Dynamic mechanical properties of structural glass fibre–epoxy composites

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Dynamic mechanical measurements at a frequency of 1 Hz have been made from 100 to 480 K on unidirectionally aligned composites of glass fibres and epoxy resin of diglycidyl ether of bisphenol A and 4,4′diaminodiphenylmethane, prepared under two conditions: (i) by mechanically mixing without a solvent; and (ii) by mixing their solutions in methyl ethyl ketone. The composites show the same three relaxation regions as the pure epoxies, but their relative magnitudes differ. The glass transition temperature of the composite prepared by the first method is less than that of pure epoxy but of that prepared by the second method is greater. This implies a decrease in the crosslink density in the former and an increase in the latter case. The ratio, G_U/G_R of the composite prepared by the first method is half of that prepared by the second method. A coefficient, $[(G_U/G_R)_{comp}/(G_U/G_R)_{polymer}]$, is used to test the various empirical equations for the reinforcement of polymers in the composites. Hashin's equation provides the most satisfactory description of the aligned fibre–epoxy composite.

(Keywords: dynamic mechanical relaxation; glass fibre–epoxy composites)

INTRODUCTION

The use of polymer composites in areas traditionally dominated by metals and ceramics has continued to grow at an accelerating rate. These now include the high-performance structural composites for use in aerospace, where design criteria require them to have specific mechanical characteristics. Obtained by mixing epoxies with a variety of lighter, but stronger, materials, the dynamic mechanical properties and static moduli of the composites can be controlled to meet the requirements of load-bearing structural applications. The mechanical behaviour of the composites depends both upon the temperature and the duration, or frequency, of deformation due to the applied load. We report such a behaviour of a composite of unidirectionally aligned glass fibres with an epoxy resin prepared by mixing the diglycidyl ether of bisphenol A (DGEBA) and diaminodiphenylmethane (DDM) in a stoichiometric ratio of 2:1.

Industrial-scale production of certain composites involves preparation of the epoxy from a solvent, one of which is methyl ethyl ketone, and it is assumed that the static and the dynamic mechanical properties of the composite remain unchanged when the solvent is used in its preparation. We include the results of a further study of the same composite prepared by mixing the resin in a solvent, and compare the behaviours of the two composites.

In addition to providing new results on fibre–epoxy composites over a wide temperature range, in which their use is intended, we also consider physical interpretation of the difference in the equilibrium modulus and relaxation features of both the epoxies and their composites prepared with and without the use of a solvent.

EXPERIMENTAL

Type D glass fibres of 10 μm diameter, which had a silicone coating of unspecified thickness, were obtained from Vetrotex, Saint Gobain, France. They were in the form of a bundle or lock with ≃2400 fibres per lock. They were wound around the three legs of an inverted tripod and were kept under slight tension to ensure straightness and desirably perfect alignment of fibres, and then impregnated with the epoxy resin.

The resin and the composites were made in Laboratoire des Matériaux Macromoléculaire, CNRS, ERA745 INSA 69261, Villeurbanne, France, by two different methods: In the first method, DGEBA and DDM, in a stoichiometric ratio of 2:1, were mixed for 15 min at 366 K. The tripod with the stretched fibres was immersed in the fluid resin to ensure complete impregnation of the fibre bundle with the epoxy. The procedure was repeated with different fibre-to-epoxy ratios. The resin was cured by keeping for 5 h at 363 K, and 3 h at 443 K, followed by cooling to ~298 K. This epoxy resin is referred to as epoxy (N) and the composite as composite (N).

In the second method, the epoxy resin was prepared by

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mixing a solution containing 1 g DDM per ml of methyl ethyl ketone with DGEBA at 353 K in a stoichiometric ratio of 2:1 of DGEBA to DDM. Several samples of the stretched fibres were impregnated in this solution and the solvent then removed by keeping under vacuum for 1 h at 358 K. The composite was then cured by keeping for 5 h at 363 K and for 3 h at 443 K followed by cooling to 298 K, as for the composite (N). In the second procedure, the solvent was evaporated from the resin under vacuum for 1 h at 358 K *prior to* impregnating the fibres in it. The composite was then cured for 3 h at 443 K and cooled to 298 K. No difference in the mechanical behaviour of the composites was observed whether the fibres were impregnated in the resin before or after the removal of methyl ethyl ketone from the resin. These composites are referred to as composites (S) and the epoxy resin used as epoxy (S).

Samples containing different weight ratios of fibre to epoxy were cut to a dimension of ~0.5 × 4 × 60 mm. The accurate composition of the composite was determined after each experiment.

The instrument used for measuring the real and imaginary components of the shear modulus has been described in earlier papers[1]. Essentially, it consisted of an inverted forced oscillation pendulum in which the lower end of the specimen was secured and the upper end was connected to a magnet which oscillated in a magnetic field of desired frequency. The assembly was counterbalanced and, therefore, the specimen was under a very slight tension. Several measurements were repeated for both the temperature and frequency variation and the results agreed to within 1%. The temperature of the sample was maintained constant to within ±0.2 K, thus ensuring that the errors from temperature variation were minimal. The relative amplitude of deformation was less than 10^{-4}.

RESULTS

The mechanical loss tangent, measured at 1 Hz, of composite (S) containing 35.1 and 44.9 vol% fibre is plotted against temperature in *Figure 1*. As in the pure epoxy resin, the composite (S) showed three relaxation peaks at 220, 325 and 430 K. These are referred to as γ, β and α-processes, respectively, in order of increasing temperature and do not imply any specific mechanisms of side-chain or segmental motions. The β-peak is slightly increased in height with increase in the fibre contents, but the α-peak which is attributed to glass–rubber transition is decreased in both its height and its temperature when the amount of glass fibre is increased from 35.1 to 44.9% by volume. These changes are less significant since their magnitude is marginally greater than the experimental uncertainty mentioned earlier. The changes are indeed small considering the ~10% increase in the fibre content.

The $\tan\phi$ at 1 Hz of composite (N) and of pure epoxy (N) is plotted against temperature in *Figure 2*. The γ-peak of the composite is narrower at the low-temperature side but its height and position (at 224 K) remains the same. The β-process in the composite is reduced to a magnitude too small to show up as a resolved peak at 329 K. The temperature of the α-peak is decreased from 453 K in the pure epoxy to 448 K in the 37.6 vol% composite and its height reduced from 0.76 to 0.47.

The corresponding plots of $\tan\phi$ at 1 Hz of epoxy (S) and 35.1 vol% composite (S) are shown in *Figure 3*. The height of the γ-peak remains the same but its temperature increases from 205 to 218 K. The height of the β-peak in the composite is decreased from 0.03 to 0.018 and its temperature increased from 308 to 325 K. The height of the α-peak is also decreased from 0.82 to 0.57 and its temperature raised from 420 to 430 K. The direction of the shift of the α-peak in composite (S) in *Figure 3* is opposite to that in composite (N) in *Figure 2*.

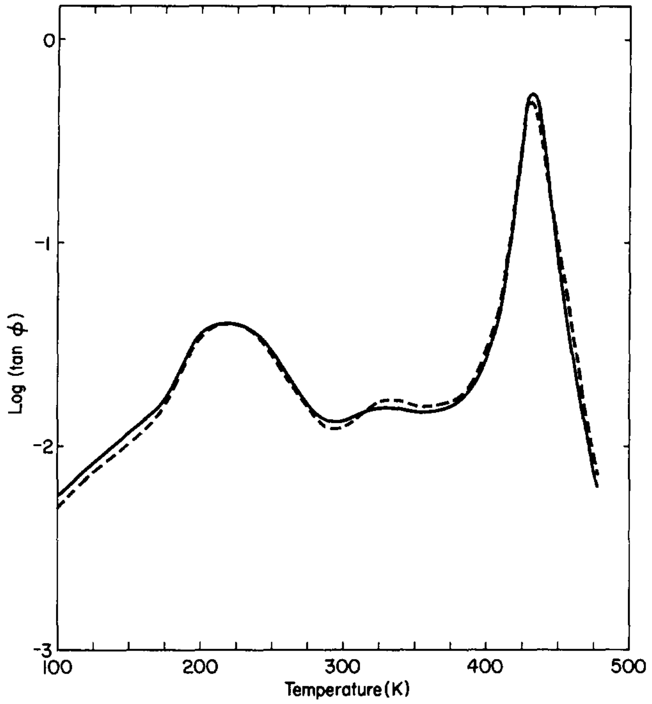

Figure 1 Mechanical loss tangent of glass-fibre–epoxy composite measured at 1 Hz is plotted against temperature. The epoxy was prepared by mixing the two components in methyl ethyl ketone. ——, Composite containing 35.1 vol% fibre; - - - -, 44.9 vol% fibre

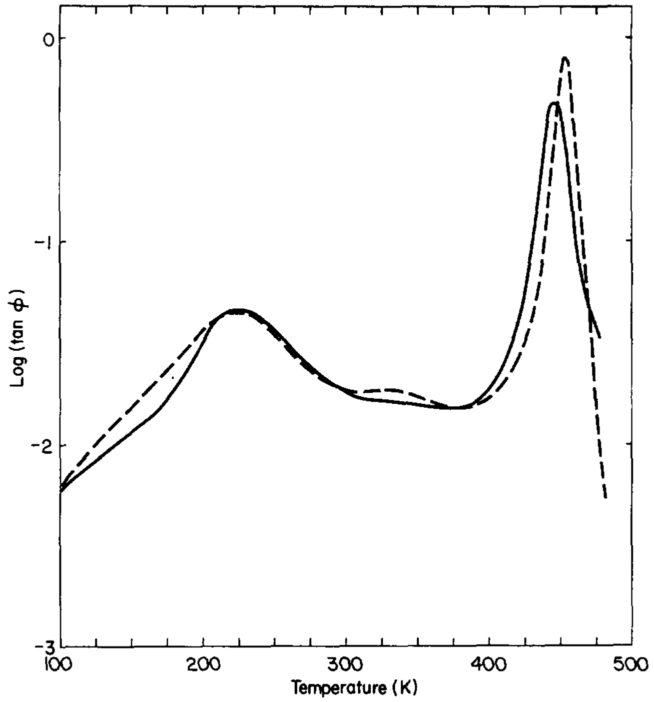

Figure 2 Mechanical loss tangent measured at 1 Hz of glass-fibre–epoxy composite and epoxy resin plotted against temperature. The resin was prepared without a solvent. ——, Composite containing 37.6 vol% fibres; - - - -, pure epoxy resin

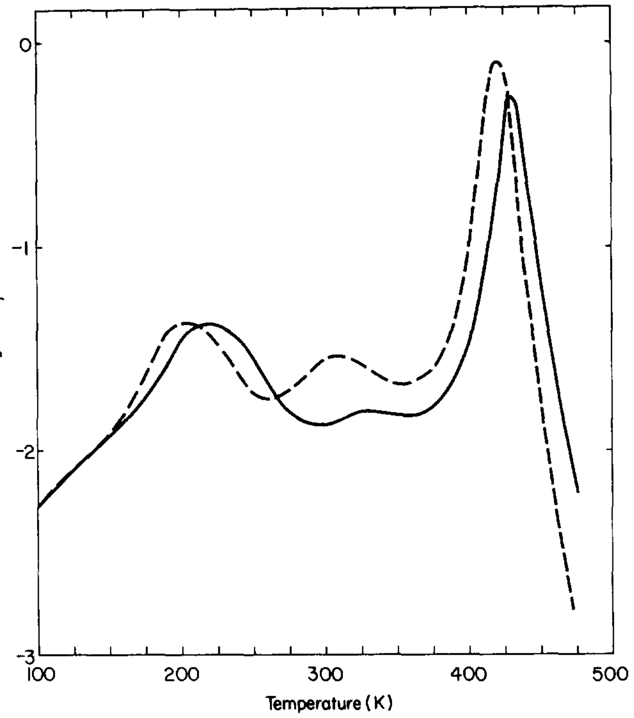

Figure 3 Mechanical loss tangent measured at 1 Hz of glass-fibre–epoxy composite and pure epoxy resin plotted against temperature. The resin was prepared by mixing the two components in a solvent. ——, Composite containing 35.1 vol% fibres; ----, pure epoxy resin

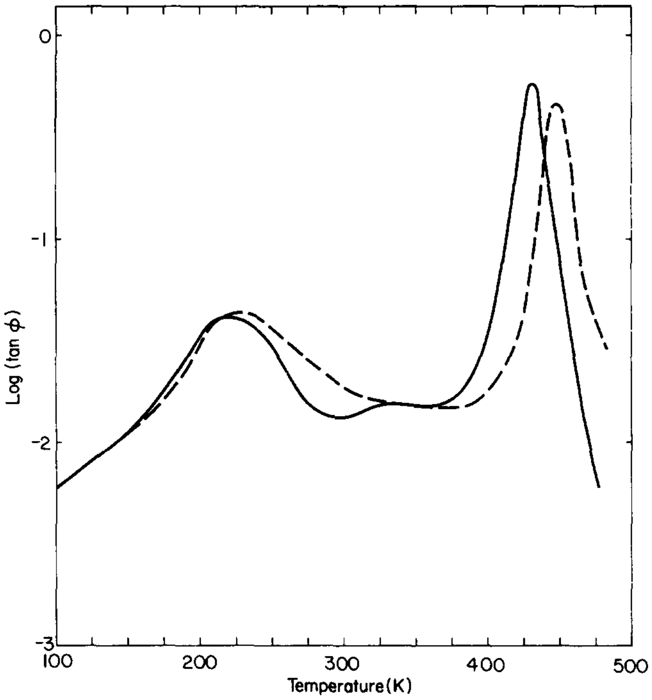

Figure 4 Mechanical loss tangent measured at 1 Hz of glass-fibre–epoxy composites plotted against temperature. ——, Composite containing 35.1 vol% fibre with epoxy obtained by mixing the components in a solvent; ----, composite containing 37.6 vol% fibre with epoxy obtained by mixing the components without a solvent

A comparison of the tan ϕ temperature behaviour of 35.1 vol% composite (S) with 37.6 vol% composite (N) is shown in *Figure 4*. In both cases the height and temperature of the γ-peak change, the β-peak remains unresolved in composite (N), but is clearly resolved in composite (S). The α-peak of composite (S) is 0.57, nearly 20% greater than of composite (N) and its temperature is ~18° lower than of composite (N).

G'' and J'' measured at 1 Hz of composites (S) and (N) and normalized to their respective values at 300 K are plotted in *Figure 5*. These show that the temperature of the peaks differs according to the parameters chosen to represent the dynamic mechanical behaviour of the epoxy composites. The recognition of such differences is important from both theoretical and applications points of view.

The real component of the shear modulus G' at a given temperature was divided by its value at 300 K, and the normalized value, i.e. $G'(T)/G'$ (300 K) for pure fibres, epoxy (N), epoxy (S), composite (N) containing 32.0 vol% fibre, and composite (S) containing 35.1 vol% fibre, is plotted against temperature in *Figure 6*. These show the shift in the temperature of, and the difference between, the relaxed and unrelaxed moduli, G_R and G_U, respectively, near the glass transition temperature. The relaxed modulus of the composites is nearly twice that of the pure epoxies whether prepared with or without the solvent.

Part of our results agree with those obtained by Babayevski and Gillham[2] from a study of the kinetics of curing of the epoxy–braid composite. In particular, the temperature of the α-peak is about the same (i.e. 453 K), but the temperature of both the β- and the γ-peaks are lower and the relative height of the γ-peak to both β- and α-peaks much higher than in *Figure 2* here. The discrepancy between our and their results may be due to several factors, namely, (i) the distribution of braided fibres in their composites, which is different from our unidirectional distribution, (ii) the experimental methods,

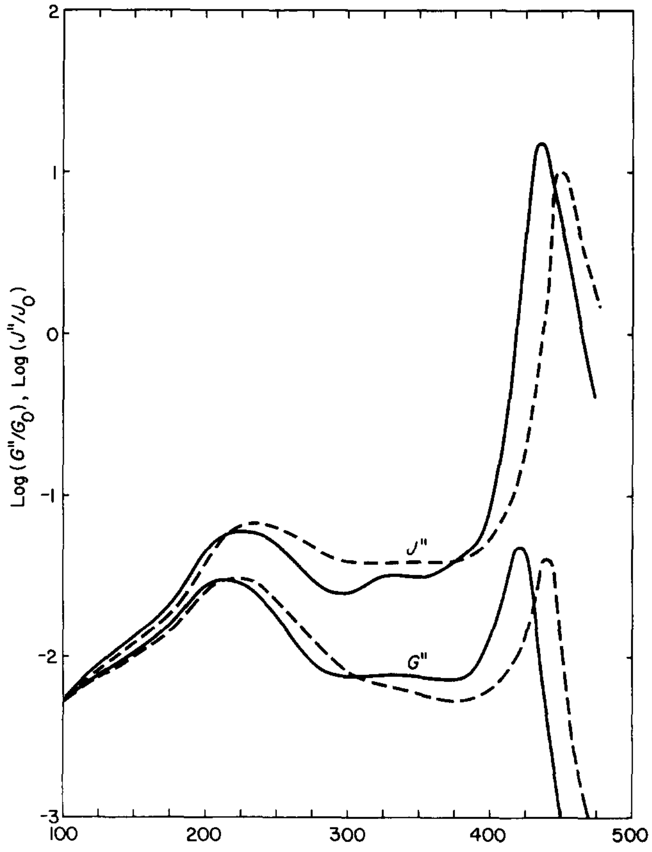

Figure 5 Normalized plots of G'' and J'' measured at 1 Hz of the two composites plotted against temperatures. The composites are the same as in *Figure 4*. G_0 and J_0 are the respective values of G'' and J'' measured at 1 Hz and 300 K

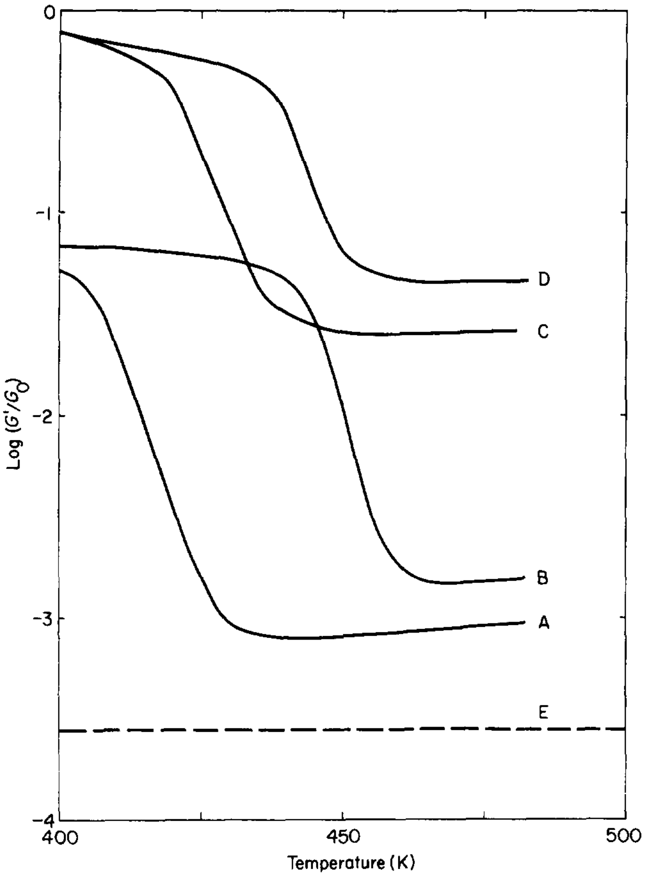

Figure 6 Plots of the shear modulus measured at 1 Hz and normalized by its value at 300 K, i.e. G'/G' (300 K) for: A, pure epoxy prepared by dissolving the components in a solvent; B, pure epoxy prepared without a solvent; C, composite prepared from a solvent containing 35.1 vol% fibres; D, composite prepared without a solvent, containing 32.0 vol% fibres; E, glass fibres alone in the form of a bundle. Plots for the epoxies, i.e. A and B, have been displaced downwards by one logarithmic cycle. $G_0 = G'$ (300 K) of the same substance except for glass fibres for which $G_0 = 30$ GPa

(iii) relatively large strains used in their torsional braid analyses and (iv) the different type of coatings on fibres. It is difficult to ascertain whether the difference is due to one or all of these factors.

DISCUSSION

The dynamic mechanical behaviour of a fibre–epoxy composite is mainly determined by the degree of adhesion of the polymer to the surface of the inorganic filler. A greater degree of adhesion of aligned fibres generally increases the modulus of the composite. While good adhesion is desirable for obtaining a high strength in a composite; it may be undesirable if high toughness is wanted. The dynamic mechanical behaviour of the composites also depends upon the difference between the coefficients of thermal expansion of the fibre and the epoxy, which can be considerable at temperatures close to T_g, and can cause residual stresses in the composite. Thus both the degree of adhesion and the residual stresses in a composite need to be considered.

The changes in the relaxation characteristics of composite (S) with the amount of glass fibres seen in *Figure 1*, though less significant, are also too small to be associated with a change in the degree of adhesion of the epoxy to the glass fibre. Since thermal cycling did not alter the position of the peaks in *Figure 1*, the magnitude of residual stresses, if present in the composite, was also insufficient to significantly affect its dynamic mechanical behaviour. Therefore, we ignore the changes in the relaxation characteristics with the amount of glass fibre and discuss the features of the composites in terms of the degree of crosslinking in the epoxies in them.

Effect of the fibre on the crosslinking in composites

In *Figure 3*, the height of both the α- and β-peaks decrease and their temperatures increase when epoxy (S) is used to form composite (S). Increased crosslinking raises the glass transition temperature[3–6] and is expected to reduce the number of loosely packed regions where segmental motions in the amorphous matrix are observed as the γ- and β-relaxations. This interpretation implies that composite (S) has a higher crosslink density than epoxy (S). Although part of the decrease in the heights of the α- and β-peaks is associated with the reduced volume fraction of the polymer in the composite, the greater decrease in the height of the β-peak relative to the α-peak is a further evidence for the greater crosslink density in composite (S) than in epoxy (S).

Measurements of composite (N) were not made as a function of its fibre content, but a comparison of the mechanical behaviour between epoxy (N) and the composite (N) in *Figure 2* shows an opposite trend, namely that both the height and temperature of the α-peak are lower in the composite than in the epoxy, the γ-peak remains at the same temperature and height, and the β-peak is either reduced in height or is completely removed. It means that the presence of the fibre–epoxy interface decreased the crosslink density in epoxy (N). It seems likely that the optimum network density reached on curing epoxy (N) is reduced by the presence of the interface. Part of the decrease in the heights of the α- and β-peaks is due to the reduced volume fraction of the polymer in the composite but the relative magnitude of the β-peak with respect to α-peak is greater in the composite (N) than in epoxy (N). This further suggests a decreased crosslinking in composite (N).

The glass transition temperature of a polymer is known to increase almost linearly with the crosslink density[3–6]. In a theory[7] that deals with the change in configurational entropy with temperature as being the dominant effect in the glass transition of polymers, the decrease in the configurational entropy caused by crosslinking may be related with the second-order transition temperature[8] at which the configurational entropy is expected to become zero. This transition temperature is raised by increased crosslinking according to the equation

$$\frac{T_2(x)-T_2(0)}{T_2(0)}=\frac{KMx/\gamma}{1-KMx/\gamma} \tag{1}$$

where x is the crosslink density, M is the molecular weight of a monomer, γ is the number of flexible bonds per monomer backbone and side chain. $T_2(x)$ and $T_2(0)$ are, respectively, the temperatures of the theoretical second-order phase transformation in a polymer with x numbers of crosslinks and zero crosslinks, respectively. The quantity K is a constant[8] of value $1.3 \pm 0.1 \times 10^{-23}$. Since T_2 is approximately equal to $0.8\,T_g$ (Ref. 7), this implies that any decrease in the temperature of the α-peak and, therefore, in T_g or T_2 of the pure epoxy resins is due to the decreased crosslink density.

Similar consideration also applies to the composites. Like other heterogeneous polymer blends, composites have two T_g values, corresponding to the two constituents. Since the T_g in such blends, or composites, does not differ from the T_g of the pure material, the change in the temperature of the α-peak observed in *Figure 4* is a reflection of a change in the crosslink density. Since T_g/T_α, the ratio of T_g to the temperature, T_α, at which the α-peak appears in measurements, at say 1 Hz, of an epoxy is likely to remain unchanged on crosslinking, $T_2(x)$ and $T_2(0)$ in Equation (1) can be replaced by $T_g(x)$ and $T_g(0)$ and, correspondingly, by $T_\alpha(x)$ and $T_\alpha(0)$, respectively, where $T_2(0)$, $T_g(0)$ and $T_\alpha(0)$ are, respectively, the theoretical second-order phase transformation temperature[7], the glass transition temperature, and the temperature of the α-peak of the uncrosslinked or gelled polymer measured at 1 Hz. $T_\alpha(0)$ for our epoxy measured by Babayevsky and Gillham[2] is $\simeq 383$ K. By substituting in Equation (1) the experimental value of $T_\alpha(x) = 430$ K for $T_2(x)$, the literature value of $T_\alpha(0) = 383$ K for $T_2(0)$, the calculated value of M/γ of 45, and of K of 1.3×10^{-23}, we obtain $x = 1.9 \times 10^{20}\ \mathrm{g}^{-1}$ for composite (S). By substituting the experimental value of $T_\alpha(x) = 448$ K for $T_2(x)$ and the same values of $T_2(0)$, M/γ and K as above, we obtain $x = 2.5 \times 10^{20}\ \mathrm{g}^{-1}$ for composite (N). The corresponding values for epoxy (S) and epoxy (N) are $1.6 \times 10^{20}\ \mathrm{g}^{-1}$ and $2.6 \times 10^{20}\ \mathrm{g}^{-1}$, respectively. Calculations show that the crosslink density of the epoxy in composite (S) is 17% higher than in pure epoxy (S), and that of the epoxy in composite (N) is 5% lower than in pure epoxy (N).

The relative magnitude of G_R in *Figure 6* for the pure epoxy is consistent with the above noted decrease in the crosslink density that occurred when the epoxy was prepared from the solvent, methyl ethyl ketone. A decrease in the G_R of epoxy (S) to nearly half the value of epoxy (N) seen in *Figure 6* corresponds to the same decrease in the M_e, i.e. the molecular weight between the fixed entanglements, according to the equation, $G \simeq \rho_e RT/M_e$, which is valid when the correction term from chain ends becomes negligible, as in the crosslinking of essentially linear molecules[3]. Such a consideration also, of course, applies to the two composites with the same volume % of fibre for which the ratio of their G_R is the same as the ratio of their crosslink density, x. But such a comparison between the pure epoxies and their composites cannot be made, since the value of G_R of a composite depends both upon the volume fraction, shape and distribution of the filler or fibre in the polymer matrix. However, a comparison that allows inclusion of some of these factors can be made as follows:

Equilibrium moduli of the composites

For a more appropriate comparison between the rubber moduli of the pure epoxies and their composites, a new sample of composite (N) containing 32.0 vol% glass fibre was made. Its dynamic mechanical behaviour was identical with that of the 37.6 vol% glass-fibre composite (N) seen in *Figure 2*, and, therefore, its tan ϕ-temperature plot is not included. The normalized value, $G = G'(T)/G'(300\ \mathrm{K})$, of the composite is plotted in *Figure 6*, from 400 to 480 K – a region of glass-rubber transition.

The relative magnitude of the glass to rubber moduli, G_U/G_R, can be obtained from *Figure 6* and these are used in our analysis. In *Figure 6*, the ratio, (G_U/G_R) at 470 K for 32 vol% composite (N) is 21, for 35.3 vol% composite (S) is 40. The ratio for epoxy (N) is 65 and for epoxy (S) is 112. For convenience of describing the effectiveness of the shape and volume of fillers on the moduli of the composites, we introduce a coefficient C, such as:

$$C = (G_U/G_R)_{\mathrm{comp}}/(G_U/G_R)_{\mathrm{poly}} \tag{2}$$

The value of C for composite (N) is 0.32 and composite (S) is 0.36. This means that the preparation from a solvent reduces the effectiveness of the composite (S) by causing its G_U to increase more than its G_R, when compared with the corresponding relative increase in G_U and G_R of composite (N).

The reinforcement in the shear modulus caused by a filler in a composite is usually described relative to the value of the pure polymer and is compared for a given volume fraction. The higher is this ratio, the lower is the effectiveness of the filler. This ratio, G_r, does not include the possibility that the amplitude of relaxation of the modulus, and the associated mechanical loss in the transition region, also depend upon the volume % of the filler. Since the coefficient C includes such effects, we would consider a comparison of C first amongst several composites and then between its experimental and theoretical values for the same composite.

Data on unidirectionally aligned fibre composites are not available to us, but Lewis and Nielsen[9] have provided accurate results on 41% glass spheres (diameter 10–20 μm) DGEBA (cured with triethylene tetramine) epoxy. From their data at 1 Hz, $G_U/G_R = 46$ for the composite and 73 for the pure epoxy, both at 300 K. Thus the value of C for their composite is 0.63 (they also observed that G_U of the composite was 2.65 times, and G_R 4.2 times, the respective G_U and G_R of the pure epoxy). The higher value of C for the sphere-filled epoxy than of the fibre-filled epoxy of our study suggest that, for the same volume of filler, the aligned glass-fibre composite is more strength-effective than glass-sphere composite.

There is a further point of interest which is related to the theoretical predictions and experimental results for the moduli of composites. This can be discussed in terms of the coefficient C, or in terms of the relative increase of G_U and G_R of the composites. For non-interacting rigid particles, the shear modulus seems to be independent of the particle size but increases with the volume fraction of the filler in accordance with several equations given in the literature. The first such empirical equation is[10,11]

$$G_{\mathrm{comp}}/G_{\mathrm{poly}} = [1 + 1.25\phi/(1 - \phi/\phi_m)]^2 \tag{3}$$

where the subscripts refer to the composite and the polymer, ϕ is the volume fraction of the filler and ϕ_m is the maximum volume fraction corresponding to close packing. Equation (3) implies that for a hard filler in a rubbery polymer, the G'-temperature plot of the composite is simply shifted by a fixed value above the corresponding plot of the polymer and, therefore, the composite and the polymer have the same value of G_U/G_R. Thus, the coefficient C is unity. Clearly Equation (3) is neither applicable to Lewis and Neilsen's results[9], as our calculation of C using their data give a value of 0.63, nor to our own results given here.

In a first approximation, the modulus of elasticity of a unidirectional glass fibre–epoxy composite with good adhesion between fibres and epoxy obey the rule of mixtures, and therefore, expressed as a series:

$$(G_{\mathrm{U,comp}}/G_{\mathrm{R,comp}}) = \frac{(\phi/G_{\mathrm{R,fibre}}) + (1 - \phi)/G_{\mathrm{R,polym}}}{(\phi/G_{\mathrm{U,fibre}}) + (1 - \phi)/G_{\mathrm{U,polym}}} \tag{4}$$

where G_U and G_R refer to the transverse shear modulus of the material indicated by the subscript and ϕ is the volume fraction of the fibres. From our studies here, $G_{\text{fibre}} = 20\,G_{\text{U,polym}} = 1300\,G_{\text{R,polym}}$. Therefore, the calculated value from Equation (4) of (G_U/G_R) is 64 for 32vol% composite (N) and 65 for the pure epoxy (N). Thus $C = 0.98$. This value is nearly three times the value of 0.32, experimentally found in our study. Evidently the series model is also inadequate for the composite. This is partly due to the difference between the crosslink density in pure epoxy and that in the composite, as discussed above, and partly due to the influence of surface forces on the interface layer structure in the composite.

Hashin[12] has also recently reviewed relations for the modulus of composites and it is useful to compare these relations for anisotropic composites with our experimental results. The expression given by Hashin[12], for the transverse shear modulus of the unidirectional fibre composite is:

$$G = G_{\text{poly}}\left\{1 + \frac{(1+\beta_1)\phi_2}{\rho - \phi_2[1 + 3\beta_1^2\phi_1^2/(\alpha\phi_2^3 + 1)]}\right\} \qquad (5)$$

where $\alpha = (\beta_1 - \gamma\beta_2)/(1 + \gamma\beta_2)$

$$\beta_1 = 1/(3 - 4v_1);\ \phi_2 = 1 - \phi_1$$

$$\beta_2 = k_2/(k_2 + 2G) \simeq 1/(3 - 4v_2)$$

$$\rho = (\gamma + \beta_1)/\gamma - 1$$

$$\gamma = G/G_{\text{polym}} = G_{\text{fibre}}/G_{\text{polym}}$$

k_2 is the transverse bulk modulus, v_1 and v_2 are the Poisson's ratios for the matrix and the fibres, respectively, ϕ_2 is the volume fraction of fibres and ϕ_1 of the matrix and subscripts 1 and 2 refer to the matrix and fibres, respectively. For our composites, $v_{\text{polym}} = 0.3$ and $G_{\text{polym}} = 1.5$ GPa in the unrelaxed, or glassy, state and $v_{\text{fibre}} = 0.2$, $G_{\text{fibre}} = 30$ GPa. $v_{\text{polym}} = 0.5$ and $G_{\text{R,polym}} = 23$ MPa for pure epoxy (N) and 13 MPa for epoxy (S). Thus from Equation (5), we calculate, $G_{\text{U,comp}}/G_{\text{U,polym}} = 1.80$ for epoxy (N) and 1.93 for epoxy (S), and $(G_{\text{R,comp}}/G_{\text{R,polym}}) = 3.89$ for epoxy (N) and 4.75 for epoxy (S). Thus, $C = 0.46$ for composite (N) and 0.40 for composite (S). These values reasonably agree with the experimental value of 0.36 for composite (S), but not with that of composite (N). The discrepancy between the calculated and experimental value of C of composite (N) may indicate poor adhesion between the fibre and the epoxy in the absence of the solvent. The solvent may change the interface properties leading to an increase in G_U of composite (S) as discussed above. In view of the approximations used in Equation (5) and of the experimental errors, Hashin's equation[12] provides the most satisfactory description of the equilibrium moduli of the composite.

It is also noteworthy that the coefficient C as introduced here provides not only a measuring scale for the stiffness of the composite, but also a method for a comparative analysis of the various empirical models for composites.

CONCLUSIONS

Dynamic mechanical spectroscopy of unidirectional fibre-glass–epoxy composite shows that the strength of the lowest temperature relaxation process in it remains almost unchanged from that in pure epoxies, while those of the intermediate temperature are substantially reduced regardless of whether the composite is prepared from a solvent or not.

The T_g of the composite is greater than of the pure epoxy when both the composite and the epoxy are prepared from the solvent, but is lesser when the solvent was not used. An analysis in terms of the entropy theory shows that the presence of glass fibres increases the crosslink density in the former but decreases it in the latter case.

The increase in G_U of the composites over that of the pure epoxy differs in magnitude from the corresponding increase in G_R. An analysis in terms of a coefficient, $(G_U/G_R)_{\text{polym}}$, shows that, of the several empirical relations for the moduli of composites, Hashin's equation provides the most satisfactory agreement with the experiment.

The moduli of the epoxy composite prepared from the solvent differ from the normally prepared composite, for the presence of the solvent during preparation has two effects, namely: (i) decreases the crosslink density in the epoxy matrix, so that (G_U/G_R) of epoxy (S) is greater than (G_U/G_R) of epoxy (N), and (ii) improves the adhesion between the fibre and matrix in the composite, so that G_U of composite (S) is greater than G_U of composite (N). The effect is much less in the rubber region and therefore the coefficient, $C(\text{S}) > C(\text{N})$.

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