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Thermomechanical properties of unidirectional composites in their transition region

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With 6 figures and 1 table

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1. Introduction

The importance of polymeric composites as structural materials is steadily growing. Polymers used as matrix materials exhibit behaviour highly dependent on temperature, which can be controlled, apart from modifiers (1), by means of fillers in disperse or continuous form. The introduction of fillers improves not only the mechanical parameters of the material but also its structural stability at elevated temperatures by decreasing thermal expansion and increasing in general glass transition temperature. The glassy region is structurally important for the material, in which it behaves almost elastically and possesses satisfactory mechanical properties. The second or glass or β -transition temperature T_g is the upper limit of the glassy region and as such is of particular interest.

Between the glassy and rubbery regions and about T_g there is a whole transition zone, in which the material behaves viscoelastically, exhibiting time-dependent properties and high energy dissipation under dynamic loading. T_g can be determined by means of mechanical tests, e. g. at T_g creep compliance curves acquire maximum slope and loss tangent its peak value, or from dilatometric tests, as at T_g the thermal expansion curve shows a break corresponding to the change of the rate of variation of volume as temperature increases. In many cases a γ -transition at much lower temperatures appears (1, 2) corresponding to the mobility of different branches of the macromolecule and at which the mechanical behaviour of the material resembles that of the β -transition.

Methods of reduced variables (3), also applicable with composites on many occasions (4, 5),

may lead to the definition of respective t_g or ω_g from a composite curve on a time or frequency scale. T_g is a most sensitive parameter towards every aspect of the material microstructure or any factor relating to external conditions or loading mode. For example, with epoxy matrices are of importance: The molecular structure of the prepolymer (2), the molecular structure of the curing agent (6), amount of curing agent in relation to stoichiometric (7) or composition of curing agent consisting of more than one reactives (8) and the heat treatment, i. e. postcuring time and temperature. At stoichiometric amounts of curing agent, polymerization reactions end within relatively short time and a stable T_g is reached, otherwise it depends strongly on the duration of curing at a given temperature. One case is reported (7), where as a result of heat treatment α and β -peaks merged into one large peak. 24 hr. postcuring at stoichiometric composition at various temperature levels caused the transition temperature to increase almost in proportion to temperature (9). Moreover, mechanical imperfections such as voids, flaws or microcracks as well as load history are expected to influence T_g considerably.

With polymer composites, all of the numerous parameters characterizing their microstructure are expected to affect T_g . The introduction of the filler particles affects the mobility of the macromolecule chains in a way depending on many factors. The size and form of the filler particles and their volume fraction in the composite, quality of adhesion etc. are only a few (10, 11, 12). The combined action of all these factors may either increase T_g , which is expected as result of reinforcement, or decrease it, as it occurs with poor adhesion. Finally, residual or shrinkage

stress fields, tending to relax upon approaching T_g in a time-dependent manner, or excessive stress levels applied at cycles, thus inflicting microchanges in the composite (13), affect T_g .

Unidirectional laminates is a convenient composite form for structural applications. They usually consist of long continuous fibres in a polymeric matrix at high volume concentrations to achieve maximum reinforcement. Above a certain concentration, however, mechanical imperfections are expected to develop within the material with aggravating results on mechanical improvement.

An enormous number of publications on unidirectional composites have been produced over the last years, which need not be mentioned here. In particular, their thermomechanical behaviour has been investigated in a number of papers (14–21). The effect of reinforcement on T_g appears also in works dealing with the dynamic mechanical properties of unidirectional composites over wide temperature range. For example in (22) an extensive experimental study was carried out on an epoxy matrix in which various fillers, such as different types of glass microspheres and fibres were embedded. It was found that, on all occasions, T_g was increasing with the amount of filler, at least up to a certain extent. Similar behaviour was observed with rubber-carbon black systems (23), by which the rubber molecules are attached to the filler particles by strong forces approaching the nature of chemical bonds (24).

In the present, based on mechanical theories, a theoretical approach to the effect of reinforcement on T_g and also the effect of imperfect adhesion is attempted, as well as experimental investigation of a glass fibre unidirectional composite to verify theoretical predictions.

2. Theoretical study

In the following E , G , k are Young's, shear and bulk moduli, ν is Poisson's ratio, t is time, T temperature, ω frequency, θ angle subtended with the direction of fibres or axial direction, α thermal expansion coefficient (in particular α_1 for $T < T_g$ and α_2 for $T_g > T$), v is volume fraction, superscript * means complex modulus, dashed and double-dashed symbols mean storage and loss modulus respectively, $\eta = \tan\delta$ is loss factor, subscripts m , f mean matrix and fibre respectively, no subscript means composite, sub-

scripts a , t denote axial and transverse direction respectively and $i = (-1)^{\frac{1}{2}}$.

Let $R(\theta, T, t)$, or $R^*(\theta, T, i\omega)$, be a mechanical parameter or a thermal expansion coefficient of the composite, which, according to some theory is usually a function valid for the particular case, is usually of two corresponding parameters of the matrix and two of the filler material, both considered as isotropic and the volume fractions v_m , $v_f = 1 - v_m$ of the constituents. Suitable expressions can be derived from the respective elastic solutions by employing a correspondence principle.

As already stated, at $T = T_g$, $R(\theta, T, t)$ or its k -th derivate acquires a peak value. Hence T_g can be derived from the equation:

$$\left[\frac{\partial^{(k+1)}}{\partial T^{k+1}} R(\theta, T, t) \right]_{T=T_g} = 0$$

or, for constant θ and T , a transition time t_g or a transition frequency ω_g , from which the corresponding T_g can be determined.

In order to investigate the effect of reinforcement on T_g of the composite, one can first derive T_{gm} of the matrix material by putting:

$$\left[\frac{\partial^{(k+1)}}{\partial T^{k+1}} R_m(\theta, T, t) \right]_{T=T_{gm}} = 0$$

and then, by examining whether the quantity:

$$\left[\frac{\partial^{(k+1)}}{\partial T^{k+1}} R(\theta, T, t) \right]_{T=T_{gm}}$$

assumes negative, positive or zero value, can see which of the three relations $T_g > T_{gm}$, $T_g < T_{gm}$, $T_g = T_{gm}$ holds.

2.1 Dynamic mechanical properties

The complex shear modulus $G_a^*(i\omega)$ of a unidirectional composite in the axial direction has at constant temperature the following components (25):

$$G_a'(\omega) = G_m'(\omega) \frac{v_m + (1 + v_f)\gamma}{v_m \gamma + 1 + v_f} \quad [1a]$$

$$G_a''(\omega) = G_m''(\omega) \frac{v_m[(\gamma + 1)^2 + v_f(\gamma - 1)^2]}{[v_m \gamma + 1 + v_f]^2} \quad [1b]$$

where $\gamma = E_f/E_m'(\omega)$.

These expressions have been derived by application of the correspondence principle of

linear dynamic viscoelasticity from the respective elastic expressions (26). The latter are rigorous for the effective elastic moduli developed on the basis of the composite cylinder assemblage model. From equations [1a], [1b] one obtains for the loss factor $\eta_G(\omega)$:

$$\eta_G(\omega) = \eta_{G_m}(\omega) \times \frac{v_m[(\gamma + 1)^2 + v_f(\gamma - 1)^2]}{[v_m + (1 + v_f)\gamma][v_m\gamma + 1 + v_f]} \quad [2]$$

Now, the loss factor is a very convenient parameter for the determination of T_g as it corresponds to the centre of the transition region and expresses the amount of energy dissipated as fraction of the energy stored in the system. Although it is not susceptible to direct molecular interpretation, is preferable to loss modulus which is a measure of energy dissipated when different systems are compared at the same strain amplitude and its peak value corresponds to lower frequencies than loss factor (or higher temperatures). At the transition frequency ω_g the loss factor assumes its maximum value, hence $\partial\eta/\partial\omega = 0$. For the matrix, ω_{gm} can be determined from the equation:

$$\left[\frac{\partial\eta_{G_m}(\omega)}{\partial\omega} \right]_{\omega=\omega_{gm}} = 0 \quad [3]$$

For this value, the derivative of $\eta_G(\omega)$ is determined from equation [2]:

$$\frac{\partial\eta_G(\omega_{gm})}{\partial\omega} = -4v_f(1 + v_f) \left[\gamma^2(\gamma^2 - 1) \frac{\eta_{G_m}(\omega)}{G'_m(\omega)} \frac{\partial G'_m(\omega)}{\partial\omega} \right]_{\omega=\omega_{gm}} \quad [4]$$

As $\partial G'_m(\omega)/\partial\omega > 0$, it is obvious that $\partial\eta_G(\omega_{gm})/\partial\omega < 0$; hence at $\omega = \omega_{gm}$, $\eta_G(\omega)$ has already gone through its peak value (fig. 1), i.e. for the composite it is $\omega_g < \omega_{gm}$ or

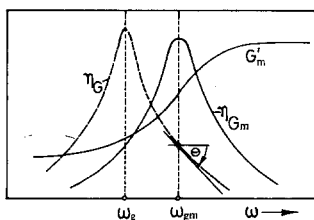


Fig. 1. Typical trends of shear moduli and loss factors for the matrix and the composite in the transition regions.

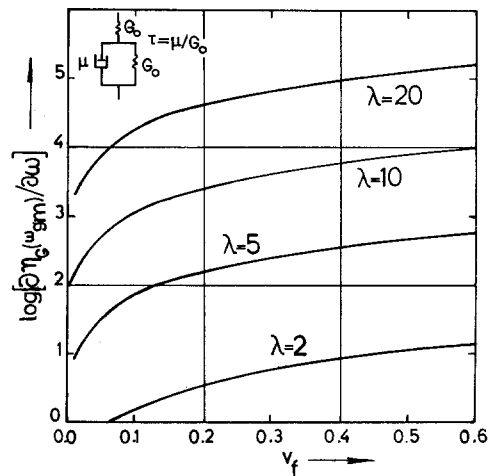


Fig. 2. Slope variation for composite shear loss factor with volume fraction of filler for various stiffness ratios of the constituents and for a matrix following a three-element linear viscoelastic model.

$T_g > T_{gm}$. Therefore, the introduction of the filler caused T_g to increase. It is interesting to see from equation [4] how $T_g - T_{gm}$ is affected: it increases with v_f and γ (while it vanishes at $v_f = 0$ or $\gamma = 1$) as well as with $\eta_{G_m}(\omega_{gm})$ and the slope $\partial G'_m(\omega_{gm})/\partial\omega$, while it decreases with increasing $G'_m(\omega_{gm})$. Hence, the slope $\partial\eta_G(\omega_{gm})/\partial\omega$ is increasing with the reinforcing effect (high v_f and low-modulus dissipative matrix).

Assuming $G^*(i\omega)$ to obey a three-parameter solid model (two identical springs and a dashpot, relaxation time τ), one has:

$$G'_m(\omega\tau) = G_0 \frac{1 + \omega^2\tau^2}{2 + \omega^2\tau^2} \quad [5]$$

$$\eta_{G_m}(\omega\tau) = \frac{\omega\tau}{2 + \omega^2\tau^2}$$

$\partial\eta_{G_m}(\omega\tau)/\partial\omega\tau$ vanishes at $\omega\tau = \sqrt{2}$ at which $\eta_{G_m}(\sqrt{2}) = \sqrt{2}/4$, $G'_m(\sqrt{2}) = 3G_0/4$ and in addition $\partial G'_m(\omega\tau)/\partial\omega\tau = \sqrt{2}G_0/8$, $\gamma = 4G_f/3G_0$. The expression holds:

$$\frac{\partial\eta_G(\omega_{gm})}{\partial\omega} = -0.5926v_f(1 + v_f)\lambda^2(1.7778\lambda^2 - 1) \quad [6]$$

which is plotted against v_f for various $\lambda = G_f/G_0$ in fig. 2.

In the transverse direction much more complicated expressions appear. With very stiff fibres ($\gamma \rightarrow \infty$) the loss factor assumes the following form (25):

$$\eta_{Gt} = \eta_{Gm} \left[1 - \frac{4}{3} (1 + \nu'_m) (1 - 2\nu'_m) \beta^2 \nu_m^3 \nu_f \times \frac{1 - \nu_f^3 + 3\nu_f \nu_m \beta (\beta + 2)}{(1 - \nu_f^2 - 3\nu_f \nu_m \beta^2) (1 - \nu_f^3) (1 + \beta \nu_f) - 3\nu_f \nu_m^2 \beta^2} \right] \quad [7]$$

where $\beta = 1/(3 - 4\nu'_m)$.

Although the algebra necessary to follow the previous procedure is very tedious, it can be shown eventually that $T'_g > T_{gm}$ as before. Moreover, the shear modulus in the θ -direction will be governed by expression derived by transformation from equations of the form:

$$\frac{1}{G_\theta} = A \sin^2 \theta \cos^2 \theta + B (\cos^2 \theta - \sin^2 \theta)^2 \quad [8]$$

where A, B are functions of the 4 elastic parameters required for the complete characterization of the orthotropic material. Equation [8] actually provides a suitable combination of the axial and transverse properties, and neither physical nor analytical reasons can justify conclusions different than the previously arrived at.

It is worth noticing that expressions other than rigorous, as the well-known law of mixtures:

$$E_a = \nu_m E_m + \nu_f E_m \quad [9]$$

lead to the opposite results. However, the complete form:

$$E_a = \nu_m E_m + \nu_f E_f + \frac{4\nu_m \nu_f (\nu_f - \nu_m)^2}{\nu_m / k_f + \nu_f / k_m + 1/G_m} \quad [10]$$

leads again to $T'_g > T_{gm}$.

It should be noted that, apart from algebraic difficulties, the method is operative if, (a) the viscoelastic functions of the matrix material are completely defined within the time (or frequency) and temperature domains considered, (b) the theory providing the mechanical parameters of the composite has been checked and found reliable within the above domains. The latter is not always a simple task, and correspondence principles are only valid within limited time and temperature ranges. For example, it is doubtful whether the previous analysis is applicable in the γ -transition without modifications. In general, γ -transition occurs at low temperatures corresponding to high

frequencies, at which the wave length of the applied vibration would be comparable with the average size of the filler particles. In this case, theories accounting for the complex phenomena thus produced should be considered (28).

2.2 Dilatometric considerations

At T'_g the coefficient of thermal expansion of the polymeric matrix undergoes a rapid change from α_1 to $\alpha_2 > \alpha_1$. With dilatometric tests, this actually occurs within a relatively narrow temperature zone having T'_g in the middle. The derivate $da_m(T)/dT$ is maximum at T'_g , hence $d^2a_m(T)/dT^2$ vanishes. Therefore, the condition:

$$\left[\frac{d^2 \alpha_m(T)}{dT} \right]_{T=T'_{gm}} = 0$$

leads to the determination of T_{gm} . An ordinary derivate has been used to denote the assumption that α is function of temperature only. This assumption is in general not true for the composite, where the thermal stress fields generated by a temperature rise tend to relax following the time-dependent patterns of the respective mechanical moduli of the matrix. Therefore, the thermal expansivities are also time-dependent, and the respective condition would be:

$$\left[\frac{\partial^2 \alpha(\theta, T, t)}{\partial T^2} \right]_{T=T'_g} = 0.$$

The expressions for the thermal expansion coefficient of a viscoelastic composite, based on the elastic formulae given in (17), are developed in (27) by replacing elastic constants with respective time operators. The extreme complexity of these results renders an analytical approach totally inoperative. On the other hand, simplifications of the form:

$$\alpha(\theta, T, t) \equiv \alpha(\theta, T)$$

implying adoption of a sort of 'isochronous' moduli, are unrealistic with a dilatometric test, by which a temperature rise at constant rate is applied on the specimen and the respective elongation is recorded. Even with simple rules of mixtures (15), fairly accurate on many occasions, the conclusions thus obtained are highly unreliable. Experimental evidence appears to be the best means in this case provided that all microstructural parameters of the composite are

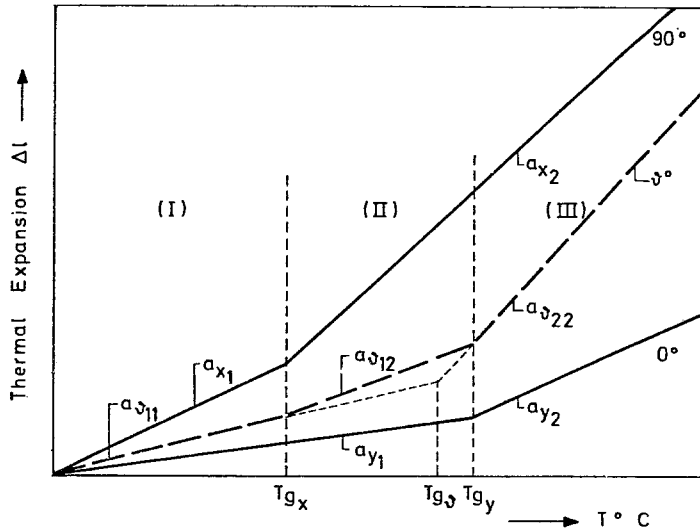


Fig. 3. Determination of glass transition temperature in the θ -direction from respective temperatures and thermal expansion coefficients along the orthotropic axes.

accurately controlled to ensure successful correlation with test results. Further investigation of this point appears necessary.

A final point is the determination of T_g in any direction in relation to the orthotropic axes. A sufficiently accurate approximation can be obtained in the following manner:

Assume that thermal expansion curves in the axial and the transverse directions are given (Fig. 3). The corresponding T_{gx} and T_{gy} divide the temperature domain in three regions. Following a well-known relation (21), the thermal expansion coefficient in the θ -direction assumes the form:

$$\left. \begin{aligned} \alpha_{\theta 11} &= \alpha_{x1} \cos^2 \theta + \alpha_{y1} \sin^2 \theta \quad (T < T_{gx}) \\ \alpha_{\theta 11} &= \alpha_{x2} \cos^2 \theta + \alpha_{y1} \sin^2 \theta \quad (T_{gx} < T < T_{gy}) \\ \alpha_{\theta 11} &= \alpha_{x2} \cos^2 \theta + \alpha_{y2} \sin^2 \theta \quad (T > T_{gy}) \end{aligned} \right\} [11]$$

corresponding to the broken line of Fig. 3. In a manner similar to the one applied to the dilatometric curve in order to determine T_g , the intersection point of the lines following $\alpha_{\theta 11}$ and $\alpha_{\theta 22}$ is found, corresponding fairly close to $T_{g\theta}$. In terms of the above parameter the following expression results:

$$T_{g\theta} = \frac{(\alpha_{\theta 12} - \alpha_{\theta 11})T_{gx} + (\alpha_{\theta 22} - \alpha_{\theta 12})T_{gy}}{(\alpha_{\theta 22} - \alpha_{\theta 11})} [12]$$

3. Experimental evidence

As stated, sufficient experimental evidence exists in the literature that T_g increases with reinforcement. However, at high volume fractions of filler opposite evidence can be found (3, 29), most probably due to imperfect adhesion, which is likely to occur with highly-filled composites. A suitable material to check the latter, as well as the rest of the points investigated in the previous analysis, was selected in the form of a composite consisting of a diglycidyl ether of bisphenol A epoxy matrix cured with an aromatic amine*) and long unidirectional E-glass fibres with 0.012 mm diameter at about 65% volume fraction. On the orthotropic characteristics and creep behaviour of such a material can be found in (30, 31). All specimens were cut from sheets 300 × 300 × 6.5 mm. The main physical properties of the constituent materials and the composite appear in table 1.

Apart from routine tests to determine mechanical moduli etc. at room temperature, the following tests were performed:

(a) Dilatometric tests on a Du Pont 990 thermomechanical analyzer (TMA) to determine

*) Araldite MY750 and HT972 hardener respectively (Ciba-Geigy Ltd.).

Table 1. Physical properties of the materials

Property	Matrix material	Fibre material	Composite	
			$\theta = 0^\circ$	$\theta = 90^\circ$
Young's modulus (GN/m ² *)	3.0	69.9	48.4	15.0
Poisson's ratio*)	0.35	0.20	0.125	0.285
Density (g/cm ³)	1.20	2.55	2.06	2.06
Thermal expansion coefficients (°C ⁻¹ × 10 ⁻⁶):				
α_1	52.5	5.0	6.5	25.8
α_2	109.0	—	9.0	76.0

*) at 20 °C

thermal expansion coefficients for the matrix and the composite up to 200 °C at angles equal to 0°, 15°, 30°, 45°, 60°, 75° and 90°.

(b) Dynamic mechanical tests for the same materials on a Rheovibron viscoelastometer

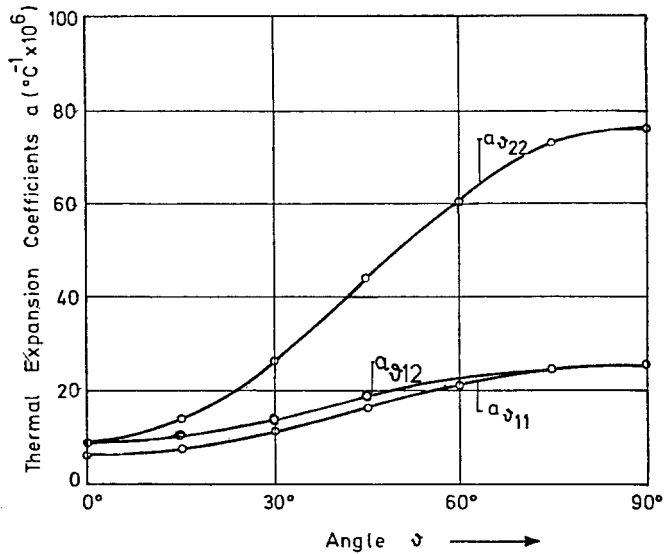


Fig. 4. Variation of thermal expansion coefficients with direction and experimental points.

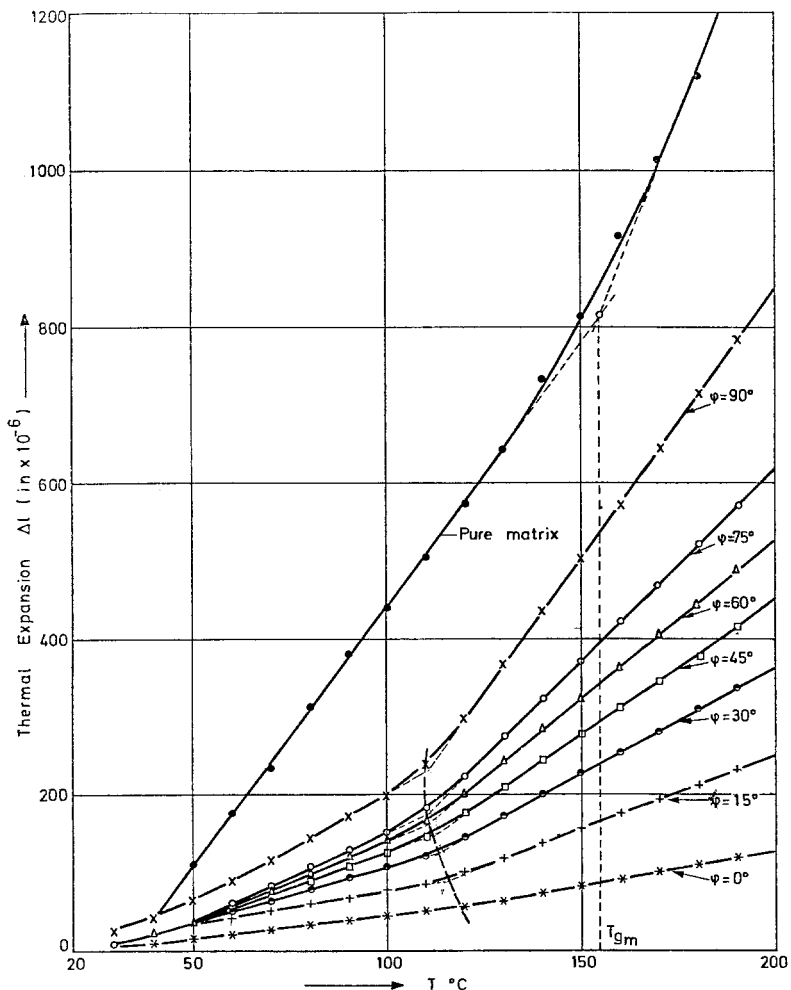


Fig. 5. Thermal expansion curves for the matrix and the composite at various directions.

employing forced extensional vibrations over wide temperature range.

(c) Dynamic mechanical tests on a Brüel and Kjaer Complex Modulus Apparatus employing flexural vibrations at resonance.

Part of the respective results are contained in (32).

4. Experimental results and discussion

As expected, thermal expansion coefficients agreed perfectly with the values predicted by equations [11]. Experimental values appear in Fig. 4.

In Fig. 5 thermal expansions curves from TMA for the various materials are presented (curves *a*). $T_{gm} = 155^\circ\text{C}$ drops to $T_{ga} = 119^\circ\text{C}$ and $T_{gt} = 110^\circ\text{C}$. T_g assumes intermediate values as plotted in Fig. 6, where experimental values agree very closely with the predictions of equation [12]. It is important to notice that aromatic amines as curing agents produce T_g higher even than expected from theoretical predictions (2).

Results from tests are also presented in Fig. 6. Longitudinal vibrations at 110 Hz gave $T_{gm} = 185^\circ\text{C}$ dropping to $T_{ga} = 178^\circ\text{C}$ and $T_{gt} = 159^\circ\text{C}$ (curves *b*) respectively. Flexural vibration method, the second is a resonance values at 100 Hz: $T_{gm} = 165^\circ\text{C}$, $T_{ga} = 159^\circ\text{C}$ and $T_{gt} = 143^\circ\text{C}$ while at 1000 Hz $T_{ga} = 168^\circ\text{C}$ and $T_{gt} = 150^\circ\text{C}$. Therefore, a very strong dependence of T_g on the testing mode appears*). This should be expected for a parameter so sensitive against a great number of internal and external factors, as T_g is. Between the two methods of dynamic testing here applied, consid-

) An interesting aspect yields the relation $E^ = 2G^* \times (1 + \nu^*)$ for isotropic viscoelastic solids: At $\omega = \omega_g$, $\partial\eta_G/\partial\omega = 0$, and the slope of the extensional loss factor is equal to:

$$\left. \frac{\partial\eta_E}{\partial\omega} \right\}_{\omega=\omega_g} = \frac{1}{D^2} \left[\nu'' \frac{\partial\nu'}{\partial\omega} - (1 + \nu') \frac{\partial\nu''}{\partial\omega} \right] \times (1 + \eta_G^2) \Big|_{\omega=\omega_g} \neq 0$$

with $D = 1 + \nu' + \nu''\eta_G$. Now, $\nu'' = E'(\eta_G - \eta_E)/2G'(1 + \eta_G^2)$ and it is always $\eta_G > \eta_E$ according to (33), but in practice $\eta_E = \eta_G$, as their difference is usually smaller than the experimental error, hence $\nu'' = 0$ and the above expression vanishes. However, it proves that ω_g or alternatively T_g are different in shear and in tension.

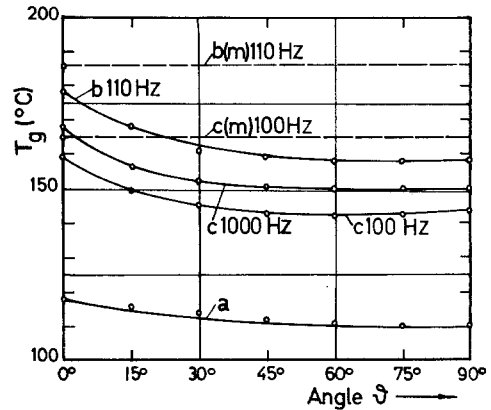


Fig. 6. Experimental values for glass transition temperature plotted against temperature: (a) from dilatometric tests with a pattern predicted by equation [12], (b) from dynamic extensional tests at 110 Hz and, (c) from dynamic flexural tests at 100 Hz and 1000 Hz.

erable differences exist. The first is a forced vibration method, the second is a resonance one utilizing results from higher harmonics to obtain values over extended frequency range. Specimen dimensions, levels of applied strain, air resistance, temperature control etc. are quite different with each method. And of course, both methods differ vastly from the dilatometric one, which is based on a completely different principle.

However, the general trends, as determined in the preceding analysis, appear to be verified. In particular, the value of T_{ga} is higher than the value of T_{gt} as the reinforcing effect is stronger. Moreover, T_g is a function of frequency, e.g. increasing with increasing frequency. An explanation for the low T_g -values obtained by 'static' tests, as dilatometric tests are, can certainly be found partly to the effect of frequency.

5. Review of work and conclusions

The mechanical properties of unidirectional composites in their glass transition region were investigated. In particular, the value of the glass transition temperature, which constitutes an upper limit for the structurally important glassy region, was examined. According to experimental evidence existing in the literature, the introduction of a reinforcing filler in a polymeric matrix causes T_g of the latter to increase, unless mechanical imperfections counterbalance the reinforcing effect or even produce T_g for the composite lower than that of the matrix. Based on mechanical theories for the mechanical modu-

li of unidirectional viscoelastic composites, it was proved that T_g increase with the effect of reinforcement, namely with filler-to-matrix stiffness ratio, filler volume fraction, loss factor of matrix and slope of modulus on a frequency scale. Hence, T_g was found lower in the transverse direction than in the axial, and similar results were found to hold for thermal expansion coefficients, although a completely thermo-mechanical analysis for this latter case is extremely complicated. In directions other than the orthotropic axes, intermediate values are expected for T_g according to the laws holding for the particular case. With dilatometric tests, an approximate method was developed, giving T_g in any direction as function of the corresponding T_g 's and thermal expansion coefficients along the orthotropic axes. The validity of the method was confirmed experimentally.

Experimental evidence was provided by means of a highly-filled unidirectional composite consisting of an epoxy matrix and glass fibres. Extensional and flexural dynamic tests as well as dilatometric tests were carried out over wide temperature ranges. As expected, imperfect adhesion between the constituent materials, usually occurring with highly-filled composites, gave reduced T_g 's in relation to the matrix, but the various methods produced vastly deviating results, confirming thus the extreme sensitivity of T_g towards any internal or external factor. It was also confirmed that T_g is increasing with the reinforcing effect, remaining on all occasions lower in the transverse direction, and with frequency.

Summary

The thermomechanical behaviour of polymeric matrices reinforced with long unidirectional fibres in their glass transition region was investigated. Based on rigorous theories for the mechanical moduli and the coefficients of thermal expansion for viscoelastic composites, it was proved analytically that the introduction of reinforcing fillers in polymeric matrices causes glass transition temperature to increase to an extent proportional to the reinforcing effect. This is confirmed by numerous experimental results available in the literature, except for highly-filled composites, where imperfect adhesion may counterbalance the reinforcing effect. Dilatometric as well as two different dynamic mechanical tests confirmed the latter. The variation of glass transition temperature as function of the various aspects of mechanical reinforcement was investigated.

Zusammenfassung

Das thermomechanische Verhalten von mit einachsig verstreckten Faserstücken verstärktem Polymermaterial im

Glasübergangsbereich ist Gegenstand der hier diskutierten Untersuchungen. Basierend auf strengen Theorien für die mechanischen Moduln und die Koeffizienten der thermischen Ausdehnung für viskoelastische Mischsubstanzen wurde analysiert, wie eine Einführung des Füllmaterials in die polymere Matrix zu einem Anstieg der Glastemperatur proportional zur Verstärkerwirkung des Füllers bewirkt. Das wird durch ein ausgiebiges experimentelles Material aus der Literatur bestätigt. Nur bei hochgefüllten Mischungen stört unvollständige Adhäsion die Verstärkerwirkung. Dies läßt sich auch dilatometrisch sowie auf zwei verschiedene andere Weisen mit mechanischen Testen zeigen.

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