Tensile test on interlayer materials for laminated glass under diverse ageing conditions and strain rates

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Abstract

Laminated glass is obtained by bonding two or more glass layers with a polymeric interlayer. The coupling between glass layers depends on the shear stiffness of the interlayer. The mechanical and optical properties of the interlayer may be affected by weathering factors. Since interlayer materials are viscoelastic, the strain rate may also affect its stiffness and ultimate strength. In this paper, tensile tests are conducted on seven different polymeric films (PVB BG-R20, PVB DG-41, PVB ES, SentryGlas, EVASAFE, EVALAM 80, and TPU) at three different strain rates. The mechanical and optical properties of unaged specimens are compared with specimens exposed to thermal cycles, high temperatures, and moisture. The unaged specimens of PVB DG-41, PVB ES, and SentryGlas had the highest stiffness, EVALAM 80 and EVASAFE had the highest ductility, PVB and SentryGlas had the highest tensile strength, and EVALAM 80, EVASAFE, and TPU were less affected by ageing factors and strain rate.

Keywords
Laminated glass; Polymeric interlayer; Glass transition temperature; Tensile test; Ageing test

1. Introduction

Laminated glass is a composite material consisting of two or more glass layers bonded using a polymeric film as interlayer. It was originally used in car windshields, because the polymeric interlayer prevented glass shards from scattering in case of accidental breakage. The first interlayer used was polyvinyl butyral (PVB). The application of laminated glass later expanded to architecture, and new interlayer materials were developed to increase some laminated glass properties such as the transparency, the flexural stiffness, the post-breakage strength, and the adhesion to other materials, like steel or timber. Nowadays, the most commonly used polymer groups for laminated glass interlayers are polyvinyl butyral (PVB), ethylene-vinyl acetate (EVA), thermoplastic polyurethane (TPU), and ionoplast (e.g. SentryGlas).
Glass is a brittle material, and its fracture is difficult to predict [1], because it is generally caused by the formation and propagation of surface flaws [2]. The mechanical behaviour of glass is linear elastic until breakage. By contrast, the polymeric interlayers are viscoelastic materials, which means that its mechanical response has an elastic and a viscous component: the elastic response is proportional to the strain, whereas the viscous component is proportional to the strain rate. The mechanical response of viscoelastic materials is time- and temperature-dependant [3]. Dynamic mechanical analysis (DMA) allows doing a characterization of the thermo-viscoelastic properties of polymers. Andreozzi et al. [4] performed dynamic torsion tests on laminated glass specimens, and Pelayo et al. [3] performed dynamic tensile tests on polymeric interlayer films. Callewaert et al. [5] evaluated the influence of load duration and temperature on laminated glass plates under flexural load. Some interlayer manufacturers provide the value of the Young modulus (elastic component) as a function of the load duration time and working temperature (Figure 1).

Figure 1. Young modulus of three commercial interlayer materials as a function of load duration time and service temperature, obtained from the Kuraray catalogue [6].
The shear stiffness of the interlayer material, as well as the level of adhesion with glass, can affect the pre- and post-breakage performance of laminated glass plates [7]. Laminated glass panels under out-of-plane bending have a higher bending stiffness and maximum load when they have stiffer interlayers [8-10]. Sable et al. [11] and Hána et al. [12] tested and simulated laminated glass plates in a four-point bending test. They identified that the type of interlayer affected not only the bending stiffness and the maximum load, but also the breakage mode and fracture pattern. With regard to the post-breakage performance, Zhao et al. [13] identified that the type of glass, the number of layers, and the mechanical properties of the interlayer material affected the failure mode, as well as the post-breakage bending stiffness and load-bearing capacity of damaged laminated glass plates.

The exposure to weathering factors, such as solar radiation, thermal cycles, or humidity, may affect the mechanical and optical properties of the polymeric interlayers, as well as its adhesion with glass [4] or other substrates such as steel [14]. Ageing factors and their effect on strength and durability are especially looked upon adhesive and laminated connections between elements [15]. Chiara Bedon [16] identified that degradation in the bonding region between glass layers could severely affect the structural performance of laminated glass.

In this paper, the authors study the effect of strain rate and accelerated ageing on the elastic behaviour of polymeric interlayers. For that reason, the specimens were subjected to uniaxial tensile tests until breakage, as previously done by Biolzi et al. [17]. The authors aim to compare the tensile properties of seven interlayer materials, under different strain rates and after exposure to different ageing factors. These results will provide relevant information for the selection of interlayer materials to be used in laminated glass. No previous papers compare the mechanical performance of so many unaged and aged interlayer materials at different strain rates.

2. Experimental study

2.1. Materials

Seven commercial polymeric interlayer materials were characterized in this study. These materials are based on four of the most commonly used polymer groups for laminated glass interlayers: PVB, EVA, TPU, and Ionomer. Table 1 shows the material base polymer, the trademark and the manufacturer of each of the tested interlayers.
Table 1. List of polymeric interlayer materials used: base polymer, trademark and manufacturer.

<table>
<thead>
<tr>
<th>Base polymer</th>
<th>Trademark</th>
<th>Manufacturer</th>
</tr>
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<tbody>
<tr>
<td>Polyvinyl butyral (PVB)</td>
<td>Trosifol® BG-R20</td>
<td>Kuraray Group</td>
</tr>
<tr>
<td></td>
<td>Trofisol® ES</td>
<td>Kuraray Group</td>
</tr>
<tr>
<td></td>
<td>Saflex® DG-41</td>
<td>Eastman Chemical Company</td>
</tr>
<tr>
<td>Ethylene vinyl acetate</td>
<td>EVALAM-80</td>
<td>Pujol Evalam</td>
</tr>
<tr>
<td>(EVA)</td>
<td>EVASAFE</td>
<td>Bridgestone Corporation</td>
</tr>
<tr>
<td>Ionomer</td>
<td>SentryGlas</td>
<td>Kuraray Group</td>
</tr>
<tr>
<td>Thermoplastic polyurethane</td>
<td>KRYSALFLEX® PE399</td>
<td>Huntsman</td>
</tr>
</tbody>
</table>

Three different PVB are tested: PVB BG-R20 is a standard PVB, whereas PVB ES and PVB DG-41 have a smaller amount of plasticiser. The main expected effect of reducing the level of plasticiser is that the glass transition temperature increases, and therefore the stiffness at a same temperature also increases. EVALAM and EVASAFE belong to the same polymer group, but have different mechanical properties because EVALAM has a higher portion of vinyl acetate. With a higher level of vinyl acetate, the behaviour of the copolymer is closer to that of rubber.

2.2. Methods

The glass transition temperature ($T_g$) was measured using the differential scanning calorimetry (DSC) technique. The methodology carried out to analyse interlayer materials is based on the standard ASTM E 1356 [18]. In this standard, a curve is represented in the heat flux-temperature diagram, and a tangent line is drawn at the inflection point in the region of the glass transition. The glass transition temperature is the mid-point between onset and endset of the inflectional tangent (Figure 2). The equipment used was a DSC 822e from Mettler Toledo under a nitrogen gas flow. The amount of sample material used was less than 5 mg and the sample was located into a 40 µl aluminium crucible. The DSC analyses were performed 30 ºC above and 50 ºC below the glass transition temperatures reported in the literature, as standards suggest. In addition, measurements were performed at three cooling rates: 20 ºC/min, 10 ºC/min and 5 ºC/min.
The water absorption test methodology was based on UNE-EN ISO 62 [19] and UNE-EN ISO 175 [20] standards. The main idea of the water immersion test was to simulate a long-term exposure to a humid weather in buildings. Three rectangular specimens (20 mm x 100 mm) of each polymer were tested. Each specimen was placed inside a closed container and filled with deionized water. Afterwards, specimens were placed in a thermostatic water bath and maintained at approximately 23 ºC during 367 hours, which corresponds to the addition of the time intervals of 1, 2, 4, 8, 16, 24, 48, 96, and 168 hours to measure the mass gain recommended by the standard UNE-EN ISO 175 [20]. In order to measure the water absorption, the samples were removed from the water containers and, after removing the excess of water, they were weighted using the analytical scale Mettler Toledo AG135, following the standard UNE-EN ISO 175 [20]. The mass increase is associated to the water absorption. Drying processes were performed in an oven Venticell (Comfort line) from MMM Group at 40 ºC.

Uniaxial tensile tests were performed following the ASTM D638 standard [21]; dumbbell shaped specimens (type IVb) were die-cut (Figure 3). Three samples of each interlayer were analysed to ensure repeatability. The equipment for the fabrication of the specimens, as well as the final result, are shown in Figure 4.
Tests were carried out in a Zwick Roell tensile testing machine (Figure 5) at 24 °C under constant displacement rates of 10 mm/min, 50 mm/min and 100 mm/min. Such velocities are recommended in ISO 527 standard [22]. Results were plotted in the stress-strain diagram, representing stress as the ratio of the applied load to the initial cross section, engineering strain as the ratio of the total deformation to the initial specimen length, and toughness as the area under the stress-strain curve. The secant modulus was the parameter used to quantify the tensile stiffness.
of the tested materials for small deflections. It was calculated by dividing the corresponding stress by the designated strain [21]. The strain interval was between 0 and 0.1 (Figure 6).

![Tensile test set-up.](image)

**Figure 5.** Tensile test set-up.

Furthermore, in order to evaluate the effect of water immersion on the mechanical properties of the interlayer materials, three tensile test specimens of each material were subjected to the same water absorption test. In this case, specimens were dried and tested after 120 h of immersion to ensure the absorption of more than 90% of the saturation water. Tensile tests to evaluate the effect of water content on the mechanical properties were performed under a constant displacement rate of 50 mm/min.

![Sketch of the calculation of the secant modulus.](image)

**Figure 6.** Sketch of the calculation of the secant modulus.

Two different temperature test methodologies were applied on tensile test specimens to evaluate the influence of temperature ageing on their mechanical behaviour. On one hand, an isothermal
temperature program (60 °C) based on UNI EN ISO 12543-4 standard [23] was used during 16 hours. On the other hand, a dynamic temperature program was established for thermal cycling tests, varying the temperature between 30 °C and 50 °C at a constant cooling and heating rate of 3.2 °C/min. A total of 80 cycles were performed following this methodology. Both temperature tests were carried out in the Venticell oven with the air flap lever open. The temperature during the test was 24 °C, as with unaged specimens. Figure 7 shows the equipment and setup of the different ageing tests described in this section.

3. Results

3.1. Glass transition temperature

Below the glass transition temperature, a polymer is more brittle and stiff, whereas over the glass transition temperature it is softer and less viscous [24]. Therefore, the glass transition temperature
of the interlayer, as well as the temperatures at which the laminated glass elements may be exposed during its service life, can affect the out-of-plane resistance [10] and post-breakage behaviour [13] of laminated glass elements. The measured glass transition temperatures of the materials are shown in Table 2. According to the literature, the glass transition of EVA polymers is within the range from -10 ºC to -50 ºC [25]. Moreover, according to its technical data sheet [26], the TPU material KRYS\textsc{tal}FLEX\textregistered{} PE399 presents a midpoint glass transition temperature measured by DSC at -36 ºC. The glass transition temperature of these materials was not measured because it was below the working range of the available equipment.

Table 2. Measured glass transition temperature of the studied interlayer materials.

<table>
<thead>
<tr>
<th>Material</th>
<th>20 ºC/min</th>
<th>10 ºC/min</th>
<th>5 ºC/min</th>
</tr>
</thead>
<tbody>
<tr>
<td>PVB DG-41</td>
<td>28.0 ºC</td>
<td>25.5 ºC</td>
<td>23.0 ºC</td>
</tr>
<tr>
<td>BG-R20</td>
<td>20.0 ºC</td>
<td>15.0 ºC</td>
<td>13.0 ºC</td>
</tr>
<tr>
<td>ES</td>
<td>29.0 ºC</td>
<td>26.3 ºC</td>
<td>24.5 ºC</td>
</tr>
<tr>
<td>SentryGlas</td>
<td>-</td>
<td>26.8 ºC</td>
<td>24.0 ºC</td>
</tr>
</tbody>
</table>

When the molecular weight of a polymer increases, so does its glass transition temperature [27]. This matches with the results obtained experimentally, since the glass transition temperature of PVB without plasticiser (PVB BG-R20) is lower than that of PVB with plasticiser (PVB DG-41 and PVB ES) [24].

For climate zones with temperate weather (long, generally hot summers and short, mild winters) a laminated glass element is expected to perform always above the glass transition temperature of EVA and TPU interlayers. On the other hand, in the case of PVB and SentryGlas interlayers, the outdoors temperature may range from below to above glass transition temperature. This must be taken into consideration in order to accurately design and calculate a structural laminated glass element.

3.2. Water absorption

Figure 8 shows the mass increase due to the water absorbed as a function of immersion time. On one hand, the mass of SentryGlas, PVB DG-41, PVB ES, and PVB DG-41 samples increased an average of 6.5%, 6.2%, 6.4%, and 4.3%, respectively, at the end of the immersion test. The water absorption was much higher at the beginning of the test: SentryGlas, PVB DG-41, and PVB ES achieved 80% of the total mass increase during the first 75 hours of immersion (19% of the total duration), and PVB BG-R20 samples saturated after approximately 55 hours of immersion. On the other hand, TPU, EVASAFE and EVALAM 80 had no relevant mass gain after 367 hours of
immersion (the average mass variation was smaller than 0.3% for each of the materials). Therefore, it is seen that an interlayer is able to absorb more or less water depending on the chemical nature of the polymer. Polymers with strong polar groups are able to bind water by hydrogen bridges and therefore be more sensitive to water exposure. For instance, interlayer materials based on PVB, due to its significant proportion of polar alcohol groups, are hygroscopic materials, which easily increase the water content, whereas EVA and TPU do not present such strong polar groups in its chemical structure, and therefore they are unable to absorb such significant quantities of water.

Figure 8. Mass increase (%) over time (hours) for the 7 interlayer materials immersed in deionized water.

Figure 9 shows the influence of water immersion on the material optical properties. Three moments of experimentation are shown for each sample. The one on the left is before test, the one
in the middle is after water immersion, and the one on the right is after drying. All interlayer materials tested were translucent before laminating, except TPU samples, which were green and rather opaque. Figure 9 shows how, after immersion, the materials that absorbed a more significant amount of water (SentryGlas and PVB) became white and opaque, whereas materials that did not absorb water (EVA and TPU) remained visually unaltered. At the end of the immersion test, SentryGlas and PVB interlayers were completely white and opaque, but when the samples were dried after the immersion they recovered the initial translucent aspect. This establishes a correlation between the water content in the interlayer material and its optical properties.

![Image](image.png)

Figure 9. Specimens before immersion (left), after immersion (middle), and dried after immersion (right).
3.3. Mechanical properties

The performance of a composite material like laminated glass depends on the properties of each one of the materials and the interaction between them. It is important to evaluate the mechanical properties of the interlayer material in laminated glass plates because it will affect its global structural response [28].

Tensile tests were carried out at three different elongation rates for unaged specimens. Another set of samples was exposed to different ageing factors separately (humidity, high temperature, and temperature cycles) in order to study the effect of each of them on the mechanical properties of the materials.

Figure 10 shows the stress-strain curves of all the unaged interlayer materials at a fixed elongation rate (10 mm/min). It shows that SentryGlas, PVB DG-41 and PVB ES have a very similar stress-strain curve, with an initial small non-linear region, followed by a softening zone, and a second region with a lower stiffness than the initial one. PVB BG-R20 has a lower stiffness for small deflections, but its stiffness keeps gradually increasing until breakage. The fact that the mechanical behaviour of PVB-BG-R20 differs so much from the other two PVB tested, shows how the addition of a plasticizer significantly affects the mechanical performance of that polymer. On the other hand, EVASAFE is the most ductile tested material, followed by the EVALAM 80, which has less than half the tensile strength of EVASAFE. TPU has an initial elastic region with progressive softening, followed by a linear region until breakage.

![Figure 10. Stress-strain curve for each one of the unaged interlayer materials at an elongation rate of 10 mm/min.](image-url)
Figure 11 shows how the different elongation rates influence the tensile strength and stiffness results. For each material and elongation rate, three specimens were tested; a curve with the average tensile strength and strain at breakage is presented for each material and elongation rate. Elongation rates used were 10 mm/min, 50 mm/min and 100 mm/min. The correlation between strain rate and mechanical properties was more visible for SENTRYGLAS, PVB DG-41, PVB BG-R20, PVB ES, and EVASAFE, but was less visible for EVALAM 80 and TPU. The highest variability in the results was obtained from TPU specimens.
Figure 11. Stress-strain curve for each interlayer material at different elongation rates.

(a) SentryGlas
(b) PVB DG-41
(c) PVB ES
(d) PVB BG-R20
(e) EVALAM 80
(f) EVASAFE
(g) TPU

Strain rate:
- 10 mm/min
- 50 mm/min
- 100 mm/min
Figure 12 shows how water immersion had a negative effect on the stiffness and ultimate tensile strength of the materials that absorbed a greater amount of water: SentryGlas, PVB DG-41, PVB ES, and PVB BG-R20. The decrease of strength and stiffness was less visible for EVALAM 80, EVASAFE and TPU. The temperature ageing, especially thermal cycles, led to an increase of stiffness for all the interlayers, except for TPU, for which the difference was less visible mostly due to the high dispersion of the results between identical tests and specimens.
Figure 12. Stress-strain curve for each interlayer material after exposure to different ageing factors.
The parameters that are considered more important for the implementation of the tested materials in laminated glass structural elements are toughness, maximum tensile stress, and initial stiffness. A higher toughness and maximum tensile stress are two key properties to increase the safety of laminated glass elements, because, in case of accidental glass breakage, the post-breakage load-bearing capacity of the interlayer allows the evacuation of civilians from the affected area and the replacement of the damaged elements. A higher initial stiffness leads to a higher transfer of shear loads between two connected glass surfaces, providing smaller out-of-plane deflections and therefore a higher out-of-plane stability.

The toughness presented in Figure 13 is the mechanical energy absorbed until breakage. EVASAFE was the material with the highest toughness, regardless of the strain rate and ageing factors, whereas TPU was the material with the lowest toughness and the highest dispersion in the results. After immersion, the toughness of SentryGlas and PVB decreased between a 76% and an 85%. EVASAFE was the only material which toughness increased after immersion (22%). There were no significant variations of toughness caused by exposure to high constant temperature and thermal cycles, except for EVALAM, which experienced an increase of 48% and 45%, respectively. None of the studied parameters had a significant effect on the toughness of TPU, since the dispersion between results in a group of specimens was greater than the difference between the mean values of each group.
Figure 13. Bar diagram displaying the average toughness for each interlayer material, elongation rate, and ageing factor, with a confidence interval of 95%.

Figure 14 shows how the addition of plasticiser to the PVB did not significantly increase the maximum tensile stress of the unaged specimens, meaning that the difference between average values was smaller than the confidence intervals. For SentryGlas and PVB, water absorption was the factor that affected most the maximum tensile stress value, reducing it between 77% and 87%. On the other hand, the maximum tensile strength of EVASAFE increased 12% after water immersion, which had no relevant effect on EVALAM 80 and TPU specimens.
Figure 14. Bar diagram displaying the average maximum stress for each interlayer material, elongation rate, and ageing factor, with a confidence interval of 95%.

Figure 15 shows that the initial stiffness of PVBs with plasticiser (PVB DG-41 and PVB ES) was between 11 and 24 times higher than without plasticiser (PVB BG-R20) for unaged specimens and up to 19 times higher for thermally aged specimens. The stiffness of SentryGlas and PVB increased between 51% and 106% when the specimens were exposed to thermal cycles. The initial stiffness of EVASAFE more than doubled the one of EVALAM 80.
4. Conclusions

In this paper, seven different interlayer materials, based on four base polymers (PVB, EVA, ionomer, and TPU) were selected to study their behaviour under different elongation rates and ageing factors. Tensile tests were carried out at three different elongation rates, and after three ageing tests (water immersion, high temperature, and thermal cycles). Additional water immersion tests were carried out to evaluate the amount of water absorbed over time and its effect on the optical properties of the interlayer.

SentryGlas, PVB DG-41, and PVB ES showed the highest stiffness. These three materials, together with PVB BG-R20, had the highest tensile strength. EVALAM 80 and EVASAFe showed the highest ductility. EVALAM 80, EVASAFe, and TPU are the materials with the lowest tensile strength, but their mechanical and optical properties are less affected by the tested ageing factors. SentryGlas, PVB DG-41, PVB ES, and PVB BG-R20 specimens showed a significant mass gain, strength decrease and transparency loss after immersion in water. After the drying process, the initial transparency was recovered. Temperature ageing (constant temperature and thermal cycling) did not significantly affect the maximum strength of these materials.
From the detailed analysis of the results from the experimental tests, it is concluded that the best candidates to be implemented for laminated glass plates for structural purposes are PVB with less plasticiser (PVB ES and PVB DG-41) and SentryGlas, because they present the highest initial stiffness and maximum stress when not exposed to humidity. For laminated glass elements with these interlayers, the exposure to humid environments could lead to a reduction of transparency and resistance at the edges of the panels. Nevertheless, such deterioration of mechanical and optical properties might be smaller in case of exposure to humidity and after the lamination process than it was with immersion in water of the polymeric film alone.

The performance of the tested interlayer materials may vary after the laminating process, since it involves high temperatures, pressures, and chemical reactions. Therefore, the results presented in this paper should be complemented by additional experimental results with laminated glass specimens. Dynamic tests at different temperatures are also needed to study the viscoelastic properties of these polymers.

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Data availability

The raw/processed data required to reproduce these findings cannot be shared at this time as the data also forms part of an ongoing study.

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