

Investigations on the viscoelastic material behaviour and linearity limits of PVB

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The consideration of the shear bond via the intermediate layer in the design of laminated glass is of great interest, as this can lead to a considerable reduction of the maximum stresses in the glass, depending on the load and static system. Polyvinyl butyral based interlayers show a typical polymeric thermo-viscoelastic material behaviour, which means that the shear coupling depends on the load (duration and magnitude) and the prevailing temperature. With small distortions below the linearity limit, the time-dependent material behaviour of the intermediate layer can be mapped using the Prony-Series. However, the linear viscoelastic range is exceeded at latest in the post-fracture behaviour and hence the Prony-Series can no longer be used straightforwardly. This article shows the experimentally determined limits of the linear viscoelastic range using the example of PVB. In addition, the Prony parameters for the linear viscoelastic range are determined.

Keywords: laminated glass interlayers, polyvinyl butyral, viscoelasticity, linearity limits

1 Introduction

The increasing use of glass as a structural element in the building industry allows the realization of almost transparent structures. In order to fulfil safety-relevant requirements with regard to fracture and post-fracture behaviour, laminated safety glass (LSG) is applied. Application situations in which LSG is required are for example fall-protection glazing, walk-on glazing and overhead glazing. According to EN ISO 12543-2 [1], LSG consists of (at least) two glass panes and a polymeric interlayer. After failure of the brittle glass, the glass fragments remain bonded to the interlayer and a residual load bearing behaviour is enabled.

In addition to the safety-relevant aspects after glass fracture, the interlayer causes a shear coupling between the glass panes in the intact laminated safety glass, which positively affects the maximum stresses in the glass panes. The importance of the shear coupling effect depends on the interlayer stiffness, the structural system (glass dimensions, thicknesses of the single layers, support conditions) and the applied load (uniform surface load,

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single load). However, for most cases with dimensions, glass and interlayer thicknesses and impact factors relevant to construction practice, shear modulus values G of 0.1 N/mm^2 may already reduce the glass stresses significantly; shear modulus values G of 10 N/mm^2 may result in full shear transfer [2], [3], [4]. This can be explained by the commonly known sandwich theories, e.g. [5-7].

The most common interlayer products in building industry are polyvinyl butyral (PVB)-based with a market share of over 90 %. Other interlayers may for example be ethylene vinyl acetate (EVA)-based, ionoplastics (e.g. SentryGlas®), thermoplastic urethane (TPU) or cast resin. In addition to the variety of products, which cannot be described in a uniform way, polymers show a temperature dependent and viscoelastic material behaviour.

Standards and guidelines offer a wide variety of approaches to respect the shear coupling effect during glass design. The prCENTS-2 [8] for example is considering different levels of accuracy. The most accurate level is considering a transient linear viscoelastic Finite element model based on the generalized Maxwell-Model, which is mathematically described by a so-called Prony-Series, and a Time-Temperature-Superposition principle (TTSP) for the interlayer to determine the maximum glass stress. The TTSP enables to transfer the Prony-Series from a reference temperature T_{ref} to a different interlayer temperature T under certain conditions (see further e.g. [9]). Alternatively, the Prony-Series can be evaluated for certain load cases, e.g. snow ($0 \text{ }^\circ\text{C}$, 30 d), and a linear elastic material behaviour with a constant shear modulus value can be assumed for the design under this specific load case. The values of the interlayer stiffnesses can be taken from technical data sheets or technical approvals in the form of Prony-Series and TTSP or tables with load-specific values, e.g. Z-70.3-230 [10], or can be determined experimentally, e.g. EN 16613 [11]. However, it should not be forgotten that the values given are based on linear viscoelastic material models, where the stiffness $G(T, t)$ depends on the prevailing temperature T and the load duration t but is independent on the load magnitude γ or τ , and therefore application limits must be defined in the form of linearity limits.

This paper focuses on the experimentally determination of the limits of the linear viscoelastic range using the example of a standard PVB. In addition, the Prony parameters and TTSP for the linear viscoelastic range are presented.

2 Polymeric Interlayers

2.1 Linear Viscoelasticity and Thermorheology

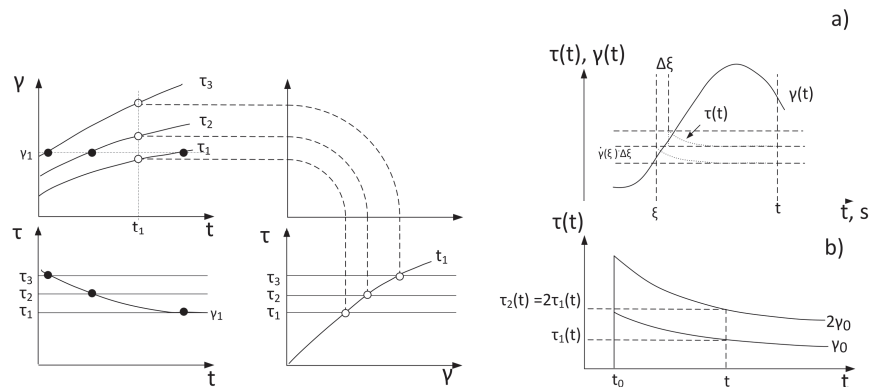


Figure 2-1 Isochronous stress-strain diagram (left), Boltzmann superposition principle (a) and linear scalability (b) (right).

Viscoelastic material behaviour can be linear or nonlinear. For strains smaller than the so-called linearity limit, the material behaviour of the polymeric interlayer can be described with the theory of linear viscoelasticity. Here, stress and strain are proportional to each other at any time and the relaxation function $G(t) = \tau(t)/\gamma_0$ is independent of the applied distortion amplitude (or stress amplitude). This can be seen in isochronous stress-strain diagrams, in which the synchronously measured stresses are plotted for different applied strains, see Figure 2-1 (left). Linear viscoelastic materials follow the Boltzmann superposition principle at constant temperature. When the history of the applied strain γ up to the time t is known ($-\infty < \xi \leq t$ where ξ is a time of prehistory), the Boltzmann superposition principle allows determining the resulting stress τ , by linear summation of the individual stress relaxation processes caused by the strain loading history, see Figure 2-1a (right). This also leads to linear scalability, which means that when the strain is e.g. doubled, the resulting stress is also doubled, see Figure 2-1b (right). [12]

The relaxation function of linear viscoelastic materials can be described by the generalized Maxwell-Model (Figure 2-2) and a so-called Prony-Series, [13], [14], [12]. The Prony parameters g_k (or G_k), $\tilde{\tau}_k$ and G_0 (or G_∞) are to be determined experimentally, e.g. in Dynamical-Mechanical-Thermal Analysis (DMTA). In DMTA, the viscoelastic body is loaded by sinusoidal oscillations and the delayed response with a phase shift of $0^\circ \leq \delta \leq 90^\circ$ is measured and evaluated to a complex modulus $G^*(\omega)$, whose real part is called storage modulus $G'(\omega)$ and whose imaginary part is called loss modulus $G''(\omega)$. The ratio of loss modulus to storage modulus is defined as the loss factor $\tan(\delta) = G''/G'$.

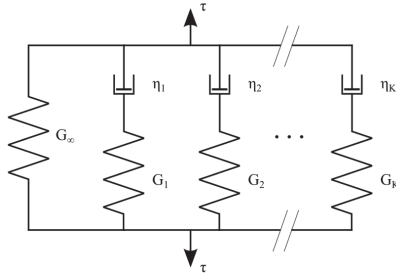


Figure 2-2 Generalized Maxwell-Model.

$$G(t, T) = G_0 \left(1 - \sum_{k=1}^K g_k \left(1 - e^{-\frac{t}{a_T \tilde{\tau}_k}} \right) \right) \quad (2.1)$$

With:

$$\tilde{\tau}_k = \eta_k / G_k$$

$$\log_{10} a_T(T, T_{\text{ref}}) = -\frac{C_1(T - T_{\text{ref}})}{C_2 + T - T_{\text{ref}}} \quad (2.2)$$

According to e.g. Schwarzl [12], Brinson and Brinson [15], Riande, et al. [16] and Williams, et al. [17], relaxation processes are based on thermally activatable molecular movements and rearrangement processes and hence a temperature increase leads to accelerated relaxation processes. For thermorheologically simple materials, all relaxation times show the same dependency on temperature and the entire relaxation curve can be shifted along the horizontal time axis by a specific temperature-dependent displacement factor $\log a_T$. In mathematical terms, all relaxation times are multiplied by the shift factor.

PVB is supposed to behave thermorheologically simple and the WLF approach provides a good adaptation to the experimentally determined displacement factors. Accordingly, the linear viscoelastic material behaviour of PVB interlayers in laminated glass can be thoroughly described via the Prony parameters and the WLF constants in the building-relevant time and temperature range.

2.2 Linearity Limits

The delimitation to nonlinear material behaviour is characterized by the linearity limit. The linearity limit is a strain-related value that represents the upper limit of the linear viscoelastic range. If this differentiation is given in relation to stress, this is referred to as compliance limit [18]. A research about the linearity limit of polymers quickly leads to

the realization that the values depend very much on the stiffness of the material. Temperatures above the glass transition temperature lead not only to a softening of the material, but also to a significantly higher linearity limit. Jansson [19] and Mezger [18] also write that the linearity limit depends on the load duration and the frequency. This means that with a longer load duration or lower frequency, lower stiffness values occur and hence the linearity limit increases. In contrast to this, the standard for the determination of dynamic-mechanical properties of plastics EN ISO 6721-1 [20] recommends a frequency of 1 Hz.

There are various ways of distinguishing between linear and non-linear material behaviour. For the evaluation of quasi static tests, Papanicolaou and Zaoutsos [21] mention the verification of the validity of Boltzmann's superposition principle and the linear relationship between stress and strain (constant stiffness) at any point in time (isochronous). If one of the two conditions is violated, the material behaviour is no longer linear. Stress-Strain-Diagrams can also be generated out of DMTA amplitude-sweeps, where the strain amplitude is increased. However, it is common to directly evaluate the storage modulus - strain-curve.

To determine the linearity limit, a maximum deviation from the stiffness in the linear range has to be defined. Additionally, it has to be defined, whether the deviation refers to the first measured value (smallest strain) or to the average value of the measuring points in the linear range. While Ehrenstein [22] defines the linearity limit as the strain level at which a deviation of 1 % from the initial stiffness occurs, Bertilsson and Jansson [23] write that the linearity limit occurs at 1 % deviation from the average value in the linear viscoelastic range. Yannas [24] on the other hand evaluates the linearity limits in his work with a deviation of 3 %. Mezger [18] suggests that the deviation of the storage modulus in the linear viscoelastic area necessary to determine the linearity limit can be set at 3 %, 5 % or even 10 % and recommends a deviation of 5 % for polymers. In the standard for determination of the complex shear modulus and phase angle for bitumen and bituminous materials, the linearity limit is defined at a deviation of the storage or loss modulus of 5 % from the initial value [25]. In the second part of the standard for the testing of lubricants the limit of the linear viscoelastic range is determined if G' deviates by 10 % from the respective plateau value [26].

Schneider, et al. [14], Grellmann and Seidler [13], Dominghaus [27] and Meissner [28] give a value for the linearity limit for polymers in the energy-elastic area of 1 % and a value of 100 % in the entropy-elastic area. However, these are only rough values, which are mostly due to much older publications. Yannas [24] dealt more closely with the linearity limits of amorphous polymers by conducting experiments on polycarbonate, with linearity limits between 0.8 % and 1.25 % in the energy-elastic state. In addition to his results, Yannas used the results of various publications from the years 1944 to 1965, in which relaxation tests were carried out on natural rubber and polyisobutylene. The determined linearity limits are 25 % to 150 % in the entropy elastic range and 10 % in the range of the glass transition temperature. Furthermore, the values for the linearity limits

were graphically displayed as a function of the difference between the examination temperature and the glass transition temperature, as shown in Figure 2-3. Schwarzl [12] also compiled a summary of these investigations, including graphic evaluations. Further information on the linearity limits of polymers found in literature are summarized in Table 2-1.

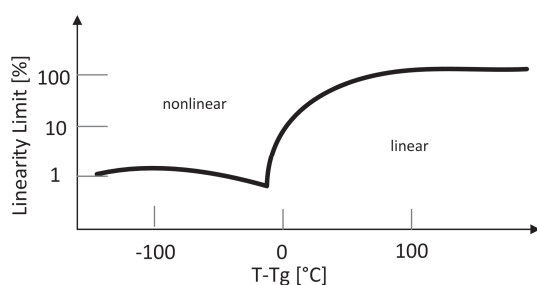


Figure 2-3 Linearity limit, reproduced from Yannas et al. [29].

Table 2-1 Linearity limits of polymers.

Reference	Material	Temp. range	Lin. Limit [%]
Mezger [18]	polymers in general	energy elastic	0.1
Retting [30]	high polymeric materials	energy elastic	0.1–0.5
Schuler [31]	amorphous thermoplastics	energy elastic	0.1–0.5
Staverman and Schwarzl [32]	different polymers	energy elastic	0.1–1
Ensslen [33]	thermoplastics	energy elastic	0.5
Bertilsson and Jansson [23]	thermoplastics	energy elastic	0.5–1
Jansson [19]	polymers	energy elastic	0.71
Yannas, et al. [29]	polycarbonat	energy elastic	0.8–1.25
Schmachtenberg [34]	amorphous materials	energy elastic	1
Yannas, et al. [29]	polyisobutylene	glass transition	10
Staverman and Schwarzl [32]	rubber	entropy elastic	10–100
Brinson and Brinson [15]	rubbery materials	entropy elastic	20–50
Yannas, et al. [29]	polyisobutylene, natural rubber	entropy elastic	25–150

3 Experimental Investigations

3.1 Sample Preparation and Test Set-Up

The standard PVB Saflex® RB from Eastman was examined.

In order to avoid shrinkage during the measurements, the PVB sheets were heated up to 100 °C and kept at that temperature for 1 hour before the samples were punched out with the aid of perforated irons $\varnothing = 8$ mm. The samples were then stored for at least 48 hours at room temperature in sealed glass containers filled with drying beads.

The experiments were performed on an Anton Paar MCR 302 rheometer using a plate-plate measuring system. The upper plate has a diameter of 8 mm and a textured surface, (see Figure 3-1 (right)). The convection oven was used in combination with liquid nitrogen for heating and cooling.

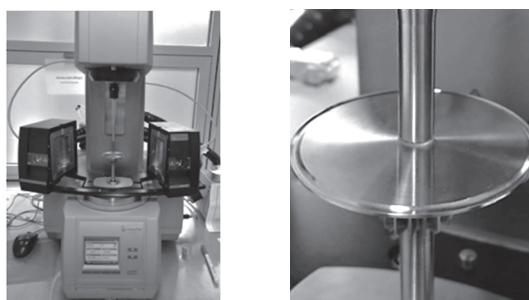


Figure 3-1 Rheometer MCR 302 (left), Plate-Plate Test set-up (right).

After sample installation, the samples were briefly loaded at a temperature of 65 °C with a normal force of 5–10 N, and unloaded (0–0.1 N). This should prevent slippage during the measurement. To relax the resulting stresses in the samples, they were then heated in a controlled manner to 100–120 °C and finally tempered to the test temperature with a cooling rate of 5 K/min.

3.2 Linear Viscoelasticity: Temperature-Frequency-Sweeps

3.2.1 Test Procedure

The temperature-frequency-sweep (TFS) was performed in a temperature range of T [100; -40] °C, with temperature steps of -5 °C. The excitation amplitude at low temperatures was 0.05 % and at higher temperatures 0.1 %. It was checked in integrated amplitude-sweeps that the linear viscoelastic range is not left during the measurement. The test parameters are summarized in Table 3-1.

Table 3-1 Test matrix temperature-frequency-sweeps.

Temperature program [°C]	[100:-40] cooling -5 °C steps Nitrogen
Frequency [Hz]	[0.1:10]
Incitation	0.1 % in T [100:40] 0.05 % in T [35:-40]
Contact normal force	0,1 N pressure
Tested samples	1

3.2.2 Test Results

A mastercurve for the reference temperature T_{ref} of 20 °C was created (Figure 3-2). The individual complex modulus $|G^*|$ frequency curves were incrementally horizontally shifted until a continuous curve was obtained. The horizontal displacement factors were then mathematically approximated by the time-temperature displacement principle of William-Landel-Ferry. The Prony-Series was determined with the institute's own Matlab script [35], taking the mastercurves of the storage modulus G' and the loss modulus G'' into account. Here, on the one hand, a Prony-Series was determined which represents the complete frequency resp. time range of the mastercurve and a reduced Prony-Series for engineering applications which is limited to 10 decades, see Figure 3-2 and Table 3-2. The equilibrium modulus G_{∞} was manually adjusted to 0, which prevents the shear modulus from being overestimated for long load periods and fulfils the theoretical requirement of thermoplastics.

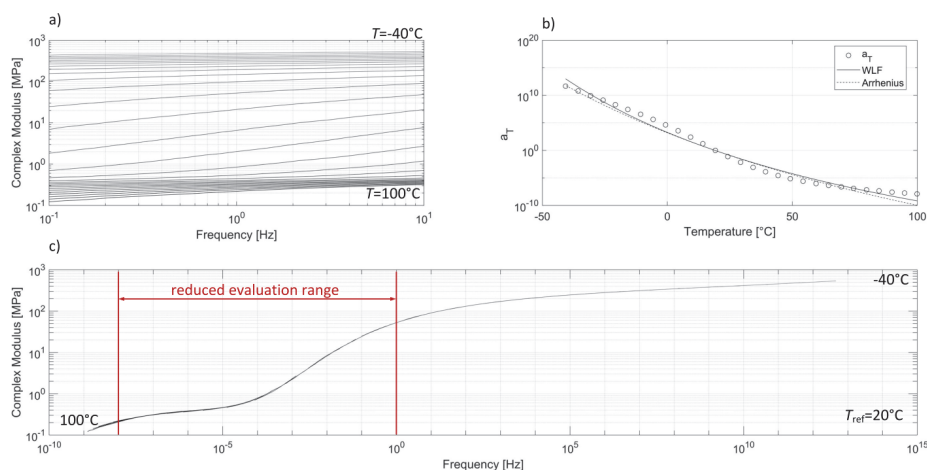


Figure 3-2 (a) measured data, (b) horizontal shift factors, (c) mastercurve at $T_{\text{ref}} = 20$ °C (exact value: $^{\circ}T_{\text{ref}} = 19.2$ °C).

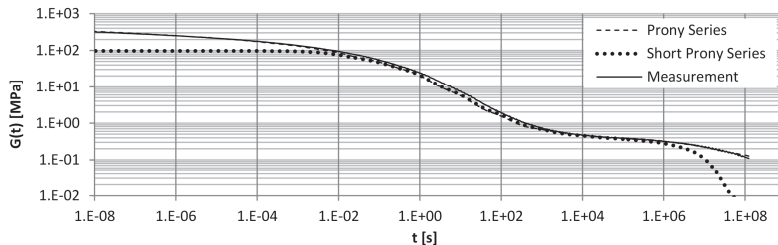


Figure 3-3 Time-dependent shear-modulus at $T_{ref} = 20\text{ }^{\circ}\text{C}$ (exact value: $T_{ref} = 19.2\text{ }^{\circ}\text{C}$).

Table 3-2 Reduced Prony-Series for engineering application.

Time-Temperature-Superposition: WLF		
C_1	33,20	-
C_2	212,42	$^{\circ}\text{C}$
Prony-Series		
G_0	97,37	MPa
i	$\tilde{\tau}_i$	g_i
1	1,00E+07	2,77E-03
2	1,00E+06	8,26E-04
3	1,00E+05	6,07E-04
4	1,00E+04	1,25E-03
5	1,00E+03	4,37E-03
6	1,00E+02	2,07E-02
7	1,00E+01	9,12E-02
8	1,00E+00	2,79E-01
9	1,00E-01	3,00E-01
10	1,00E-02	2,99E-01

The measurement data at a frequency of 1 Hz for all temperatures of the TFS were used to determine the glass transition temperature. The maximum of the loss factor led to $T_g = 30\text{ }^{\circ}\text{C}$, see Figure 3-4. However, it should be pointed out again that the temperature increment was $\Delta T = -5\text{ }^{\circ}\text{C}$.

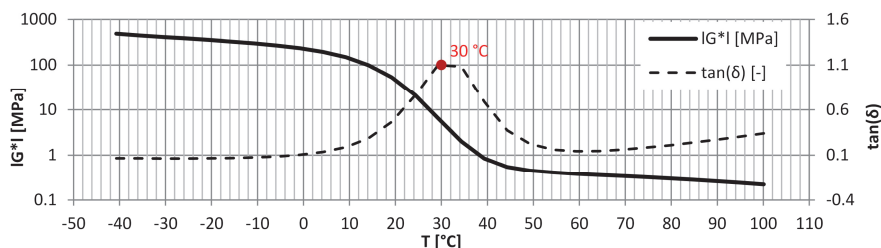


Figure 3-4 Temperature dependency, $f = 1\text{ Hz}$.

3.3 Linearity Limits: Amplitude-Sweeps

3.3.1 Test Procedure

The amplitude-sweeps were performed with linearly increased distortions for different temperatures T and different frequencies f . The most temperature levels were examined at a frequency of 1 Hz. For deviating frequencies (0.1 Hz) only a few tests were carried out in different temperature ranges. The test parameters are summarized in Table 3-3 and Table 3-4.

Table 3-3 Test matrix amplitude-sweeps.

Strain amplitude range	[0.1–35] %, dependent on T
Contact normal force	0,1 N pressure

Table 3-4 Nb.° of samples at temperature T [°C] and frequency f [Hz].

T [°C]	f [Hz]	
	0.1	1
8	-	1
10	-	1
15	-	1
17.5	-	1
20	1	2
22.5	-	1
25	-	1
27.5	-	1
30	1	2
32.5	-	2
35	-	1
37.5	-	1
40	-	1
45	-	1

3.3.2 Test Results

The linearity limit γ_{lin} was defined to the strain value at which the deviation of the storage modulus G' from the mean storage modulus in the linear viscoelastic area $G'_{lin-visko-el}$ reaches 3 %. To determine the mean storage modulus of the linear viscoelastic area $G'_{lin-visko-el}$ under consideration of a certain measuring noise, the deviation of the actual measurement point from the mean value of all previous measured values is determined. $G'_{lin-visko-el}$ is then calculated from the average of the values with a deviation less than

0.5 %. The deviation for all measured values is then determined from this mean modulus. If the deviation reaches 3 %, this point is defined as the linearity limit.

Figure 3-5 (left) shows the procedure for the Saflex® RB interlayer at a temperature of 20 °C and a frequency of 1 Hz. To check that the stiffness decrease is not caused by slippage, the amplitude-sweeps were driven both forward (from small to large strains) and backward (from large to small strains). If both curves lie on top of each other, it can be assumed that no slippage has occurred.

Figure 3-5 (right) shows the linearity limits determined as a function of temperature for two different frequencies (1 Hz and 0.1 Hz). At a frequency of 1 Hz, 14 different temperature levels were investigated. The selected temperature levels cover temperatures in the entropiy-elastic as well as in the energy-elastic area and the glass transition area. It becomes apparent that the linearity limit increases with increasing temperature. This correlates to the literature references described before, see e.g. Figure 2-3. The linearity limit extends over a range of approx. 0.5 % at low temperatures and 35 % at high temperatures. At temperatures below 15 °C, the linearity limit seems to increase again, but this could be due to the experimental setup. A new sample was used for each amplitude sweep, which means that there may be small differences in the sample preparation and in the sample installation. At the frequency of 0.1 Hz, only two temperatures were investigated. The linearity limits are higher compared to the measurement data at 1 Hz. Remembering the thermorheology (time resp. frequency temperature analogy), a lower frequency results in a lower stiffness. Evaluating the TTSP with the parameters given in Table 3-4 the change in frequency from 1 Hz to 0.1 Hz leads to a shift factor of $\log_{10}a_T = 1$, which in turn corresponds to a temperature decrease of $\Delta T = 6.21$ °C.

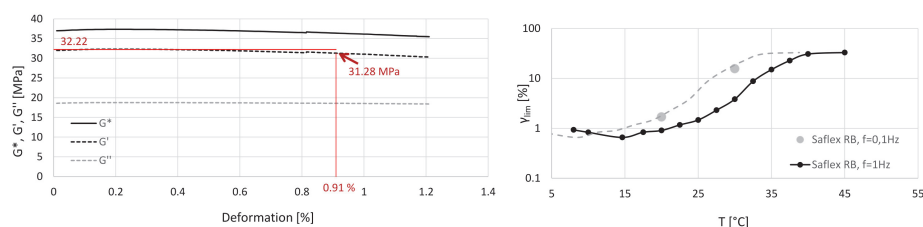


Figure 3-5 Amplitude Sweep, $T = 20$ °C, $f = 1$ Hz (left), Horizontal shift of linearity limits $\log_{10}a_T = 1$, $\Delta T = 6.21$ °C (right).

By shifting the black line in Figure 3-5 (right) with the calculated shift factor, the gray dashed line is obtained. As can be seen this line correlates well with the measured values. That shows that the procedure used for the shear modulus can also be applied to the linearity limits. Hence, the linearity limit can be specified via the linear viscoelastic stiffness (e.g. storage modulus) irrespective of the prevailing temperature and frequency, as shown in Figure 3-6.

3.3.3 Comparison with other PVB-based interlayers

For the structural PVB Saflex® DG, tests in the linear viscoelastic range were carried out and a Prony-Series and TTSP were derived. In addition, a large number of amplitude-sweeps were performed to determine the linearity limits. A total of 3 different frequencies 1 Hz, 0.1 Hz and 10 Hz were investigated. The PVB types Saflex® DG and Saflex® RB differ primarily in their plasticizer content.

Figure 3-6 shows that all linearity limits, both for the standard product Saflex® RB and for the structural product Saflex® DG plotted as a function of the storage modulus, coincide. This can be explained by the fact that a variation of the plasticizer content affects the stiffness in the same way as a temperature change; the $G(t)$ curve shifts horizontally along the time axis.

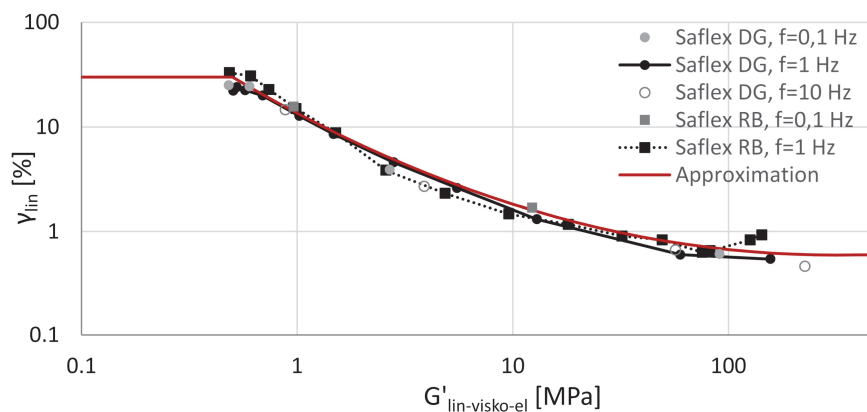


Figure 3-6 Linearity limits Saflex DG in dependency of storage modulus;

Approximation: $G'_{lin-visko-el} \leq 0.5$ MPa: $\gamma_{lin} = 30$ % $G'_{lin-visko-el} \geq 252$ MPa: $\gamma_{lin} = 0.6$ %

0.5 MPa $< G'_{lin-visko-el} < 252$ MPa: $\log(\gamma_{lin}) = 2,1834 \cdot 10^{-1}x^2 - 1,0879x + 1,1308$ with $x = \log(G'_{lin-visko-el})$.

4 Summary and Outlook

The linear viscoelastic stress-strain behaviour of a standard PVB as used in glass construction was shown as an example for the material Saflex® RB and the corresponding Prony-Series was given. The process for determining the linearity limits was discussed and shown for the selected material. Comparing the two chemically similar materials Saflex® RB and Saflex® DG it was found that their linearity limits behave analogously. The extent to which non-linear visco-elastic material behaviour occurs in material characterization tests and in common engineering applications should be checked before application, e.g. with numerical investigations.

After glass fracture, the linearity limit is reached locally in many areas of the foil, see further e.g. Delincé, et al. [36]. The film locally delaminates from the glass panes and undergoes large deformations in areas between two or more fragments.

There are different model approaches in order to combine the linear and non-linear viscoelastic properties in one model and to use them in numerical investigations, for example to predict the post-fracture behaviour of laminated safety glass. Often the material model of Schapery is used [15], who derives a constitutive model for non-linear viscoelastic materials in Schapery [37]. The linear viscoelastic constitutive model, which also underlies the Prony-Series, is extended by four strain dependent nonlinearity factors $h_\infty(\varepsilon)$, $h_1(\varepsilon)$, $h_2(\varepsilon)$, $a_\varepsilon(\varepsilon)$.

$$\sigma(t, \varepsilon) = h_\infty E_\infty \varepsilon(t) H(t) + h_1 \int_0^t \tilde{E}(\psi - \psi') \left\{ \frac{d[h_2 \cdot \varepsilon(\tau) H(\tau)]}{d\tau} \right\} d\tau \quad (4.1)$$

With:

$$\psi = \int_0^t \frac{dt}{a_\varepsilon(t)}$$

And

$$\psi' = \int_0^\tau \frac{d\tau}{a_\varepsilon(\tau)}$$

In equation (4.1) E_∞ is the equilibrium modulus, $\tilde{E}(t)$ is the linear shear modulus at a given time t and $H(t)$ is the Heavyside function at a given time. For all nonlinearity factors equal to one, the constitutive law for linear viscoelasticity results.

If one looks at the individual effects of the nonlinearity functions $h_\infty(\varepsilon)$, $h_1(\varepsilon)$, $h_2(\varepsilon)$, $a_\varepsilon(\varepsilon)$ on the curve of the elastic modulus as a function of the load duration (as shown in Figure 3-2c), one can see that the effect of the function $a_\varepsilon(\varepsilon)$ is similar to the effect of the temperature displacement factor a_T in equation (2.1). These displacement factors cause a change in the relaxation times and thus a horizontal displacement of the curve in Figure 3-2c. However, the factors h_0 , h_1 , h_2 have a vertical effect on the curve in Figure 3-2c.

Schapery [38] used creep and creep recovery tests to determine the parameters, using test results in the linear viscoelastic range to determine the linear model parameters and test results in the non-linear viscoelastic range to determine the non-linear model parameters. Hiel followed this procedure in [39]. Golden, et al. [40] used DMTA measurement data

in the linear and non-linear range to determine the model parameters. The use of one model for both the linear and the nonlinear viscoelastic range seems to be physically reasonable as well as practical and easy to apply, since the same Prony parameters can be used. In further investigations of the nonlinear viscoelastic behaviour of the interlayers of laminated safety glass this approach shall be pursued.

5 Acknowledgement

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