Mechanical Modelling and Experimental Characterization of Laminated Glass with Multi-material Polymeric Interlayers

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Abstract

Multi-Material Polymeric Interlayers (MMPI) combine different polymers between two or more glass plies. Typically, a soft polymer sheet is used as the core layer, while two stiffer sheets encase this core. MMPIs are widely used in various industries, such as construction and automotive, for their ability to impart hybrid characteristics to laminated glass structures. For instance, an acoustic monolayer PVB can be used as the core layer to enhance sound insulation, while a stiff PVB is applied as the outer layer to increase structural stiffness. Modelling the mechanical behaviour of MMPIs is challenging due to their time- and temperature-dependent rheological properties. Accurately predicting the relaxation function of a stacked interlayer requires combining the relaxation functions of each material through a comprehensive viscoelastic analysis, where the current strain depends not only on the actual stress but also on the entire stress history. A novel approach involving fractional calculus is here used for a comprehensive viscoelastic characterization of MMPIs. The fractional derivatives are numerically approximated using the L1 formula, which allows a variable time-step in the computations. The basic assumption is that the relaxation functions are represented by continuously connected power-law branches. This is a faithful representation for the response of many commercial polymers, which simplifies parameter determination from experimental data and enables easier and more computationally efficient numerical implementation. The fractional approach offers significant advantages in accuracy, efficiency, and simplicity compared to the traditional method using Prony series of exponential functions, making it a promising method of analysis. In this study, we combine the single relaxation functions of the interlayers in a laminated glass package to obtain the MMPI's, comparing model predictions with experimental data. We also highlight potential differences from the quasi-elastic approach, which models the polymers as linear-elastic materials with a temperature- and time-dependent secant shear modulus.

Keywords

Laminated glass; polymeric interlayer; multi-material interlayer; viscoelastic properties; fractional calculus.

Article Information

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1. Introduction

Laminated glass consists of a layered structure made up of two or more glass sheets bonded together by one or more thin, flexible thermoplastic polymer interlayers. These interlayers are considered "flexible" because they lack significant axial or flexural stiffness, and their function is to provide shear coupling between the glass layers. The most used commercial interlayers include polyvinyl butyral (PVB), Ionoplast SentryGlas® (SG), and Ethylene-Vinyl Acetate (EVA). These materials are available in various formulations, depending on the amount of plasticizers and metal salts added, as well as the specific processing methods used (M. Martin, 2020). The interlayer capacity of coupling the glass plies varies between the upper bound of full coupled glass plies (*monolithic limit*) and the opposite lower bound of free-sliding plies (*layered limit*). The viscoelastic materials provide a condition that varies in time within these two limit cases. The traditional approach to interpreting the relaxation function of a polymeric material involves the use of Prony series, founded on the Maxwell-Wiechert model. This is the most used framework for describing linear viscoelasticity (Biolzi, Cattaneo, Orlando, Piscitelli & Spinelli, 2020). It accounts for the fact that relaxation does not occur at a single time but over a range of times. The relaxation curve is represented as a sum of exponential terms, each modelled as a spring-dashpot element, with different decay times. Additionally, a single spring represents the material's stiffness at infinite time, when all dashpots are fully relaxed. However, experimental studies on a broad range of materials, particularly on most commercial polymers used as interlayers, suggest that the relaxation function can be effectively approximated by segments of power laws in time (Viviani, Di Paola, and Royer-Carfagni, 2022). When plotted on a bi-logarithmic scale of secant shear modulus versus time, these power laws appear as a polyline, with each segment fully defined by two parameters: its slope and its intercept with a vertical axis. When the relaxation function of a viscoelastic material follows power laws, rheological models based on fractional calculus become highly effective. A fractional derivative, which is a derivative of any arbitrary order (real or complex), aligns with Boltzmann's convolution integral when the relaxation function is a power law.

Multi-Material Polymeric Interlayers (MMPI) for laminated glass are often employed when a single-foil interlayer fails to deliver the desired combination of properties. The primary application of MMPI is to improve acoustic insulation. In such applications, a soft PVB core layer, specifically optimized for acoustic damping, is sandwiched between standard PVB layers that are tailored for strong adhesion to glass, toughness, and ease of lamination processing. Naturally, the mechanical behaviour of MMPI differs from that of single-layer products. It is important to note that very short load durations become particularly relevant in practical applications at low temperatures, as this effectively shifts the performance curve toward longer times. It is difficult to estimate the mechanical performance of such MMPI because every material shows its own long-term behaviour, and, when combined, the overall relaxation curve is intermediate. The thickness of each layer in the composite also plays a significant role in the determination of the mechanical response. It is possible to estimate such relaxation curve through Dynamic Mechanical Thermal Analysis (DTMA), but this method is usually expensive and time consuming; moreover, they can be errors in the measurement of the experimental data, because the testing machine is sophisticated and needs proper calibration. A numerical simulation able to predict the relaxation curve with good accuracy, starting from the relaxation curves of the single materials, can help in the design of MMPI and provide guidance for optimization.

Here, we present a numerical tool based on the power law interpolation of the relaxation curve of polymeric materials, which can predict the relaxation curve of an MMPI using as input data the relaxation curves of the single materials, usually available from the producers. Specific reference is made to three main products by Kuraray GmbH: Ionoplast SentryGlas®, Stiff PVB and Acoustic PVB at various operating temperatures (20^{o} C, 30^{o} C and 40^{o} C).

2. The model

Interlayers composed of several individual layers of different materials can exhibit complex mechanical behavior under various loading conditions. This is dictated by factors such as material properties, layer thickness, and the arrangement of the individual layers. Understanding the mechanical behavior of multilayers is crucial for designing and optimizing structures that utilize these materials. We demonstrated a theoretical method for predicting the mechanical behavior of a multilayer in a relaxation test, starting from the material model parameters and layer thicknesses. This method is also implemented in a finite element framework, which allows us to predict the relaxation function of a complex MMPI starting from the experimental relaxation function of each single material determined via DTMA analysis.

2.1. Description via fractional calculus

The viscoelastic properties of interlayer materials are characterized by their relaxation function. This function reflects the material's long-term creep behavior and demonstrates how stiffness diminishes over time. For interlayers, the relaxation function is typically determined experimentally by applying a constant strain and observing the corresponding stress reduction, specifically the decay in the secant elastic modulus of the uniformly strained sample over time. Alternatively, it can be obtained through dynamic experiments and Time-Temperature Superposition (TTS), as outlined in the DMA approach also mentioned by EN 16613. In structural modelling, the relaxation function of the polymer serves as a critical input. Boltzmann's superposition principle is used to describe linear viscoelastic behavior.

In laminated glass beams and plates, the relaxation curve determines the time-dependent shear-coupling capacity of the polymeric interlayer.

The conventional method for interpolating the experimental points involves the use of Prony series, which is expressed as a summation of exponential terms of the form $R_i e^{-t/\vartheta_i}$, where $R_i$represents the i-th relaxation shear modulus and ϑ_i denotes the corresponding relaxation time (Gant & Bower, 1997). This series corresponds to the Wiechert model of viscoelasticity, composed of an array of Maxwell units arranged in parallel with a spring of stiffness R_0. Such spring represents the residual stiffness of the viscoelastic material as time approaches infinity. Therefore, the relaxation function is given by

$$R(t) = R_0 + \sum_{i=1}^{N} R_i e^{-t/\vartheta_i} \qquad (1)$$

The form of the relaxation function indicates that an alternative interpolation can be achieved using continuously connected branches of power laws, each expressed as $C_\alpha t^{-\alpha}$, with $0 < \alpha < 1$ (Santi, Bennison, Haerth & Royer-Carfagni, 2023). In a bi-logarithmic plot, a power law appears as a straight line, implying that the curves in Figure 1 can be approximated by a polyline consisting of three segments, as illustrated for the material Acoustic PVB. Each segment is characterized by two parameters: the slope α of the line and C_α $[MPa\ s^\alpha]$, which represents the stiffness value at $t = 1\ s$. If the observation interval for the phenomenon is narrowed, or for materials such as Ionomers for which the curve can be well approximated by a single power law, fewer branches may suffice for the approximation (Santi & Royer-Carfagni, 2024).

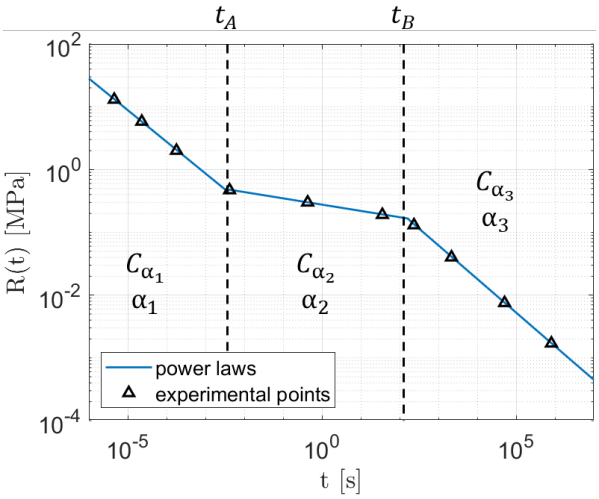

Fig. 1: Relaxation curves for a stiff PVB, tested at 20° C, approximated with branches of power laws. Three branches are needed to represent the relaxation curve for the entire observation time.

There is a mathematical description of the equations of viscoelasticity founded on fractional calculus, because Boltzmann's convolution integral coincides with the Caputo fractional derivative of order α when the relaxation function is expressed by a power law, that is

$$ {}_0^C D_t^\alpha [f(\cdot)](t) = \frac{1}{\Gamma(1-\alpha)} \int_0^t (t-\bar{t})^{-\alpha} \dot{f}(\bar{t}) d\bar{t} \qquad (2) $$

However, to establish such a precise correspondence, it is necessary to formally write the power-law terms by defining the coefficients in terms of Euler's Gamma function Γ, which is the generalization of the factorial *n*!, to non-integer or complex values of *n*.

We choose to use the power law approximation to represent the relaxation function of the polymeric materials, because such method presents several advantages over the classical approach for the purpose of this work: an easier and more intuitive representation of the curves that allows to build numerical faster and more reliable numerical models.

In the viscoelastic interlayer, from Boltzmann superposition principle, one obtains the constitutive equation takes the form:

$$ \tau(z,t) = \tau(z,0) R(t) + \int_0^t \frac{\partial \epsilon}{\partial \bar{t}} R(t-\bar{t}) d\bar{t} \qquad (3) $$

In general, it is supposed that at $t = 0$ the structure is undistorted, so that $\tau_{yz}(z,0) = 0$. If the relaxation function is a simple power law or a piecewise function of power laws, one obtains

$$ \tau(z,t) = \int_0^t \frac{\partial \gamma_{yz}}{\partial \bar{t}} \frac{C_\alpha}{\Gamma(1-\alpha)} (t-\bar{t})^{-\alpha} d\bar{t} = C_\alpha \, {}_0^C D_t^\alpha [\epsilon(z,\cdot)](t) \qquad (4) $$

This is the definition of Caputo fractional derivatives, as indicated in (2).

2.2. Shear response of multi-material interlayers

To predict the mechanical behaviour of a MMPI on the laminated structure, it is necessary to study the coupling effect of the interlayer on the two glass plies it connects. The relaxation functions of the materials that compose the MMPI are well known, and they represent the starting point of the analysis. The goal is to predict a single relaxation function that describes the coupling effect of the package, that is, the evolution of the stiffness in time. It is possible to extrapolate that relaxation function through

DTMA analysis and use it for the model validation, by comparing the numerical result and the experimental points.

The proposed model consists in a laminated glass structure with a MMPI composed by two sheets of Acoustic PVB used as outer layers and one single sheet of stiff PVB used as core layer. In Figure 2 we represent the relaxation-function's power-law interpolation of the two materials, $R_1(t)$ for Acoustic PVB (in yellow) and $R_2(t)$ for Stiff PVB (in violet), juxtaposed to the curve derived from their combination and their respective experimental points in red triangles. The curves are represented at different temperatures (20°C (a), 30°C (b), 40°C (c)), obtained by controlling the temperature of the machine platen used in the test.

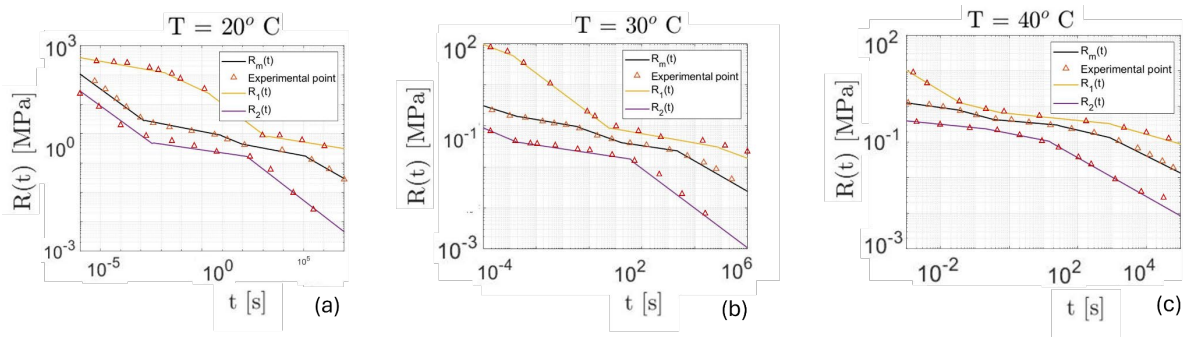

Fig. 2: Power law approximation of the relaxation function for two materials: $R_1(t)$ for Acoustic PVB (yellow) and $R_2(t)$ for Stiff PVB (violet), as well as the curve derived from their combination(black), with the experimental points represented by red triangles. The curves correspond to different environmental temperatures: 20°C (a), 30°C (b), 40°C (c).

The laminated glass package is subjected to a shear stress on the top surface, while the bottom surface is fixed by preventing the displacement in x-direction, as shown in Figure 3. The proposed system can be simplified by considering a single layer of material "2" with total thickness equal by the sum of the thicknesses in the original configuration and respective relaxation functions R_1. It is measured the displacement of the two materials ϵ_1 and ϵ_2 in x-direction, assuming that the total displacement of the upper glass layer is $\epsilon = \epsilon_1 + \epsilon_2$. This system is compared to the equivalent system where, instead of considering the two materials, the interlayer is composed by only one single material with thickness equal to the sum of the thicknesses of the two layers "1" and "2" in the previous scheme, i.e., $h_m = h_1 + h_2$, and with the equivalent relaxation function R_m measured from DTMA by considering the two materials. The total displacement of the upper glass layer should be the almost same in both systems $\epsilon \cong \epsilon_m$. Consider that there are always some uncertainties in the results, because the relaxation functions used as an input data are derived from experiments, subjected to a certain error in the measure.

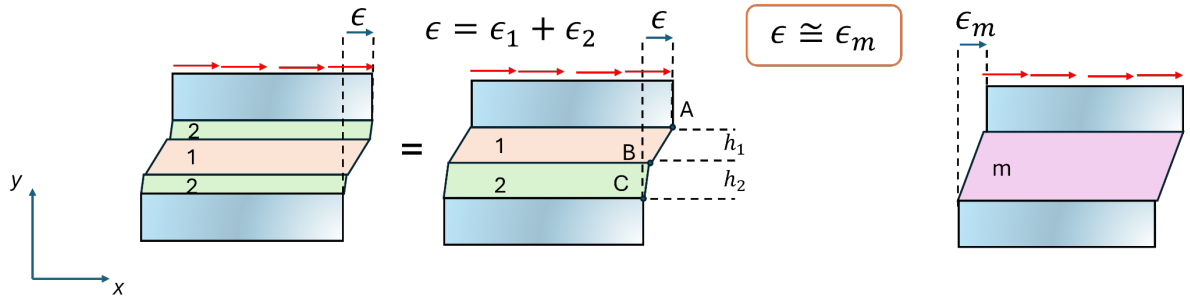

Fig. 3: Laminated glass sandwich package composed by MMPI. The green material "1" used in the core is Trosifol Extra Stiff PVB, while the orange material "1" is Acoustic PVB. The structure is subjected to shear stress on the top surface.

Considering the power law approximation of the experimental relaxation function, it is possible to describe the constitutive law of the polymeric interlayer with Equation (4), where the shear stress $\tau(z,t)$ in the material is equal to the α time derivative of the shear deformation multiplied by the stiffness constant C_α. The shear deformation is therefore

$$\epsilon_1(z,t) = \frac{1}{h_1}[u_A(z,t) - u_B(z,t)] \tag{5}$$

referred to the interlayer "1"; the relationship for the material "2" is analogous.

In elastic solids, stress is proportional to the zero-order derivative of strain, while in liquids, stress is proportional to the first derivative of strain. Therefore, it is natural to assume that for viscoelastic materials, stress is proportional to a real-order derivative, intermediate between 0 and 1 of the strain over time. This hypothesis leads to the conceptualization of a mathematical model of Spring-Pot, whose schematic representation is shown in Figure 4. The constitutive equation for the Spring-Pot can be described through Caputo's' fractional derivative definition in the generalized form $\tau(t) = C_\alpha {}_0^C D_t^\alpha \epsilon(t)$, where C_α and α can be obtained through a best-fit of experimental data. Although the Spring-Pot serves as an analytical model for simulating the viscoelastic behaviour, it lacks a complete physical interpretation. In classical models, it is possible to distinguish the contribution of the solid phase from that of the fluid phase, but in the Spring-Pot, such a distinction is not straightforward. Indeed, the term C_α, which represents the proportionality coefficient between stress and the fractional derivative of strain, lacks a clear physical definition, as it does not correspond to either an elastic modulus or a viscosity parameter.

We can schematize the laminated package with MMPI with two Spring-Pots in series, where the total displacement of the system is equal to the sum of their displacements and the stress is the same in every element, as indicated in Figure 4. The system of the laminated glass package with only one interlayer can be represented with one spring pot. The constants needed for the constitutive equation can be derived from the experimental data of the relaxation curve, by interpolating the points with a piecewise function of power laws, which correspond to lines in the bi-log graph. The slope of the lines is α , and C_α represents the stiffness value at $t = 1\ s$, in a specific temporal branch.

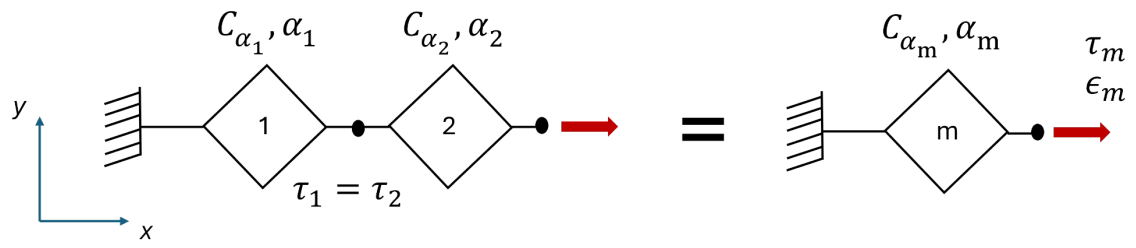

Fig. 4: Scheme of the problem presented in figure 1. The two materials the compose the interlayer are modelled as full viscoelastic materials with the fractional derivative constitutive law here represented as a spring pots.

The fractional derivatives in time of the constitutive equation (4) are numerically approximated using the L1 formula, which enables the use of nonuniform time meshes. This approach provides several key benefits. First, a time mesh in logarithmic scale progression allows for an accurate representation of each branch of the relaxation function over a wide range of time scales; in contrast, constant time steps would fail to interpolate all branches with comparable precision, resulting in a loss of accuracy in the numerical solution. Second, the variable time-step approach significantly reduces computational effort by requiring fewer steps to follow the relaxation function for long periods of observation, enabling efficient calculation over extensive observation times.

The numerical problem has been solved through a finite element code. specifically written in MATLAB and Python. This implements the constitutive equation of the polymeric interlayer based on fractional derivatives (numerically approximated with the L1 formula), made possible by the approximation of their relaxation function with power laws. The Finite Element Fractional Viscoelastic (FVFV) model is described in detail in (Santi & Royer-Carfagni, 2025), where it has been used for the simulation of the long-term viscoelastic behaviour of a laminated glass structure in a four-point bending test, demonstrating its accuracy by comparison with experimental results. The numerical approximation of fractional derivatives can be also performed via the Grünwald-Letnikov approximation (Scherer, Kalla, Tang & Huang, 2011), which is very efficient since it provides the direct construction of a triangular matrix that operates on the discretized array of values of the relevant variables. The triangular structure of this matrix facilitates the numerical solution. However, one of the major drawbacks of this method is that it is based on a discretization in constant time steps: when the interval of observation is wide, too many steps would be needed to describe the long-term response. On the other hand, enlarging the time step results in a loss of accuracy. The major advantage of using the L1 formula in the finite element code is that it is compatible with a non-constant time-mesh size. A comparison between the Grünwald-Letnikov and the L1 formula approaches is presented in (Santi & Royer-Carfagni, 2024).

The finite element scheme for the numerical resolution of this problem is based on a two-node element with three degrees of freedom, corresponding to the displacement along the x-direction of the points A, B and C indicated in Figure 3.

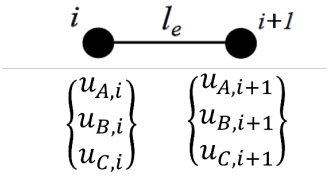

Fig. 5: Finite element scheme of the model used to solve the numerical problem in figure 1. The element is described with node with four degrees of freedom each.

This mathematical treatment allows to calculate the deformation of the two different systems represented in Figure 4 and compare their deformations. Once we know the deformation of the system with the two spring pots in series, it is possible to calculate the stress trend over the observation time, and in the end, by reversing Equation (3), it is possible to find the trend of the effective relaxation function $R(t)$, which describes the behaviour of the laminated glass structure with the MMPI. For the model validation, the relaxation function $R(t)$ is compared to the relaxation curve's experimental points, preliminary obtained with DTMA and showed in Figure 2.

It is important to point out that the FEFV model furnishes a full viscoelastic characterization of the stress in the polymeric material, which implies that the strain in a specific observation time step does not only depend on the stress in the same time step, but also to the previous state of stresses starting from the beginning of the load application on the material. This is mathematically described by the convolution integral of the Boltzmann superposition principle (Equation 3).

3. Numerical results and model validation

The deformation trends in time obtained through the FEFV model are showed in Figure 6, where the three cases at different environmental temperatures 20°C (a), 30°C (b), 40°C (c) are compared, when using the relaxation functions of the two materials (R_1= Stiff PVB and R_2= Acoustic PVB) in the spring pots series (black dashed line,) and when using the relaxation function of the same two combined materials with red dashed lines, as showed in Figure 4. The shear deformations are obtained with Equation 5, using the output values of the FEFV model; in the first case the total deformation is obtained by summing up the deformations of the two materials, i.e., $\epsilon = \epsilon_1 + \epsilon_2$.

The two deformation trends are expected to be almost the same ($\epsilon \cong \epsilon_m$); from Figure 6 it is possible to observe that this is verified with always some uncertainty, which results from the inevitable measurement errors I the DTMA analysis of the relaxation function, used as an input data for the numerical model.

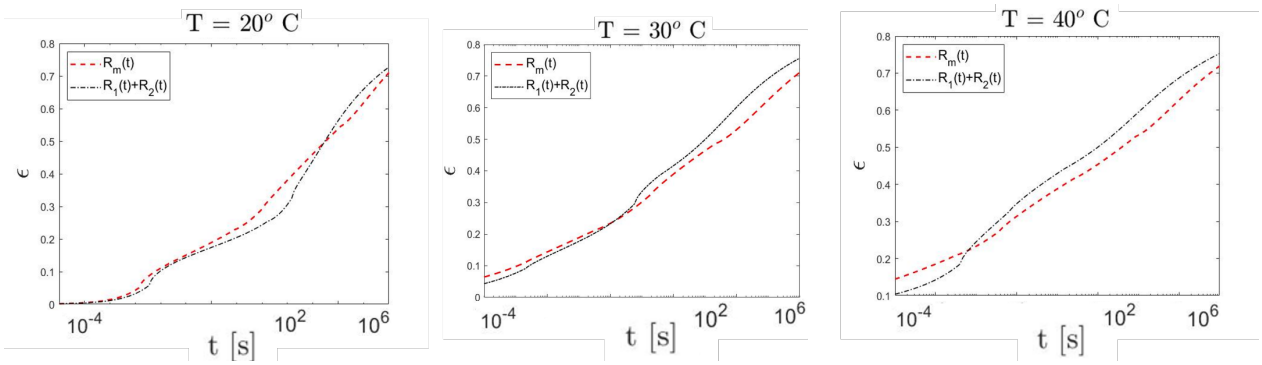

Fig. 6: Deformation $\epsilon(t)$ for the two different cases of Figure 4, for the three considered temperatures 20°C (a), 30°C (b), 40°C (c).

The obtained results confirm that the fractional viscoelastic model produces reliable results. The second step consists in calculating the relaxation curve from the previously obtained strain trends. This is possible by inverting Equation 4 using as an input data the deformation $\epsilon = \epsilon_1 + \epsilon_2$ that describe the mechanical response of the laminated glass structure with the MMPI. The result is shown in Figure 7 at different environmental temperatures 20°C (a), 30°C (b), 40°C (c), and compared with the experimental results of the relaxation curve of the same MMPI obtained through DTMA analysis. We conclude that the FEFV model can accurately predict the long-term response of the composite material given the known relaxation function of the two separates materials and their thickness ratio. The simulated relaxation function well fits with the experimental points represented with the red triangles.

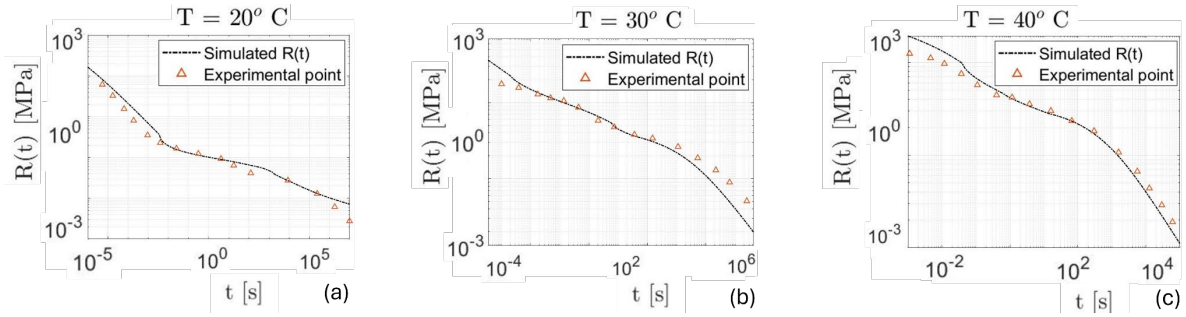

Fig. 7: Simulated relaxation function of the MMPI obtained through the FEFV model, represented with black dashed line, compared with the red triangles representing the experimental points of the relaxation functions of the same interlayer obtained though DTMA, at different temperatures 20°C (a), 30°C (b), 40°C (c). The polymeric materials used are Stiff PVB and Acoustic PVB.

The proposed model is based on a full viscoelastic analysis of the polymeric material. In order to emphasize how important it is to consider such a material model in this type of analysis, a comparison with the simple elastic model already proposed in (Schuster, Härth, Thiele & Bennison, 2023) is now developed. Here, the relaxation function is simply calculated as the inverse stiffness at time *t* of the multilayer, which is approximated from the inverse addition of the individual stiffnesses at time *t,* i.e.,

$$\frac{h_1+h_2}{R_m(t)} = \frac{h_1}{R_1(t)} + \frac{h_2}{R_2(t)}. \qquad (6)$$

The result of this comparison is shown in Figure 8. Observe that in the early stages of observation time the two approaches give the same result, because the memory effect of the polymer has not yet a

great influence. Moving to longer observation times, the difference between the two solutions becomes more evident. The elastic model gives reliable results for an observation time of approximately $\sim 10^2$ seconds, but it is not sufficient to describe the long-term response of the MMPI. Instead, the FEFV model is in good accordance with the experimental results for the entire observation time.

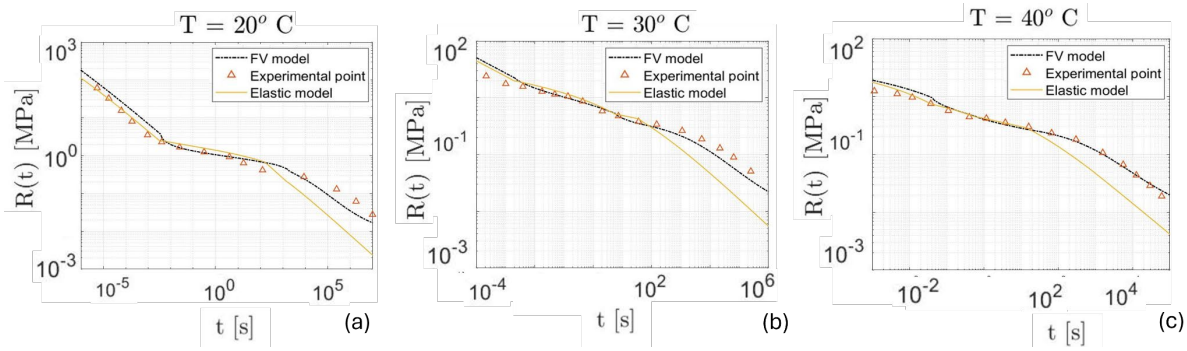

Fig. 8: Simulated relaxation function of the MMPI obtained through the FEFV mode,l represented with black dashed line, and the simulated relaxation function obtained with the elastic model, indicated with yellow line; comparison with the red triangles representing the experimental points of the relaxation functions of the same interlayer obtained via DTMA, at different temperatures 20°C (a), 30°C (b), 40°C (c). The polymeric materials used are Stiff PVB and Acoustic PVB.

Since the model is validated, a final analysis is now proposed to show the potential of this approach in predicting the mechanical behaviour of an MMPI. A comparison has been made between two different types of interlayers, the one previously analysed composed by Stiff PVB and Acoustic PVB, and another one where the Stiff PVB has been replaced by a Sentry Glass sheet with the same thickness. Our aim is to show the influence of this material on the mechanical performance of the global package at different environmental temperatures 20°C (a), 30°C (b), 40°C (c). Figure 9 shows the result of this comparison, also considering for completeness the elastic solution previously discussed. In the first row (Figures 9 a, b, c) we compare the two solutions corresponding to the two different materials, juxtaposed to the experimental points and the elastic solutions only in the first case. It is possible to observe that the sentry Glas does not have a big influence on the mechanical performance of the MMPI at 20° C since the two solutions almost overlap; its influence becomes more evident by increasing the temperature, when the relaxation function of Stiff PVB drops out and stiffness is lost (Figure 2). On the contrary, the response of Sentry Glas tends to be more stable at higher temperatures, as it is clear from the graphs of the second row (Figures 9 d, e, f). Here, the comparison between the FEFV and the elastic solution in the second case (MMPI is composed by Sentry Glas and Acoustic PVB) is also shown, . Now the elastic solution is not able to correctly predict the relaxation function of the composite and badly fails for long observation times.

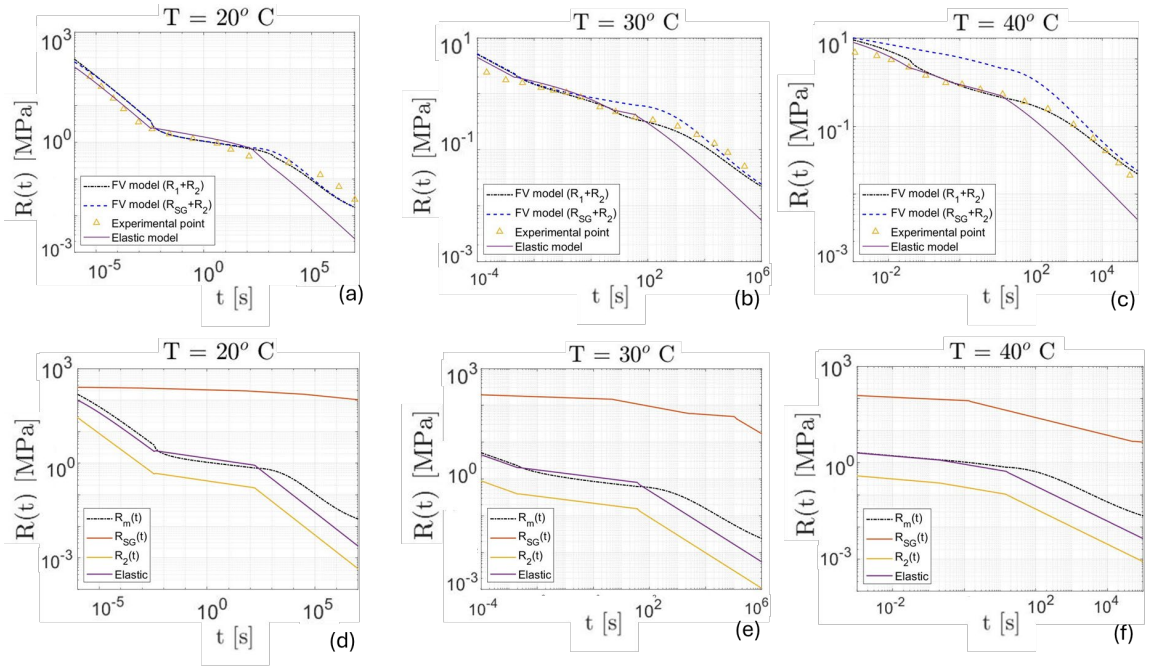

Fig. 9: Simulated relaxation function of the MMPI obtained through the FEFV model, represented with black and blue dashed lines, and the simulated relaxation function obtained with the elastic model, represented with purple line; comparison with the yellow triangles, representing the experimental points of the relaxation functions of the same interlayer obtained though DTMA, at different temperatures 20°C (a), 30°C (b), 40°C (c). Comparison between two different cases: when the MMPI is composed by R_1= Stiff PVB, R_2= Acoustic PVB and R_1= Sentry Glas, R_2= Acoustic PVB.

4. Conclusions

A novel approach to characterize the mechanical performance of a MMPI, able to predict the corresponding relaxation function, has been proposed. The approach is based on an innovative method to interpolate the experimental relaxation curve from DTMA analysis, which uses piecewise branches of power laws. Using this method, it is possible to write the constitutive equation of the polymeric material through fractional derivatives to interpret their viscoelastic response. In particular, the stress is proportional to a real-order derivative, intermediate between 0 and 1 of the strain over time. This approach considers a full viscoelastic characterization of the material that it is crucial for this kind of analysis. The model can be solved through a finite element numerical analysis, that we named FEFV model. The predicted relaxation function is in very good agreement with the experimental points. On the other hand, if an elastic model is considered, results are not reliable. This approach can also be used to predict the relaxation function of any kind of MMPI that combines two or more different polymers with different thickness; it permits to readily evaluate the mechanical performance without having to perform expensive and time-consuming experimental campaigns. This model can be very helpful in the design of new types of MMPI that can combine different properties on the laminated glass structure, including better acoustic performance. Having such powerful tool can helps customers to find the best solution for a MMPI for their specific application. by furnishing a reliable and easily interpretable solutions.

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