(\alpha) = Ce^{-\alpha}$
  $G' = \frac{2C\left(\frac{\pi\omega}{e} - 2\log(\omega)+2\right)}{4(\log(\omega)-2)\log(\omega)+\pi^2+4}$, $\quad G'' = \frac{2C(-2\omega+2\omega\log(\omega)+e\pi)}{e\left(4\log^2(\omega)-8\log(\omega)+\pi^2+4\right)}$
- $c(\alpha) = C\alpha$
  $G' = \frac{2C\left(\pi^2(\pi\omega-2)+4(\pi\omega+2)\log^2(\omega)-8\pi\omega\log(\omega)\right)}{\left(4\log^2(\omega)+\pi^2\right)^2}$
  $G'' = \frac{4C\left(\pi^2\omega+\log(\omega)(\pi(\pi\omega-4)+4\omega(\log(\omega)-1)\log(\omega))\right)}{\left(4\log^2(\omega)+\pi^2\right)^2}$
- $c(\alpha) = C\alpha^2$
  $G' = \frac{2C\left(\pi^3\left(\pi^2-8\right)\omega+16\pi\omega\log^4(\omega)-64(\pi\omega+1)\log^3(\omega)+8\pi\left(12+\pi^2\right)\omega\log^2(\omega)-16\pi^2(\pi\omega-3)\log(\omega)\right)}{\left(4\log^2(\omega)+\pi^2\right)^3}$
  $G'' = \frac{4C\left(2\pi^3(\pi\omega-2)+\log(\omega)\left(\pi^2\left(\pi^2-24\right)\omega+8\log(\omega)\left(\omega\log(\omega)\left(2(\log(\omega)-2)\log(\omega)+\pi^2+4\right)+6\pi\right)\right)\right)}{\left(4\log^2(\omega)+\pi^2\right)^3}$

- $c(\alpha) = C\alpha^n$
  $G' = \frac{(-2\log(\omega)-i\pi)^{-n-1}\left(-\log(\omega)+\frac{i\pi}{2}\right)^{-n}(C(\pi+2i\log(\omega))(-2\log(\omega)+i\pi)^n B - c(\pi-2i\log(\omega))(-2\log(\omega)-i\pi)^n A)}{\pi+2i\log(\omega)}$
  $G'' = \frac{C(-2\log(\omega)+i\pi)^{-n}\left(-\log(\omega)-\frac{1}{2}(i\pi)\right)^{-n}((\pi-2i\log(\omega))(-2\log(\omega)-i\pi)^n(A)+(\pi+2i\log(\omega))(-2\log(\omega)+i\pi)^n(B))}{4\log^2(\omega)+\pi^2}$
  with $0 < \omega < 1$.

where C is a real constant, $n \geq 0$, log is the natural logarithm, $A = \Gamma(n+1) - \Gamma\left(n+1, \frac{i\pi}{2} - \log(\omega)\right)$, $B = \Gamma(n+1) - \Gamma\left(n+1, -\frac{1}{2}(i\pi) - \log(\omega)\right)$.

It should be remarked that for more complex functions one can obtain the solution numerically for each value of ω. These closed form solutions for G' and G'' should be used as a guide to find more suitable functions for $c(\alpha)$.

3.3 Fit to Experimental Data

In this subsection we present the fitting of the DOVM to the experimental data obtained by Ng and McKinley [28] (G' and G'') for a strong wheat flour, mixed to a constant time (360s/peak-mixed) and a fixed water ratio by weight (66%).

For the fitting we have used the function:

$$c(\alpha) = \frac{S}{\sqrt{\pi}a} \exp\left(-\frac{(\alpha-b)^2}{a^2}\right) \tag{29}$$

with S the strength of the bread-dough the critical gel point. The reason for choosing this function is due to the fact that when $S = 1$, this function approximates the delta Dirac function $\delta(\alpha - b)$. The smaller the value of a, the better is the approximation.

Based on the results presented in [28], we have choosen an $b = \frac{1}{5}$ as the most important order of the fractional derivative.

Figure (11) shows the experimental data (G' and G'') and the fitting obtained with the Maxwell, Fractional Maxwell (FMM) and DOVM models. Nat that it was possible to obtain a closed form solution for G' and G'' using equation (29) (the mathematical expressions are too long to be presented here). The parameters used are: $\eta = 3000$, $\lambda = 0.03$ for the Maxwell model; $\mathbb{V} = 23865$, $\mathbb{G} = 8121$, $\alpha = 0.24$, $\beta = 0.2$ for the Fractional Maxwell Model; $S = 6000$, $a = \frac{1}{100}$ and $b = \frac{1}{5}$ for the DOVM.

The results obtained show that the fits obtained for the FMM and DOVM are much better when compared to the fit obtained for the Maxwell model. The reason for that is because the Maxwell model is more suitable to model materials for which the relaxation modulus shows an exponential decay (for example: polymer melts and polymer solutions) while the Fractional Maxwell Model is suitable for modelling materials that show both types of relaxation, an exponential and an algebraic decay (see [9, 16]). The DOVM seems to be only suitable to model materials for which the relaxation modulus shows an algebraic decay. Also, the Maxwell model has 2 parameters, the Fractional Maxwell

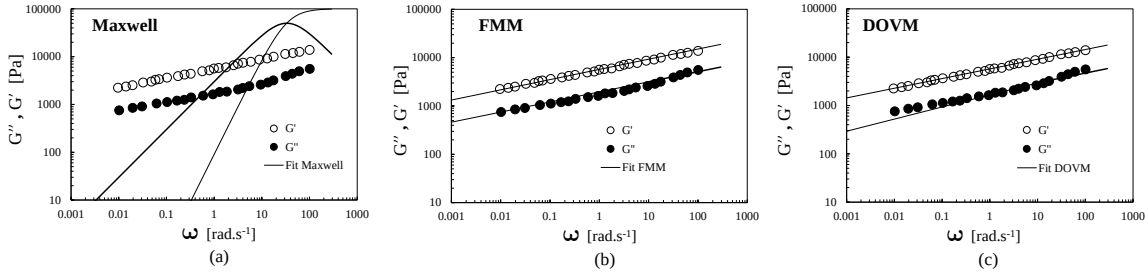

Figure 11: Fit to the Storage Modulus (G') and Loss Modulus (G'') obtained experimentally for bread dough [28]. (a) Fit obtained with Maxwell model; (b) Fit obtained with the Fractional Maxwell Model; (C) Fit obtained with the DOVM.

Model has 4 parameters, and DOVM has (in this particular case) 2 parameters (S and b).

To quantify the error incurred during the fitting process we used a mean square error given by:

$$\varepsilon = \sum_i \left[\log G'_i - \log G'_{fit}(\omega_i)\right]^2 + \sum_i \left[\log G''_i - \log G''_{fit}(\omega_i)\right]^2. \tag{30}$$

The errors are given by $\varepsilon_{Maxwell} = 221.95$, $\varepsilon_{FMM} = 7.54 \times 10^{-2}$, $\varepsilon_{DOVM} = 4.04 \times 10^{-2}$, for the Maxwell, FMM, and DOVM, respectively. Based on these results, we may say the DOVM model shows a good performance with a less number of parameters. However, one should not forget that we have first to derive the function $c(\alpha)$.

In the future, more fittings should be performed considering a broader range of materials. This will allow us to derive more robust weighting functions $c(\alpha)$.

4 CONCLUSIONS

The final considerations and main conclusions of this work are:

- A generalised viscoelastic model for small deformations was derived from the Boltzmann theory;
- The new model has a more general relaxation function, allowing its use for modelling a broader range of viscoelastic materials showing a power law relaxation;
- The storage and loss functions were derived for special cases of $c(\alpha)$ (the function that distributes the weights through the different order derivatives);
- The model is only valid for small deformations and requires further studies in order to understand how to derive the $c(\alpha)$ functions for different viscoelastic materials.

- The model can be easily generalised for large deformations (rheologically admissible constitutive equation) by using for example the Lodge rubber-like liquid form, combining the linear viscoelastic relaxation modulus presented in this work with finite strain kinematics.

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