Viscoelastic properties of EVA interlayer used in laminated glass structures

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Abstract. Laminated glass is getting used more extensively in a current architecture due to its transparency, aesthetic impression, and post-breakage behaviour. Examples of such structures are balustrades, stairs, or facade panels. These are usually loaded in bending. Polymeric interlayer embedded between glass plies has no flexural stiffness, but it can itself transfer shear stress. The rate of this transfer depends on the shear stiffness of interlayer which is time and temperature dependent parameter. Producers of interlayers often do not specify this quantity thus engineers rather neglect the shear interaction of glass plies in perpendicularly loaded laminated glass panels. This paper provides the values of shear stiffness modulus of common interlayer EVASA\(\text{F}^{\text{E}}\) by Bridgestone\(^{\text{T}}\) in time and temperature domain. This shear stiffness is expressed through Maxwell model whose parameters are based on the series of Dynamic mechanical thermal analysis (DMTA) performed on single lap small-scale specimens in Klokner institute CTU in Prague. Results show that shear stiffness of this interlayer is able to provide significant shear coupling of glass plies in broad temperature range of short-term perpendicularly loaded laminated glass structures.

1. Introduction
Laminated glass is used in a current architecture even as a load bearing element. It is a composition of glass plies bonded with polymeric interlayer. Typical applications of these structures are balustrades, roof panels, stairs, or canopies. The direction of applied load is usually perpendicular to central plane and stress-state and deflections are significantly affected by the shear stiffness of a particular interlayer [1]. This parameter is time and temperature dependent quantity [2] and it is not stated in technical sheets of most of interlayers. Increase of temperature as well as long-term loading induce molecular movement in a polymeric material resulting in reduction of its shear stiffness [2]. Therefore, laminated glass structure loses its bending stiffness and main tensile stress in glass increase. Drop of temperature and short-term loading have opposite effects [3]. To design load bearing laminated glass in bending taking shear interaction of glass plies into account, an engineer is referred to national standards, e.g. CNR-DT 210 [4], or to the current European draft prEN 16612 [5]. The mechanical tests of interlayers can be performed on small-size samples of a single interlayer [6], on small laminated glass specimens [7], or on large specimens [8]. They can be basically sorted into two groups: (i) static long-term creep or relaxation experiments, and (ii) dynamic tests called Dynamic mechanical thermal analysis (DMTA) where the specimen is cyclically loaded in various frequencies and temperatures. Static tests do not require complicated evaluation methods, but they cannot be usually performed in broad time and
temperature range. Contrary DMTA results need to be thoroughly evaluated, but they enable to express material stiffness in long relaxation times and broad temperature range. It is more appropriate to perform the experiments of an interlayer in shear or torsion. From the shear or torsion test, the shear modulus $G$ is determined with more precision in comparison with tensile tests where the conversion using Poisson ratio is necessary [9]. This paper is focused on viscoelastic description of ethylene-vinyl acetate interlayer EVASAFE® made by Bridgestone™ based on series of displacement controlled DMTA experiments performed on small-scale single lap laminated glass specimens. Particularly, Prony series of constructed Maxwell model are provided and relaxation shear modulus $G$ at reference temperature 20 °C in time is illustrated. Response of the constructed model was further compared to the response of EVASAFE® in static single lap displacement-controlled shear tests performed in various loading rates and temperatures using the identical specimens and testing equipment as in DMTA. All experiments and analyses were performed in Klokner Institute CTU.

2. Theory of Viscoelasticity

Response of viscoelastic material subjected to shear cyclic loading in linear viscoelastic region is described by Boltzmann superposition principle [10]

$$\tau(t) = \int_{0}^{t} G(t - s) \dot{\gamma}(s) \, ds,$$

where $\tau(t)$ is the shear stress in time in [MPa], $s$ is the time of the loading step from the interval $<0; t>$ in [s], $\dot{\gamma}(s)$ refers to the engineering shear strain rate in [s$^{-1}$] at the certain time $s$, and $G(t)$ is the relaxation function in [MPa] of a viscoelastic material in the time domain. This function is usually considered in the form of the generalized Maxwell model [11] according to the equation

$$G(t) = G_\infty + \sum_{i=1}^{n} G_i \exp(-t/\tau_i),$$

where $G_\infty$ is the equilibrium shear stiffness modulus in [MPa] in long relaxation times, $G_i$ denotes the shear stiffness of spring in [MPa] together with the relaxation time $\tau_i$ in [s] of the $i$-th Maxwell element, and $n$ is the number of Maxwell elements in the Maxwell model. Relaxation time $\tau_i$ expresses the ratio $\eta_i/G_i$, where $\eta_i$ [Pa.s] is the viscosity of the dashpot of $i$-th Maxwell element. The structure of Maxwell model is shown in figure 1.

![Maxwell model](image)

Figure 1. Maxwell model with its Prony parameters.

To include the temperature effect into the Maxwell model, the relaxation time $\tau_i(T_{\text{ref}})$ of $i$-th element determined for the reference temperature is modified as $\tau_i(T) = \alpha_T(T) \cdot \tau_i(T_{\text{ref}})$ for thermorheologically simple material, where $\alpha_T$ denotes the shift factor for temperature $T$ [12]. This quantity may be obtained from the following Williams-Landel-Ferry (WLF) equation

$$\log \alpha_T = -C_1(T - T_{\text{ref}})/(C_2 + T - T_{\text{ref}}),$$
where constants $C_1$ and $C_2$ are free non-dimensional WLF parameters [13]. WLF equation provides reliable results of extrapolated relaxation times for temperatures above Glass transition temperature $T_g$. For lower temperatures, the horizontal shift factor $\alpha_T$ should be expressed using different formulas [12].

In DMTA the interlayer is loaded by cyclic deformation according to the equation (4), where $\gamma(t)$ denotes shear strain in time $t$ [s], $\gamma_0$ is the shear strain amplitude [-], and $\omega$ denotes the angular frequency [rad/s]. If shear strain $\gamma(t)$ is implemented into Boltzmann superposition principle in equation (1), its integration leads to the shear stress $\tau(t)$ through the complex modulus $G^*$ [MPa] with its real part $G'$ (storage modulus) and imaginary part $G''$ (loss modulus) [10] as shown in equation (5).

$$\gamma(t) = \gamma_0 \cdot \sin \omega t$$  \hspace{1cm} (4)

$$\tau(t) = G^* \cdot \gamma(t) = \gamma_0 \cdot (G' \cdot \sin \omega t + G'' \cdot \cos \omega t)$$  \hspace{1cm} (5)

General viscoelastic loop of stress-strain dependence with its important points as well as stress and strain courses in time observed during DMTA are displayed in figure 2. Viscoelastic materials during cycling exhibit time delayed response to the applied strain characterized by their phase shift $\delta$ [rad] as shown in figure 2.

DMTA is usually performed at a certain range of angular frequencies and temperatures. Several curves of complex modulus $G^*$ are obtained in that range depending on a testing equipment, see the example in figure 3. For thermorheologically simple materials, horizontal shifts of measured relations for the value $\log \alpha_T(T)$ are performed to get a smooth Master curve at reference temperature $T_{ref}$, which enables to express the complex modulus $G^*$ (resp. shear stiffness of material $G$) in wide frequency (resp. time) domain. This procedure is called Time-Temperature-Superposition-Principle (TTSP) as a sufficient approximation for engineering purposes [14]. As soon as Master curve $G^*(\log \omega)$ for a chosen reference temperature is completed, it is possible to approximate this curve by the response of the Maxwell model loaded in harmonic oscillatory loading by angular frequency $\omega$ according to the equation (4). Shear response $\tau(t)$ of that model is then expressed from the equation (5) by the equation (6) [11], where $n$ denotes the number of Maxwell elements. Relaxation curve of fitted model is then expressed by the equation (2).

$$\tau(t) = \gamma_0 \cdot G_\infty \cdot \sin \omega t + \sum_{i=0}^{n} \frac{G_i \cdot \omega^2 \cdot \pi_i^2 \cdot \sin \omega t}{1 + \omega^2 \cdot \pi_i^2} + \sum_{i=0}^{n} \frac{G_i \cdot \omega \cdot \pi_i \cdot \cos \omega t}{1 + \omega^2 \cdot \pi_i^2}$$  \hspace{1cm} (6)
3. Test Set-up

To find the viscoelastic parameters, DMTA tests of six single lap small glass specimens EVASAFE® in the range of temperatures -5 °C to +50 °C and testing frequencies between 0.05 Hz and 4.95 Hz were completed in Klokner Institute CTU in Prague. Experiments were performed in hydraulic testing system MTS with climatic chamber TIRA TEST T250/1. Shear strain of interlayer γ was controlled according to the equation (4). The shear area A of the interlayer was 50 x 50 mm with its thickness 0.81 mm. Engineering shear strain γ of the interlayer was determined from its thickness h and mutual displacement of glass panes u measured by displacement sensors MMR 1011 as γ = u/h. Shear stress τ acting on the interlayer was calculated from the applied force F and shear area A as τ = F/A. Temperature in a climatic chamber was controlled by sensors Pt100 glued directly on the sample surface. As it has been stated, static displacement-controlled tests of the identical interlayer had been performed using identical testing equipment, specimens, and MTS system. Stress-strain relations obtained from those tests will serve for the evaluation of EVASAFE® Maxwell model. Detailed description of performed DMTA tests and static tests including methods of results evaluation has been specified by Hána et al. [15].

4. Results and Discussions

DMTA results, particularly storage modulus-frequency relations of representative specimen are shown in figure 6. Loss modulus was much lower than storage modulus (G" ≪ G'), thus it was not considered in master curving process. In the sense of regression analysis, WLF parameters C₁ and C₂ and the appropriate master curve at reference temperature 20 °C were constructed, see figure 7.
Figure 6. Measured storage modulus $G'$ of representative specimen EVASAFE at testing temperatures.

Figure 7. Constructed Master curve at reference temperature 20 °C for representative specimen.

Master curves of another testing specimens are of similar course. Glass transition temperature of ethylene-vinyl acetate is below 0 °C [16] thus master curving process using WLF equation is justified [12]. It is visible that a clear Master curve emerges in the range of high stiffness, while at the range of low stiffness it shows a discontinuity. It reflects the fact that EVASAFE® shows signs of thermorheological complexity and vertical displacements and rotations of the experimental curves are also necessary to get a clear Master curve. Similar discontinuity of Master curve in low frequencies of another EVA-based cross-linked interlayer evguard® has been revealed by Schuster et al. [16]. Based on the WLF constants and Master curve, generalized Maxwell model was constructed. To reduce the number of unknown Prony parameters, relaxation times $\tau_i$ were chosen. Homogenous distribution of relaxation times in frequency (time) domain was adopted [17]. It was chosen to use 22 relaxation times $10^{-10+k}$, where $k = 1:22$, to cover the range of testing frequencies. Fitting the shear moduli $\{G_i\}$, and $G_\infty$ then leads to linearized problem. The objective function to be minimized is given by the equation (7)
\[ F(\{G_i\}, G_\infty) = \sum_{j=1}^{m} (G'(o_j) - G_j')^2 + \sum_{j=1}^{m} (G''(o_j) - G_j'')^2, \]  

where \( m \) is the number of all experimental measurements, \( G_j' \) and \( G_j'' \) are the measured storage and loss moduli for frequency \( o_j \), and \( G'(o_j) \) with \( G''(o_j) \) are the storage and loss moduli given by Maxwell model for frequency \( o_j \). Minimizing condition of this function based on calculus takes the form

\[ \frac{\partial F}{\partial G_\infty} = 0; \frac{\partial F}{\partial G_i} = 0; i = 1, 2, ..., n. \]  

After solving the set of equations (8), the set of \( \{G_i\} \), and \( G_\infty \) with the corresponding \( \tau_i \) was obtained, see Table 1. When these are used as input parameters in the equation (2), the shear modulus \( G(t, T = 20 \, ^\circ C) \) in time domain is obtained, see Figure 8. To express the shear modulus in other tested temperatures, relaxation times according to TTSP can be modified as \( \tau_i(T) = \alpha_T(T) \cdot \tau_i(T_{\text{ref}}) \).  

**Table 1.** Prony parameters of investigated interlayer EVASAFE®.

| \( \tau_i \) (s) | \( G_i \) (MPa) | \( G_\infty = 0.362 \, \text{MPa} \) | \( \tau_i \) (s) | \( G_i \) (MPa) | \( \tau_i \) (s) | \( G_i \) (MPa) |
|----------------|----------------|-----------------|----------------|----------------|----------------|----------------|
| 1.00E-09       | 5.899          | 1.00E-02        | 0.365          | 1.00E+06       | 0.159          |
| 1.00E-08       | 2.882          | 1.00E-01        | 1.670          | 1.00E+07       | 0.194          |
| 1.00E-07       | 0.789          | 1.00E+00        | 0.063          | 1.00E+08       | 0.220          |
| 1.00E-06       | 0.136          | 1.00E+01        | 0.284          | 1.00E+09       | 0.060          |
| 1.00E-05       | 1.412          | 1.00E+02        | 1.322          | 1.00E+10       | 0.985          |
| 1.00E-04       | 17.767         | 1.00E+03        | 0.653          | 1.00E+11       | 0.141          |
| 1.00E-03       | 0.071          | 1.00E+04        | 1.260          | 1.00E+12       | 0.396          |

**Figure 8.** Relaxation function for the reference temperature 20 °C.
Short-term shear modulus at 0 °C – \( G(1s, 0 \, ^\circ C) = 18.2 \, MPa \) ensures monolithic response of laminated panel in bending. According to Galuppi and Royer-Carfagni [18] for the shear modulus values in range \( G = 1 – 10 \, MPa \), the assumption of significant shear interaction of the individual glass plies in laminated panel is justified. Short-term shear moduli of tested interlayer, \( G(1s, 20 \, ^\circ C) = 7.0 \, MPa; G(1s, 40 \, ^\circ C) = 3.9 \, MPa; G(1s, 60 \, ^\circ C) = 2.1 \, MPa \) fulfill this assumption. Contrary, shear interaction of glass plies in long-term loaded laminated glass panel \( (G_c = 0.36 \, MPa) \) would be limited [18].

To verify the presented model, its shear stress response to the constant shear strain rate in tested temperatures was calculated and compared to stress-strain relations of already mentioned static displacement-controlled shear tests. In static tests, thickness of the interlayer was also 0.81 mm, temperatures were kept as 0; 20; 40; 60 °C and loading strain rates of the interlayer \( \dot{\gamma} \) based on set MTS crosshead displacement were calculated as 0.0407 s\(^{-1} \) (for displacement 2.0 mm/min), 0.0102 s\(^{-1} \) (for displacement 0.5 mm/min), and 0.0025 s\(^{-1} \) (for displacement 0.125 mm/min). Representative results from static tests are in figure 9. If relaxation function of the constructed Maxwell model from the equation (2) is given into the integral equation (1), its evaluation results in the equation (9) which reflects the evolution of shear stress in constant strain rate. This way enabled to obtain shear stress-strain relations for tested temperatures in figure 10. When comparing both figures, one can see that static tests provide similar relations as Maxwell model in the range of engineering shear strains to 20%. This value is far over that observed in intact laminated glass panel (normally up to 1% [11]). For higher strains, Maxwell model is stiffer which might be a cause of exceeding the limits of linear viscoelasticity (usually between 20% and 50% [12] for softer polymeric material) or lower real shear strain rate of the interlayer.

\[
\tau(t, T) = \dot{\gamma} \cdot \left[G_{\infty} \cdot t + \alpha_T(T) \cdot \sum_{i=1}^{n} G_i \cdot \tau_i \cdot [1 - \exp(-t / (\alpha_T(T) \cdot \tau_i))]\right]
\] (9)

![Figure 9. Stress-strain relations from static shear tests in various strain rates.](image1)

![Figure 10. Response of constructed Maxwell model to the applied various strain rates.](image2)

5. Conclusion
This paper provided Prony parameters of the constructed WLF and Maxwell model of polymeric interlayer EVASAFE® based on the series of DMTA shear tests performed on small single lap
laminated glass specimens at CTU in Prague. Experiments showed that tested interlayer exhibits signs of thermo-rheologically complex behaviour. Short-term relaxation modulus $G$ based on constructed model ensures significant shear interaction of glass plies in a laminated panel in the range of 0 – 60 °C. Prony parameters were further used to calculate the theoretical stress-strain relations of the constructed Maxwell model loaded by various constant shear strain rates at various temperatures. These strain rates were identical with those from static displacement-controlled shear tests calculated on the set value of MTS crosshead displacement (2.0 mm/min, 0.5 mm/min, and 0.125 mm/min). Static shear tests were performed on the same testing specimens and interlayer as DMTA. Presented model provided similar stress-strain relations with static shear tests for engineering strains to 20% at all tested temperatures. For higher strains, response of Maxwell model to the applied load was stiffer. To validate the presented Maxwell model, sets of relaxation experiments of laminated glass with EVASAFE® at various temperatures are necessary. Experimentally verified models of polymeric interlayers is the way how to design safe and economic glass structures.

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