Determination, verification and durability of PVB interlayer modulus properties

Wim Stevels[1], Pol D' Haene[2]

[1] Eastman Chemical BV, Watermanweg 70, 3067 GG Rotterdam, The Netherlands, wimstevels@eastman.com
[2] Solutia bvba, Ottergemsesteenweg-Zuid 707, 9000, Gent, Belgium, ppdhae@eastman.com

Interlayer modulus properties are important for the calculation of the load resistance of laminated glass configurations. The determination of interlayer properties can be made on the interlayer material directly, or indirectly through measurements on laminated glass test specimens. One challenge relates to the four-decade range of modulus values that interlayer materials go through with variations of temperatures and duration that are relevant to building elements. Furthermore, in determination of properties directly on the interlayer, e.g. by dynamic mechanical analysis, methodological choices and data treatment play an important role, apart from experimental and material variation. To some extent, this also holds true for modulus determination on laminated glass specimens, with the added challenge that the properties of a relatively soft material are derived from the response of a rigid assembly. In this contribution, an overview is provided of material characterization of all major PVB interlayer formulations (conventional, structural, acoustic) by dynamic mechanical analysis, and validation of the models developed by four-point bending. In addition, it is shown that the properties determined do not change upon exposure to the weathering conditions of product standard EN ISO 12543.

Keywords

PVB interlayers, material model, Prony series, acoustic PVB, stiff PVB, glass design.

1 Introduction

Over the last five years, a number of developments have taken place that have influenced the determination of interlayer modulus properties, and their use in laminated glass design.
Significant research has been undertaken by academia and industry, to optimize, compare and expand measurement methods and data processing methodology, complemented with sensitivity analysis and, in some cases, inclusion of these data in standards. An extensive review of predominantly academic work and standards has been recently published by Kuntsche et al. [1]. Other relevant academic publications are available, and examples of additional or complementary industrial work are mostly available as conference contributions, as cited by Stevels and D'Haene [2]. The combined effort has led to a better understanding of best practices for methods and data treatment and validation.
After many years of discussion and development, EN 16612 [3] and the associated interlayer characterization standard EN 16613 [4] were published in 2019. Significant changes to the scope as compared to the enquiry versions of 2013 and 2017 have occurred, which limits this standard to guidance for the lateral load resistance of linearly supported glazed elements used as infill panels, in a class of consequence lower than those covered in EN 1990. Particularly to the use of interlayer properties, emphasis has shifted from use of effective thickness methods and stiffness families with default shear transfer coefficients to direct use of interlayer properties in FEM design. Much progress has been amde on the development of a Eurocode on glass design, resulting in (pre-)publication of a precursor technical specification for further development in the coming years [5]. Different levels of using interlayer properties are likely to be recognized, including layered limit states, effective thickness models and - for the purposes of finite element calculations - viscoelastic models based on Prony series.
It was with these developments in mind, that the properties of three different PVB (conventional, acoustic, stiff) formulations have been determined using current methodologies, with the results expressed as a viscoelastic model in Prony series format. These models were benchmarked against independently determined viscoelastic models, and validated by four-point bending experiments on laminated glass. The latter approach was also used to verify the stability of the viscoelastic properties upon exposure of the laminated glass specimen to the UV-radiation test of EN ISO 12543-4.

2 Experimental approach

Dynamic mechanical analysis (DMA) experiments were carried out on three different Saflex® interlayers from commercial production, all at a nominal thickness of 0.76 mm. These interlayers were respectively Saflex Clear 0.76 mm (formerly Saflex RB41- conventional PVB type), Saflex Structural 0.76 mm (formerly Saflex DG41 or DG41XC - stiff PVB type) and Saflex Acoustic (formerly Saflex QS41 - acoustic trilayer PVB type). For the latter product, an additional separate effort in validation was made that is described elsewhere [6]. The geometry used was a parallel plate set-up as per ISO 6721-10 [7], having a diameter of 8 mm. Using a larger plate diameter in combination with low temperature experiments and higher frequencies generally results in an overload of the transducer which is not recommended. For each temperature it was verified that the applied strain was maintained within the linear viscoelastic region. Experimentally, frequency sweeps were carried out in a frequency interval ranging from 0.1 rad/s to 100 rad/s. The temperature of the experiment was increased from -30°C to 90°C, starting far below the glass transition temperature of the interlayers. For the dynamic experiments, the shape of the measured sine wave was monitored to assure that no slippage at the interface of the sample occurred. Only horizontal shifts of the frequency shifts curves on the frequency axis were used to generate the mastercurve. Time-temperature superposition principles were applied using the Williams-Landel-Ferry ("WLF") equation to determine shift factors a_T. The relaxation spectrum was directly generated by the instrument software from the measured shear and loss storage modulus data. The relaxation spectrum was determined from the obtained master curve, at a reference temperature of 20°C, using a model containing 10-11 relaxation times and shear relaxation moduli. During the fitting process both parameter sets were considered as variables. The resulting relaxation data were interpolated to relaxation times regularly spaced in decades for user convenience and associated with the corresponding shear relaxation modulus values. The results of the model were compared to automatically generated mastercurves at different temperatures for reasonable fit. Reference viscoelastic models were provided by the Institute for Statics and Construction, Technical University Darmstadt, Darmstadt, Germany based on independent experimentation and evaluation (Schuster and Schneider, [8]). Results from four point bending experiments were provided by Institute of Structural Engineering, University of the German Armed Forces, München, Germany based on independent experimentation and evaluation (Kraus et al. [9]). Measurement reports can be provided upon request to the authors.

3 Results and discussion

In fitting the experimental data, a Prony series comprising 10-11 elements was chosen to balance accuracy with practicality as a model for G(t). This function is a mathematical representation of the molecular motion taking place in the polymer, which can be described mechanically as a series of Maxwell models:

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

$$\tau_i = \frac{\eta_i}{G_i} \, and \; Gi = G0 * gi$$

in which the modulus Gi reflects the relative magnitude of each process while the relaxation time τi indicates its timescale. Curve fitting was optimized over the number of elements disregarding the exact values of the relaxation time, which were later interpolated to more rounded relaxation times, while reviewing retainment of fit. For all products, a temperature shift function was developed based on the WLF equation of type:

$$log_{10} a_\tau(T, T_{ref}) = -\frac{C_1 * (T - T_{ref})}{C_2 + T - T_{ref}}$$

Thus, for each Prony series, values are provided for g_i, τ_i, G_0, G_∞, C_1 and C_2. The approach followed will be outlined in more detail for stiff PVB in 3.1, whereas in the interest of brevity the resulting models only will be provided in 3.2 for conventional and acoustic trilayer PVB.

3.1 Stiff PVB type

Thus far, Prony series for this stiff PVB (Saflex® Structural) were restricted to a German national approval [10] and a single academic publication [11]. The former provides interlayer properties allowed for design, not necessarily material properties, and the latter used creep relaxation experimentation on multiply laminates, which although feasible, is a labor intensive and time-consuming approach. The Prony series developed for stiff PVB type (Saflex Structural) using DMA is represented in Table 1, along with the WLF parameters C1 and C2.

Table 1. Model parameters for the evaluation shear relaxation modulus G(t,T) for Saflex Structural (formerly Saflex DG and Saflex DG XC) interlayers

	G0 = 576 MPa	**G∞= 0.23 MPa**
	Tref = 20 °C	**C1 = 21.3**
		C2 = 66.0
i	**relaxation time**	**gi (G_i/G_0)**
	(s)	
1	1.000E-01	0.1713
2	1.000E+00	0.1960
3	1.000E+01	0.2101
4	1.000E+02	0.2054
5	1.000E+03	0.1503
6	1.000E+04	0.0543
7	1.000E+05	0.0101
8	1.000E+06	0.0018
9	1.000E+07	0.0005
10	1.000E+10	0.0003

The shear relaxation modulus derived from the model for this stiff PVB is plotted in Figure 1 for temperatures of 0, 20, 30 and 40°C, as well as data from the independently developed model [8]. Over this temperature domain, values for shear relaxation modulus range over four decades. The shape of the mastercurve is consistent over both models, and at the reference temperature of 20°C, the models align reasonably well. The model of Table 1 is more conservative at higher temperatures and longer durations as compared to the independently developed model, as readily apparent in the 20 – 40°C range. The largest discrepancy is observed at around the glass transition temperature of 30°C. Apart from potential differences in samples, equipment and measurement practices, the curve fitting of the temperature shift function plays a major role in the outcome of the models. In order to optimize fit, a 5th order polynomial function was used for the curve fitting of the external data (dotted lines). In terms of glass design, the less conservative numbers that are generated by the model of Table 1 at low temperatures are unlikely to be critical, as the values exceed the threshold for full shear coupling by a wide margin, at least for durations up to 1 month.

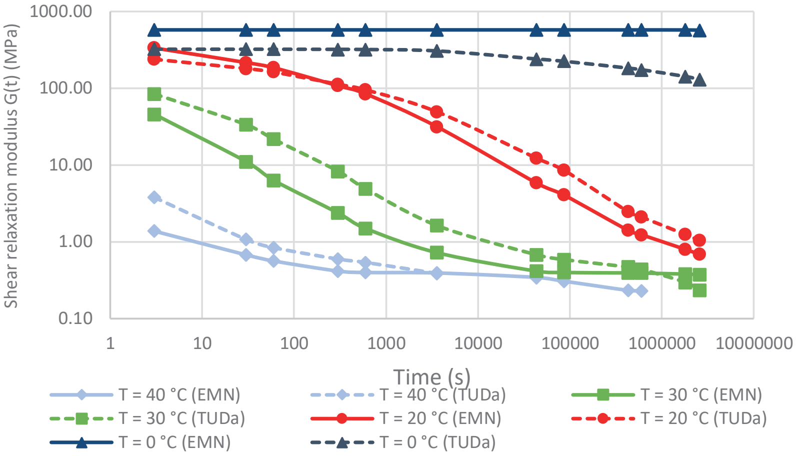

Figure 1. Shear relaxation modulus for stiff PVB for durations between 3s and 1 month at different temperatures for the model of Table 3 (solid lines) and the independent model (dotted lines, [8])

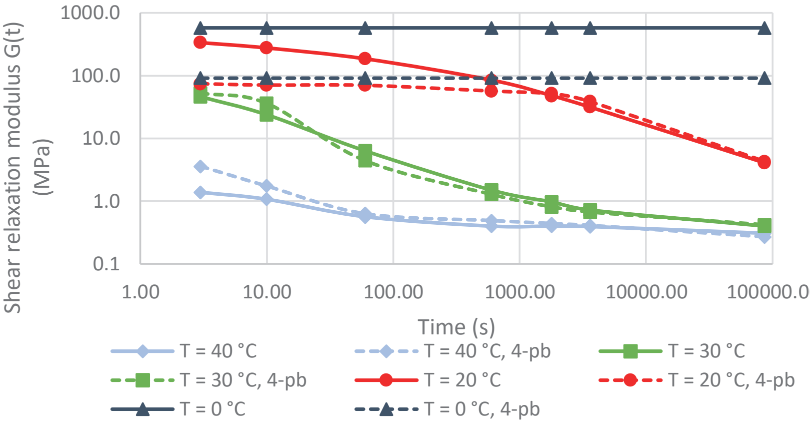

Figure 2. Shear relaxation modulus for stiff PVB for durations between 3s and 24 hours at different temperatures for the model of Table 3 (solid lines), and as derived from four point bending experiments (dotted lines, [9])

To validate the model of Table 1, four-point bending experiments on laminated glass specimens of 360*1100 mm were executed over a 0 to 40°C temperature range at the University of the German Armed Forces (Munich, Germany) following the generic guidance of EN 16613 taking into the remarks made in Botz et al. [12], with details provided in Kraus et al. 2019 [9]. A comparison of the modulus values derived from these measurements is given in Figure 2 (note changed scale of x-axis vs. Figure 1). While the exact value of the interlayer modulus cannot be determined above the fully coupled limit using four-point bending, the relaxation behavior of the laminate is very accurately predicted in the 20-40°C range.

3.2 Conventional and acoustic PVB types

A similar approach as in 3.1 was followed for conventional (Saflex® Clear RB) and acoustic trilayer PVB (Saflex Acoustic QS) types. A full description in the work is available from Stevels and D'Haene [2] – including addtional references to other work. The resulting models are provided in Table 2.

4 Model limitations

Mathematically, the models developed can be evaluated for any duration and temperature. However, the relaxation times and the number of relaxation times chosen, and the curve fitting were directed at accurately reflecting changes as relevant for the range of temperatures and duration that glass in buildings routinely experience. Time temperature superposition can only be applied to materials going through a single relaxation process (e.g. glass transition), and care must be taken to not extend the generation of base data outside of this range. In cases where either the elastic limit state or the viscous limit state is approached, care should be taken to apply these models for the generation of absolute modulus values. In practical terms, the validation of the models can only take place over a certain time and temperature range as well. In the time/frequency domain, these models were not developed to accurately reflect the interlayer behavior for very short-term loads (e.g. bomb blast), or very long term loads (e.g. permanent). In the temperature domain, data can be generated in the range in the 0 to 70°C range, although the upper and lower limit may differ slightly per product type. The models for conventional and stiff PVB can be considered fully validated for a temperature range between 10 and 60°C, and durations from 3 seconds up to 1 month. For the acoustic trilayer product, the applicable range is more limited to durations of up to 1 hour and temperatures up to 40°C.

Table 2. Model parameters for the evaluation shear relaxation modulus G(t,T) for conventional [8] and acoustic trilayer PVB

	Conventional PVB	**Trilayer AcousticPVB**		
	G_0 = 97.37 MPa	**G_∞ = 0.008 MPa**	**G_0 = 368 MPa**	**G_∞ = 0 MPa**
	T_{ref} = 20 °C	**C1 = 33.2**	**T_{ref} = 20 °C**	**C1 = 35.9**
		C2 = 212.4		**C2 = 266.3**
i	**relaxation time**	**g_i (G_i/G_0)**	**relaxation time**	**g_i (G_i/G_0)**
	(s)		(s)	
1	1.00E-02	0.2995	1.000E-09	0.2029
2	1.00E-01	0.2997	1.000E-08	0.2030
3	1.00E+00	0.2790	1.000E-06	0.2597
4	1.00E+01	0.0912	1.000E-04	0.2331
5	1.00E+02	0.0207	1.000E-03	0.0672
6	1.00E+03	0.0044	1.000E-02	0.0236
7	1.00E+04	0.0013	1.000E+00	0.0042
8	1.00E+05	0.0006	1.000E+01	0.0035
9	1.00E+06	0.0008	1.000E+02	0.0015
10	1.00E+07	0.0028	1.000E+04	0.0004
11			1.000E+06	0.0004

5 Durability

Three laminated glass specimens were produced on a commercial line for a durability test to EN ISO 12543-4 (UV-radiation) in a 66.2 configuration (360 * 1100 mm) using a 0.76 mm stiff PVB type (Saflex® Structural) as interlayer. These panels - as produced - were subjected to four-point bending experiments in creep mode using a 250 N load at 20, 30 and 50 °C, with deflections monitored for respectively 72, 48 and 24 hours [13]. Panels were conditioned for 24 hours before applying the load to reduce any effect of creep under self-weight. A FEM study was executed to assess deflections under the load applied in the fully coupled and decoupled limit states and found to be respectively 1.2 and 5.8 mm. After characterization of the initial behavior of the panel (results marked 'I' for initial), the panels were exposed to the conditions of the UV-radiation test. This test has a duration of 2000 hours and requires a radiation intensity of 700 W/m2 at 45 °C. The set-up is illustrated in Figure 3. After exposure for 2000 hours, re-characterization of the panels as above was executed as outlined above. Results were expressed as either deflection (Figure 4) or modulus (Figure 5).

Practically, it proved difficult to get reliable readings of the deflection at very short time frames in this experiment, especially if the interlayer is relatively stiff. Potential causes are friction of the panels on the supports upon applying the load and poor damping of vibrations after applying the load. Readings stabilize after 10 s – 1 min. The deflections measured are within the range for the modelled limit states. For the experiment at 20 °C, the measured deflection is very close to the fully coupled limit state at short durations, which is to be expected based on the interlayer properties.

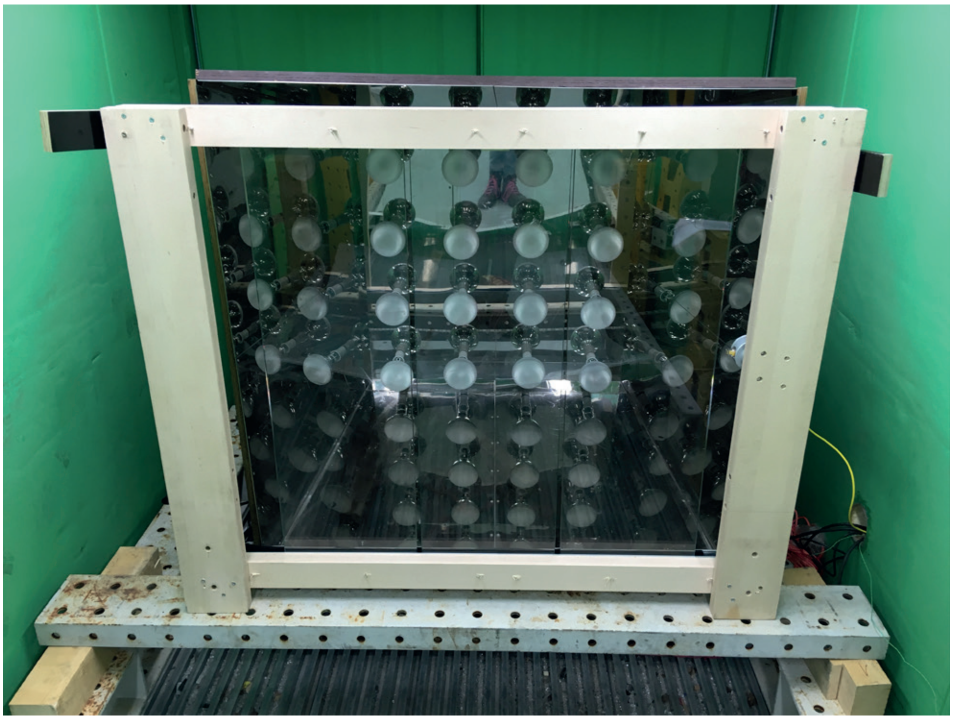

Figure 3. Overview of the test-up of the UV-radiation. Note that the edges of the three different glass specimens are marked with a thin blue line to make them visible in the picture.

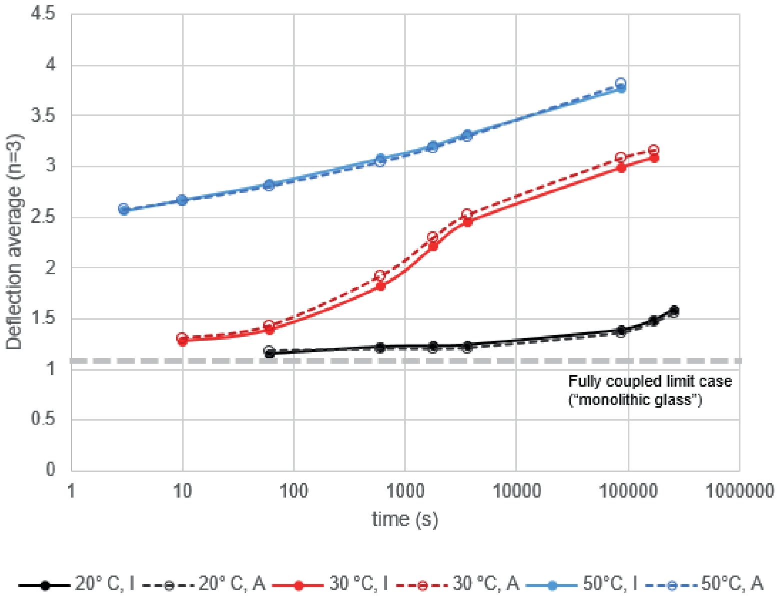

Figure 4. Deflections as measured on the panels under load prior to exposure (marked I for initial state, solid lines) and after exposure (marked A for aged state, dotted lines) to EN ISO 12543-4.

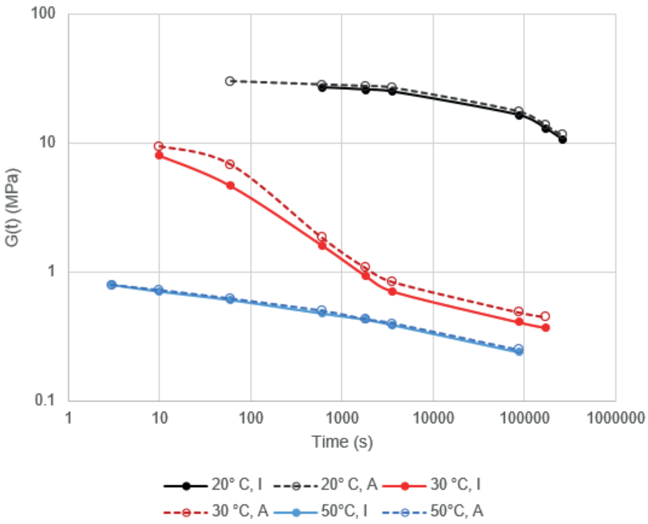

Figure 5. Modulus values derived from the deflections as measured in Figure 4 on the panels under load prior to exposure (marked I for initial state, solid lines) and after exposure (marked A for aged state, dotted lines) to EN ISO 12543-4

Overall, the deflections indicate that at 20 °C relatively little relaxation takes place over the duration measured, at 30 °C significant duration takes place over the duration the measurement, and at 50 °C a signifcant amount of relaxation has already taken place prior to obtaining the first reliable data point. This is consistent with the properties as reported in 3.1.

Most importantly, the measured deflections or modulus values before and after exposure to the UV-radiation test are similar within the accuracy of the experiments, indicating robustness of the viscoelastic properties for glazing in use comprising this interlayer. It should be noted that conventional and acoustic PVB types are closer to viscous regime/ decoupled limit in use and therefore the curves of Figure 4 and 5 would be more closely spaced. Among the three different PVB types discussed, stiff PVB types are the most critical case, and the conclusions with regards to the robustness of viscoelastic properties would hold for the other PVB types as well.

6 Conclusions

Material models generating shear relaxation models were developed for all three main commercial PVB types. These models were compared to independently developed models. The model that was more conservative at longer durations/higher temperature was selected for validation with four-point bending experiments at different temperatures. Generically, good agreement was achieved except for assemblies where full shear coupling is achieved. In those cases, four-point bending experiments cannot accurately provide interlayer modulus values.

Although the models can be used to generate modulus values for a wide range of load scenarios, care should be taken to not use load scenarios outside the validated temperature or time range.

The mechanical properties of glass laminates did not change upon exposure to UV radiation to EN ISO 12543-4, indicating robustness of the viscoelastic properties for glazing in use comprising the PVB interlayers studied.

7 References

[1] Kuntsche, J.; Schuster, M.; Schneider, J.: Engineering design of laminated safety glass considering shear coupling: a review, Glass. Struct. Eng. 2019-4, 209-228, 2019.

[2] Stevels, W.M.; D'Haene, P.: Interlayer and verification of PVB interlayer modulus properties, accepted – publication pending.

[3] European Committee for Standardization: EN 16612 Glass in Building – Determination of the load resistance of glass panes by calculation and testing (2019).

[4] European Committee for Standardization: EN 16613 Glass in Building – Determination of interlayer

mechanical properties (2019).
[5] European Committee for Standardization, prCEN/TS xxxx-1:2019 Design of glass - Part 1: Basis of design and materials
[6] D'Haene, P.; Stevels, W. Modelling laminated glass based on multilayer interlayers: effective modulus approach. In: Proceedings Glass Performance Days (Tampere Finland), 2019, pp. 412-415.
[6] International Organization for Standardization: ISO 6721-10 Plastics – Determination of Dynamic Mechanical Properties_ Complex shear viscosity using a parallel plate oscillatory rheometer (2011).
[8] Schuster, M.; Schneider, J.: Investigations on the behaviour of the Saflex® PVB in laminated safety glass after glass fracture Report_Linear Viscoelasticity: DMTA / Rheometer, Proprietary report (2019).
[9] Kraus, M.; Botz, M.; Pauli, A.; Siebert, G.: Investigations on the behaviour of the Saflex PVB in laminated safety glass after glass fracture_Linear Viscoelasticity: Four-Point Bending Tests, Proprietary report (2019).
[10] German building institute: Verbund-Sicherheitsglas aus der Produktfamilie Saflex DG mit Schubverbund. Generic building approval Z-70.3-230, Berlin 2016.
[11] Piscitelli, L. Serviceability and post-failure behaviour of laminated glass structural elements, Thesis University of Braunschweig – Institute of Technology (2018).
[12] Botz, M.; Kraus, M.A.; Siebert, G.: Experimental determination of the shear modulus of polymeric interlayers used in laminated glass. In: Proceedings GlassCon Global 2018, pp.31- 38.
[13] Pauli, A.; Siebert, G.: Investigations on the behaviour of Saflex Structural in laminated safety glass after ageing due to UV radiation, Proprietary report (2020).