Dynamic Mechanical Thermal Analysis of EVA and PVB Polymeric Interlayers in Low Temperatures

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Abstract. Many examples of glass load bearing structures such as beams, panes, balustrades, columns or even stairs can be found in a current architecture. These members are usually made of laminated glass panels. Glass plies of a laminated panel are bonded with polymeric interlayer significantly influencing shear forces transfer between them. It principally depends on a polymer shear stiffness which is affected by an ambient temperature and load duration. There is still general lack of knowledge regarding shear stiffness of most polymeric interlayers. Civil engineers thus design laminated glass members with excessive caution neglecting positive shear coupling of the glass plies provided by the interlayer. This approach leads to uneconomical and over-sized glass bearing structures profoundly preventing an extensive use of laminated glass in civil engineering. There are many polymer interlayers of different chemical composition available on the market. Mechanical properties of most of them are unfortunately not available for civil engineers dealing with laminated glass constructions design. This paper is focused on dynamic shear modulus of EVA (ethylene-vinyl acetate) and PVB (polyvinylbutyral) interlayer and their experimental investigation as a function of temperature and loading ratio. One possible way of determining this modulus is a shear dynamical thermal analysis (DMTA) which further enables to derive time and temperature dependent shear stiffness of EVA and PVB. This experimentally investigated property helps engineers design safer and cheaper glass constructions, possibly extending the use of laminated glass in a current architecture.

1. Introduction
Laminated glass which is made of two or more glass plies bonded with a polymeric interlayer is the subject of current research [1]. Process of bonding is usually performed in autoclave machine under the pressure 0.8 MPa and temperature 140 °C. Laminated glass as a load bearing structural element becomes necessary to be used because of its residual load bearing capacity in the accidental situation. In case of one glass ply breakage, the remaining glass plies are still able to carry some part of the load and thus enable users to leave an endangered area. Furthermore, glass shards remain attached to the interlayer and do not fall down, possibly causing harmful injuries. Laminated glass can be therefore installed above the heads of structure users. Calculation and design of laminated glass structural members is, apart from monolithic ones, much more complicated since transfer of shear forces provided by the interlayer is not a well-known fact. Civil engineers mostly design laminated safety glass on the safe side neglecting positive shear coupling of glass plies provided by the interlayer especially due the lack of its properties. This approach results into expensive and over-sized laminated glass members. Many various polymeric
interlayers are these days available on the market. From a global point of view, they differ in a chemical composition profoundly affecting their shear stiffness which plays a crucial role in stress distribution of laminated glass panels. The interlayer shear stiffness is time and temperature dependent since polymeric interlayers are in fact viscoelastic materials [3] thus these external effects influence the overall performance of laminated glass panels. This makes analysis of these structures rather complicated. This paper introduces the experimental data obtained from Dynamic mechanical thermal analysis of two types of EVA interlayers and one type of PVB interlayer in low temperatures performed at Klokner Institute CTU in Prague, describes how the experiments were done and gives the evidence of interlayers viscoelastic behaviour.

2. Experimental program

2.1. Materials and equipment
Two types of EVA interlayers such as Evalam and Evasafe as well as one type of PVB interlayer such as Trosifol BG-R-20 were experimentally examined. Evalam interlayer will be stated as EVA L, Evasafe interlayer will be stated as EVA S and Trosifol BG-R-20 will be stated as Trosifol hereafter in the text. Test specimens were made of 2 annealed float glass panes bonded in autoclave with the examined interlayers at shear area 50 x 50 mm. Dimensions of specimens are shown in figure 1. Thickness of each interlayer was measured at eight representative specimens taking three points of shear area under the microscope. The average thickness values of tested interlayers were determined as follows: Trosifol thickness 1.49 mm, EVA L thickness 0.643 mm and EVA S thickness 0.827 mm. Two potentiometric linear transducers MMR 1011 were stuck on the sides of the specimen to be able to measure the relative slippage of the glass plies \( u \) in [mm] in the acting force direction, see figure 2a. Temperature in the insulating chamber was measured by temperature sensor Pt 1000. To measure the glass surface temperature directly, two Pt 100 sensors were stuck on the sides of the specimen, see figure 2b. In total, there were three specimens with EVA L, three specimens with EVA S and three specimens with Trosifol interlayer dynamically loaded. Low temperatures were ensured by the presence of liquid nitrogen supply into the insulating chamber which was stored in Dewar’s vessel, see figure 3.

![Figure 1. Nominal dimensions of the testing specimen and acting force direction](image-url)
2.2. Test set-up

For DMTA tests, the specimens were put into the metal jaws of the loading device MTS 500B with Tira Test TS 201 chamber as shown in figure 2b and dynamically loaded. Dynamic in this context has no connection with inertial terms or resonance [4]. The tests were controlled with displacement of the glass panes. The amplitude of MTS loading cylinder displacement was set as 0.18-0.2 mm depending on the tested interlayer stiffness for each loading cycle. This value evoked the maximum values of tensile force 1.8-2.5 kN measured by MTS force meter. Temperature and loading frequencies were changed during the tests. The range of testing frequencies $f$ was set as 0.05 – 4.95 Hz for every tested interlayer. Frequency step between each loading cycle was set as 0.05 Hz, therefore each specimen was loaded in 99 different testing frequencies (99 loading cycles). Prestressing tensile force between each loading cycle was set as 1.2 – 1.5 kN depending on the testing specimen with its duration of 10 s. The interlayers were tested in the temperature range +20 °C to -10 °C with a temperature step of 5 °C. The temperature
was held constant during each testing frequency range (99 cycles). Cylinder’s displacement evoked interlayer’s shear strain $\gamma$. To make it simple, the example of this shear strain as a function of time is drawn in figure 4.

Figure 4. Shear strain example of the interlayer during DMTA

2.3. Evaluating method

Force transferred between the glass plies caused a shear strain of the interlayer, see figure 5. This strain was calculated from the glass plies slippage $u$ and the interlayer thickness $t$ as shown in equation 1 referring to $\gamma$ as an engineering shear strain expression. Based on measured values of glass plies slippage, large deflections were not considered. Additional bending moment resulting from the geometry was neglected due to its low effect.

$$\gamma = \tan \gamma = \frac{u}{t}$$

Figure 5. Engineering shear strain $\gamma$ of the interlayer and its calculation formula

In the dynamic mechanical thermal analysis tests (DMTA), the shear strain of the interlayer was in each loading cycle controlled according to the equation 2

$$\gamma = \gamma_{\text{max}} \sin(\omega t),$$

where $\gamma_{\text{max}}$ is the maximum shear strain of the interlayer evoked by 0.18-0.2 mm loading cylinder’s displacement, $\omega$ is the loading angular frequency [rad/s] and $t$ is the instantaneous time [s] in each cycle. The appropriate shear stress can be then calculated from the equation 3 [4].

$$\tau = \tau_{\text{max}} \sin(\omega t + \delta),$$

where $\tau_{\text{max}}$ is the maximum shear stress of the interlayer evoked by 0.18-0.2 mm loading cylinder’s displacement and $\delta$ is the phase-lag between the oscillating strain input and stress response. Each loading cycle, particularly shear stress and strain dependence can be graphically displayed through the hysteresis loop which is drawn in figure 6.
Figure 6. Hysteresis loop showing shear stress-strain dependence in each loading cycle with its important points [4]

The reader can notice that there are some important points marked on the hysteresis loop enabling to find out the required time and temperature dependent shear modulus $G(t,T)$. Loop symmetry axis slope indicates the value of dynamic complex shear modulus $G^*(\omega)$. The slope of the line passing through the origin of the coordinate system and the point at which the maximum shear deformation is achieved indicates the value of the shear storage modulus $G'(\omega)$ being directly proportional to the average energy storage in each loading cycle. Shear loss modulus value $G''(\omega)$ can be determined as the intersection of the hysteresis loop with vertical shear stress axis. This value is directly proportional to the average dissipation or loss of energy as heat in each loading cycle. Mathematical expression of the aforementioned modules can be described using the following equations. For linear viscoelastic materials in sinusoidal shear strain input $\gamma$ according to equation 2, Boltzmann superposition principle expressing the overall response to the applied load as the sum of the individual loads increments responses can be used [4]. Shear strain rate can be written according to the equation 4

$$\gamma' = \frac{d\gamma}{dt} = \omega \gamma_{max} \cos \omega t. \quad \text{(4)}$$

Then the shear stress response can be expressed using equation 5

$$\tau(t) = \int_0^\infty G(t - t')\gamma' dt' = \int_0^\infty G(t - t')\omega \gamma_{max} \cos \omega t \cos \delta dt'. \quad \text{(5)}$$

When the equation 5 is further modified, equation 6 is given in the form

$$\tau(t) = G' \gamma_{max} \sin \omega t + G'' \gamma_{max} \cos \omega t = G^*(\omega) \gamma(t) = \tau_{max} \sin(\omega t + \delta), \quad \text{(6)}$$

where $\gamma_{max}$ is the maximum shear strain input, $\omega$ is the loading angular frequency, $i$ is the complex unit, $t$ is the instantaneous time, $t'$ is the time of loading increment on the time axis, $\delta$ is the phase angle between stress and strain input and $G^*(\omega)$ is the dynamic complex shear modulus with its real and imaginary parts $G'(\omega); G''(\omega)$. These modules can be also expressed as loading cycle frequency $f$.
dependent keeping the relation $\omega = 2\pi f$ in mind. Equation 5 enables to derive these modules from the hysteresis loop. When $\omega t = \pi/2$, then $\gamma_{\text{max}}$ is achieved and then $\tau = G'\gamma_{\text{max}}$. Contrary, for time $t = 0$ is $\gamma = 0$, therefore $\tau(0) = \gamma_{\text{max}}G''$ which is displayed on the hysteresis loop. All three aforementioned modules were evaluated for every loading cycle from the hysteresis loop using software MATLAB®, according to this procedure. Even though DMTA analysis is rather time consuming, it delivers a wide range of dynamic shear modulus values but still in a frequency domain. But the interlayer shear modulus varies with the applied load duration, thus conversion to real time domain is required. This can be performed by Fourier transform for linear viscoelastic materials [5]. This conversion is a part of the next survey. If an accurate conversion is not available, the following approximations with the increasing accuracy enable a quick and simple way to get the shear modulus estimation [6]

\[
G(t = 1/f) = G'(f),
\]

\[
G(t = 1/f) = G'(f) - 0.4G'(f/2),
\]

\[
G(t = 1/f) = G'(f) - 0.4G''(0.4f) + 0.014G''(10f).
\]

3. Results

The following text concludes the stiffness-frequency curves of the representative specimens obtained by DMTA, compares their shear storage modules as a function of frequency and low temperatures, and shows failure mode of specimens.

3.1. Experimental curves

As mentioned previously, there were three specimens of each interlayer in the aforementioned temperatures and frequency range tested with a frequency step of 0.05 Hz between the individual cycles. Each cycle was controlled by the loading cylinder displacement. Figure 7 shows storage modulus-frequency curves of EVA L interlayer. It becomes noticeable that the interlayer’s stiffness increases with the decreasing temperature. The most significant strengthening comes between -5 °C and -10 °C. In task of frequency dependence, there is no profound shear storage modulus growth for temperatures to 0 °C. For the lower ones, this modulus obviously begins to be frequency sensitive.

![Figure 7. Storage modulus-frequency curves of representative EVA L interlayer](image)
Graph in figure 8 shows storage modulus-frequency curves of EVA S interlayer. These relationships are also strongly temperature dependent. When the temperature goes down, the interlayer’s stiffness increases. The most dramatic storage modulus increase is noteworthy between 0 °C and -5 °C. Regarding frequency, the storage modulus does not attain any significant changes for temperatures 20 °C, 15 °C and 10 °C. For the lower ones, it tends to increase with the increasing frequency which is another aspect of strengthening. When comparing frequency-stiffness relations of EVA L and EVA S, these are in general of a similar shape but relations of EVA S are shifted upwards attaining higher storage modulus values. EVA S frequency-stiffness curves also exhibit a more pronounced strain rate input dependence in a broader temperature range when compared to EVA L relations.

![Figure 8. Storage modulus-frequency curves of representative EVA S interlayer](image1)

Storage modulus-frequency curves of Trosifol are displayed in figure 9. When the temperature is decreased, shear storage modulus increases dramatically. The most recognizable stiffness growth can be detected between 10 °C and 5 °C. Measurement in lower temperatures could not be performed because of specimen’s breakage. Shear storage modulus is further frequency sensitive. When the load is applied faster, the stiffness tends to increase in all tested temperatures. Apart from EVA interlayers, Trosifol’s relationships are shifted far upwards. This confirms its stiffer response.

![Figure 9. Storage modulus-frequency curves of representative Trosifol interlayer](image2)
3.2. Numerical values
To compare the experimental data in detail, certain values of shear storage modulus are shown in table 1 as a function of temperature and loading period of each cycle. The loading cycle period is reciprocal to the loading frequency $f$. Table 1 shows numerical values of shear storage moduli of tested interlayers for representative periods of loading cycles. When the temperature decreases, the shear storage modulus achieves higher values and when the loading cycle takes a longer time, the shear modulus decreases in all cases of interlayers. Keeping the direct proportionality between shear storage modulus $G'$ and real shear modulus value in a time domain $G(t)$ in mind, this performance correlates with viscoelastic material response. Molecular movement and rearrangement of viscoelastic materials are thermally activated processes and temperature increase causes polymer’s relaxation time reduction and its more compliant response [6]. When comparing numerical values of storage moduli of EVA interlayers, one may notice that EVA S storage moduli are about two times higher than EVA L moduli. It results from the fact that EVA S stiffness-frequency curves are shifted upwards to higher storage modulus values when comparing figure 7 and figure 8. Shear storage modules of Trosifol are much higher than EVA interlayers and their differences increase with decreasing temperature. But, apart from EVA, Trosifol’s storage moduli depend on time period of loading cycle more considerably.

Table 1. Experimental values of shear storage moduli for representative EVA L, EVA S and Trosifol

| Temperature | EVA L | EVA S | Trosifol |
|-------------|-------|-------|----------|
|             | $G'$  | $G'$  | $G'$    |
| f [Hz]      | [MPa] | [MPa] | [MPa]   |
| Cycle period (1/f) [s] |       |       |         |
| 1           | 4.0   | 4.6   | 5.7     |
| 0.2         | 3.7   | 4.3   | 5.2     |
| 0.05        | 3.6   | 4.1   | 5.0     |
|              | 9.8   | 11.0  | 12.6    |
|              | 9.4   | 10.5  | 11.9    |
|              | 9.2   | 10.1  | 11.5    |
|              | 33.0  | 80.2  | 142.9   |
|              | 17.0  | 53.3  | 115.9   |
|              | 9.3   | 37.9  | 90.9    |
| 20 °C        |       |       |         |
| 15 °C        |       |       |         |
| 10 °C        |       |       |         |
| 5 °C         |       |       |         |
| 0 °C         |       |       |         |
| -5 °C        |       |       |         |
| -10 °C       |       |       |         |

3.3. Failure mode
When the samples with EVA interlayers were dynamically tested, their stiffness enabled to perform the tests in negative temperatures with no specimen breakage whereas the samples with Trosifol could only be tested up to the temperature of 5 °C. When trying to perform the test in lower temperatures, the interlayer responded in a very stiff manner therefore higher tensile force than 2.5 kN was induced in MTS jaws resulting into glass specimen breakage. This is shown in figure 10. The collapse occurred suddenly with no previous warning.

Figure 10. Sample failure loaded at 0 °C
4. Conclusions
In this paper, experimental results concerning Dynamic mechanical thermal analysis (DMTA) of EVA and PVB interlayers in low temperatures were discussed in detail. Evalam, Evasafe and Trosifol interlayers were tested in the temperature range 20 °C to -10 °C and frequency range 0.05-4.95 Hz. Shear storage-frequency values of these interlayers were introduced and compared. Tested interlayers are generally classified as viscoelastic materials which was confirmed by DMTA analysis. When the ambient temperature was decreased, all polymers’ response was stiffer. When the load was applied faster which meant frequency input increased, all polymers stiffened accordingly. This was also demonstrated through the numerical values of shear storage moduli for representative frequencies. Trosifol interlayer achieved much higher storage moduli in comparison with both EVA interlayers and exhibited a far stiffer shear response to the applied load. This stiffer response evoked glass overloading in tension, which resulted into specimen’s failure thus Trosifol specimens could not be tested in negative temperatures. Even though the interlayer dynamic moduli become known, their conversion to a real time domain is necessary to get their shear relaxation moduli expressing the applied static load duration. Obtained shear moduli as a function of time can be further verified by static creep or relaxation tests. In practice, it is necessary to check out the type of the interlayer used in laminated glass panels perpendicularly loaded. This paper showed that different chemical composition results in a different polymer stiffness which is a serious factor in tensile stress distribution of laminated glass panels. Generally speaking, stiffer interlayers should be used mainly in case of load bearing structural elements such as floors, roof panels, staircases and balustrades. On the other hand, softer ones may be used in short-term loaded structures such as facades or windows.

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