JOURNAL OF MATERIALS SCIENCE 27 (1992) 4582–4590

Plastic deformation in highly cross-linked rubber-modified epoxy resins

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Plastic yielding behaviour of three different cross-link density rubber-modified epoxy resins, at different rubber levels and temperature, were investigated. All the systems studied show decrease in Young's modulus, E, and yield stress, σ_Y, with increasing temperature and rubber content. The deformation process was analysed using both Bowden and Argon theories. Molecular parameters from each. theory were then compared with chemical structures of the epoxy systems.

1. Introduction

Despite the highly cross-linked nature of epoxy resin, plastic yielding will occur when it is being subjected to compression loading. Numerous theories have been put forward in explaining the plasticity of glassy polymers [1–4] mainly based on models specific to the molecular deformation process. Two theories which relate plastic flow to specific response at molecular level were developed by Argon and co-workers [5–7] and Bowden and co-workers [8, 9]. Although both theories were developed primarily to predict yielding of single-phase thermoplastic polymers, Young and co-workers have recently shown that the theories can also be applied to hybrid–particulate thermosetting composites [10, 11].

In this paper, plastic yielding from a relatively low cross-link density epoxy to the highly cross-linked tetraglycidyl diamino diphenyl methane–diamino diphenyl sulphone epoxy system, and the influence of different rubber contents in each system was investigated using both Argon and Bowden theories.

2. Experimental procedure

Diglycidyl ether of bisphenol A (DGEBA) resin (MY750) and tetraglycidyl diamino diphenyl methane (TGDDM) resin (MY720), were modified with carboxyl-terminated butadiene acrylonitrile (CTBN) rubbers. The types and properties of the CTBN are given in Table I and the structures of the resins and hardeners are shown in Fig. 1.

The epoxy and epoxy–rubber systems were cured either with piperidine or diamino diphenyl sulphone (DDS) and BF_3MEA.

For DDS-cured systems, a prereact of rubber–epoxy was premixed at 80 °C for MY720 resin. Triphenyl phosphine catalyst was added in the case of rubber–MY750 resin. The reaction was carried out under a nitrogen atmosphere and followed by determining the carboxyl content [10] at regular intervals.

The reaction was stopped when the COOH content had reduced to 0.05 or below (usually 3–4 h). The prereact was cooled and stored at 5 °C.

The prereact was used in combination with additional resins to obtain the desired rubber content. Mixing was carried out by heating the mixture (prereact, resin and curing agent DDS) to 100 °C and stirring for 15 min. The temperature was then raised to 135–140 °C and degassed until all the DDS had dissolved. The mixture was cooled to 110 °C and BF_3MEA catalyst was added. After further stirring for 3 min the mixture was poured into preheated glass moulds and cured in a heated oven for the required curing schedules.

For the piperidine-cured system, when rubber-modified, the desired rubber content was added to 100 parts of resin and the mixture heated and degassed to 80 °C for 30 min before being cooled to 30–40 °C. Piperidine was added and the mixture stirred for 5 min before it was poured into a preheated glass mould and cured.

For cast resin without rubber, the premixing stage with the rubber was omitted.

All the formulations and curing schedules used are given in Table II. Included are also the denotations which are used to describe the different formulations.

2.1. Young's modulus measurements

Young's elastic modulus, E, was measured in flexural mode in accordance with BS2782. Test specimens cut from cast plaques were 3 mm thick, 12 mm wide and 60 mm long. The test span was 50 mm. Testing was carried out in an electrical fan-heated chamber with a

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TABLE I Properties of Hycar CTBN elastomers (B. F. Goodrich Chemical Company)

	CTBN 1300 × 13	CTBN 1300 × 8
Molecular weight	3500	3500
Functionality	1.85	1.85
Acrylonitrile content (%)	27	17
Solubility parameter (cal cm)	9.14	8.77
Specific gravity at 25° C/25° C	0.96	0.948

liquid nitrogen cooling facility which enable the modulus from − 80 to + 80 °C to be measured. The modulus of each specimen was determined using Equation 1

$$E = \frac{L^3 P}{4bt^3 d} \qquad (1)$$

where d is the displacement, p the load at displacement, d, L the span, b the width of the specimen and t the thickness of the specimen.

TABLE II Formulations of the unmodified and rubber-modified epoxies

Epoxy	Type of rubber	Curing agent	Rubber content (%)	Notation
MY750		Piperidine	0	P750
MY750	1300 × 13	Piperidine	10	P750-13 (10)
MY750	1300 × 13	Piperidine	20	P750-13 (20)
MY750	1300 × 13	Piperidine	30	P750-13 (30)
MY750	1300 × 8	Piperidine	10	P750-8 (10)
MY750	1300 × 8	Piperidine	20	P750-8 (20)
MY750	1300 × 8	Piperidine	30	P750-8 (30)
MY750	—	DDS	0	D750
MY750	1300 × 13	DDS	10	D750-13 (10)
MY750	1300 × 13	DDS	20	D750-13 (20)
MY720	—	DDS	0	D720
MY720	1300 × 13	DDS	10	D720-13 (10)
MY720	1300 × 13	DDS	20	D720-13 (20)
MY720	1300 × 13	DDS	30	D720-13 (30)

2.2. Yield stress measurement

Cylindrical samples used for compression yield stress, σ_y, measurement were machined to ensure that the top and bottom surfaces were flat and parallel. The specimen height (19.5 mm) to diameter ratio was 2:1. The samples were deformed in a compression cage between polished steel plates in a Instron testing machine. Polytetrafluoroethylene (PTFE) tapes were placed between the surfaces of the samples and steel plates to reduce friction. A constant crosshead displacement rate of 0.5 mm min^{-1} was used. Measurements were made over a temperature range from − 80 to + 80 °C.

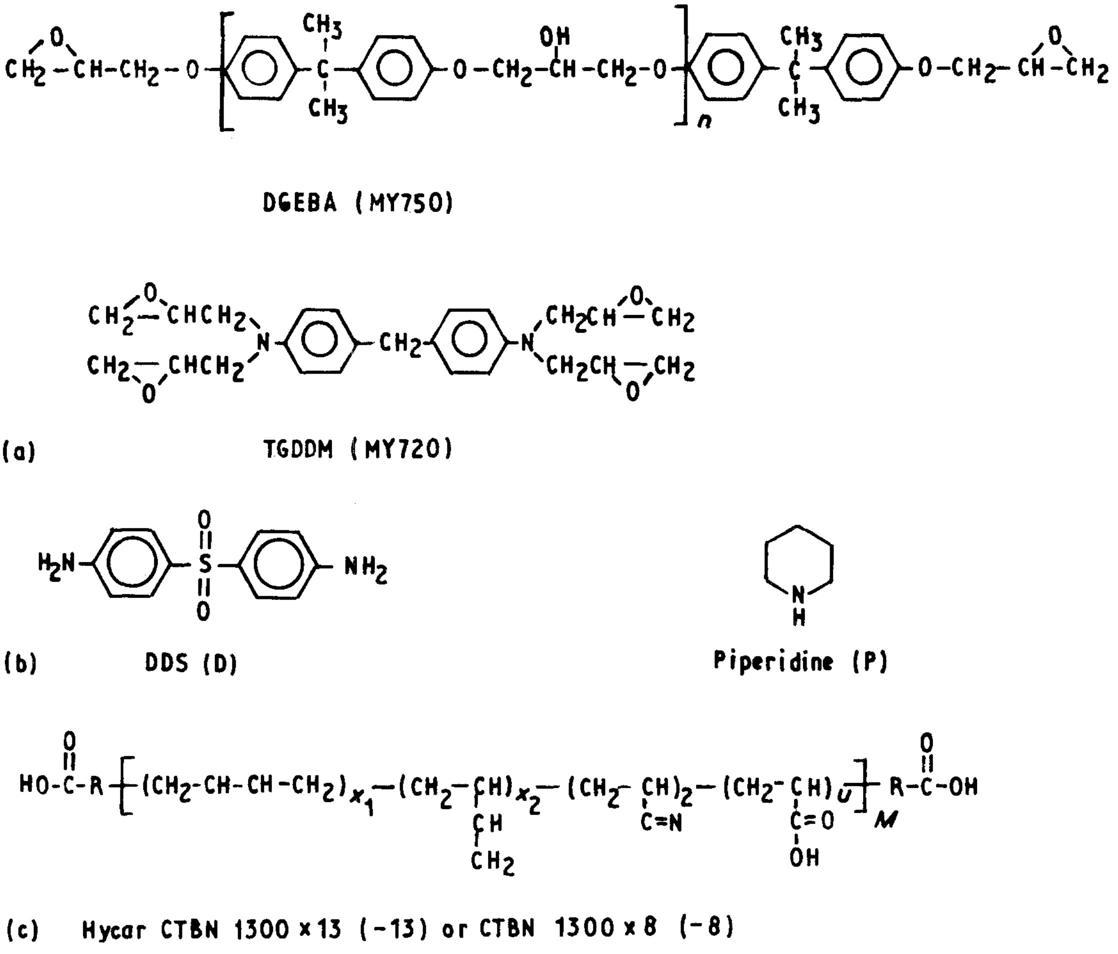


Figure 1 The structures of (a) epoxy resins, (b) curing agents and (c) CTBN rubbers. Notations used in this paper are given in parentheses.

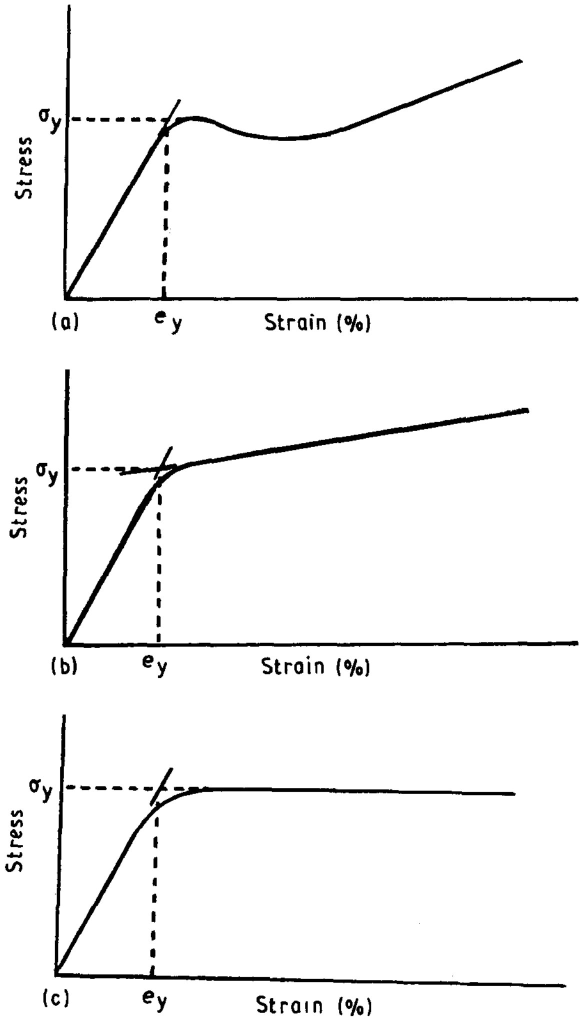


Figure 2 Determination of yield stress and yield strain from stress–strain curves.

Different compression stress–strain curves were exhibited by different resin systems at different temperatures and rubber contents. The procedures used to obtain the yield stress are shown in Fig. 2.

3. Results and discussion

The rubber in the rubber-modified systems has been previously observed to undergo phase separation [12, 13]. The resin's cross-link density from the lowest to the highest, in the order of P750 < D750 < D720, is not expected to be influenced significantly, if at all, by the incorparation of rubbers.

3.1. Elastic modulus measurements

The variation of Young's modulus with temperature for the systems studied is shown in Fig. 3a–d. The relative modulus of epoxy resin increases with increasing cross-link density, but decreases with increasing rubber content. The variation of the modulus with temperature for the unmodified resin is relatively small from − 80 to + 80 °C. For rubber-modified epoxy resin systems, depending on the systems, a sharp drop in modulus from − 80 to − 20 °C can be observed, followed by a relatively linear variation with temperature to 80 °C.

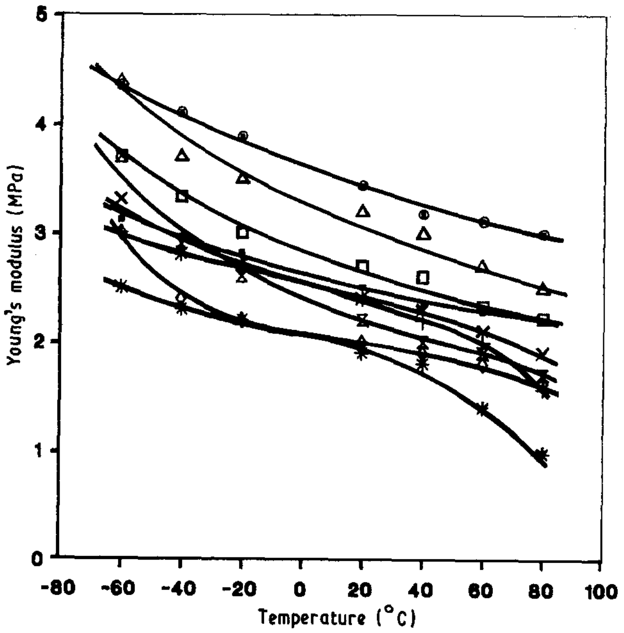


Figure 3 Variation of Young's modulus with temperature for the unmodified and rubber-modified epoxies. (■) P750, (+) P750-8 (10%), (*) P750-8 (20%), (□) D750, (×) D750-13 (10%), (◇) D750-13 (20%), (◉) D720, (△) D720-13 (10%), (⌧) D720-13 (20%).

The sharp fall in modulus at − 80 to − 20 °C can be related to the glass transition of the rubber particles which has been reported to be from − 20 to − 50 °C depending on the resin systems [13].

3.2. Compression yield stress

Most of the systems tested exhibit the behaviour of Fig. 2b, having a smooth and gradual transition from elastic to plastic behaviour, which remains largely uninfluenced by test temperatures below 80 °C. P750 neat resin and P750-8 [10] systems, however, demonstrate clear yield behaviour, as in Fig. 2a, at all temperatures.

The results for compression yield stress are shown as a function of temperature in Fig. 4. All systems showed a decrease in yield stress with increasing temperature. The decrease in yield stress is more gradual for the three unmodified epoxy resins at all temperatures, but a much sharper decrease is observed from − 80 °C to about − 20 °C for the rubber-modified systems. The higher cross-link epoxy resin showed higher yield stresses at all temperatures.

The rubber-modified epoxy resins showed a lower yield stress than their respective neat resins. The reason for this is that the rubber particles possess a lower shear modulus compared to the epoxy resins, which means that the rubbery particles are unable to support a significant share of the stress. Hence the yield stresses of the rubber-modified epoxies are much lower than those of the respective unmodified epoxies.

3.3. Argon's theory of yielding

Argon's theory viewed the randomly coiled molecules in the initial undistorted state of a polymer as containing a large number of kinks. The alignment process

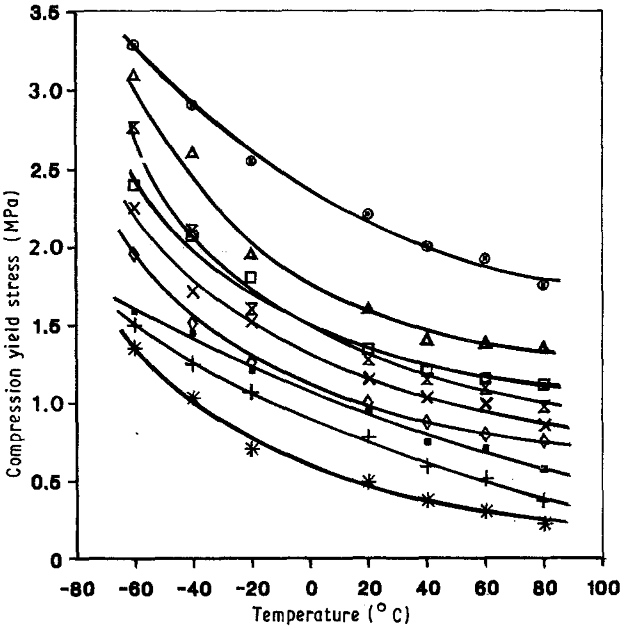


Figure 4 Variation of yield stress with temperature for the respective unmodified and rubber modified epoxies. For key, see Fig. 3.

was considered to be a systematic reduction of such kinks in pairs, each time a molecular unit undergoes a local rotation. The rotating molecular unit must displace neighbouring molecules, which oppose such motion by intermolecular elastic interaction. The intermolecular energy change associated with the double kink formation, Fig. 5, was modelled by the formation of wedge disclination loops after Li and Gilman [14].

Simpler forms of equations were given by Argon to facilitate the comparison of the theory with experimental results. The forms of the equations are given as

$$(\tau/G)^{5/6} = A - B(T/G) \qquad (2)$$

where T is the absolute temperature, A and B are constants, and are given by

$$A = \left(\frac{0.077}{1-\nu}\right)^{5/6} \qquad (3)$$

and

$$B = A\left(\frac{16(1-\nu)k}{3\pi\omega^2 a^3}\ln\left(\frac{\dot{\gamma}_0}{\dot{\gamma}}\right)\right) \qquad (4)$$

where ν is Poisson's ratio, K Boltzmann's constant, ω the net angle of rotation of the molecular segment between the initial conformation and the activated conformation, and a the mean molecular radius. $\dot{\gamma}$ is the shear strain rate with the one exponential frequency factor, $\dot{\gamma}_0$ taken to be $10^{13}\ \mathrm{s}^{-1}$. Both values of A and B from the theory can be obtained by rearranging Equation 4; the molecular radius, a, can be determined as

$$a^3 = \frac{A}{B}\frac{16(1-\nu)k}{3\pi\omega^2}\ln\left(\frac{\dot{\gamma}_0}{\dot{\gamma}}\right) \qquad (5)$$

The critical separation at yield of a pair of kinks (wedge disclinations) on the polymer molecule, z^*, at

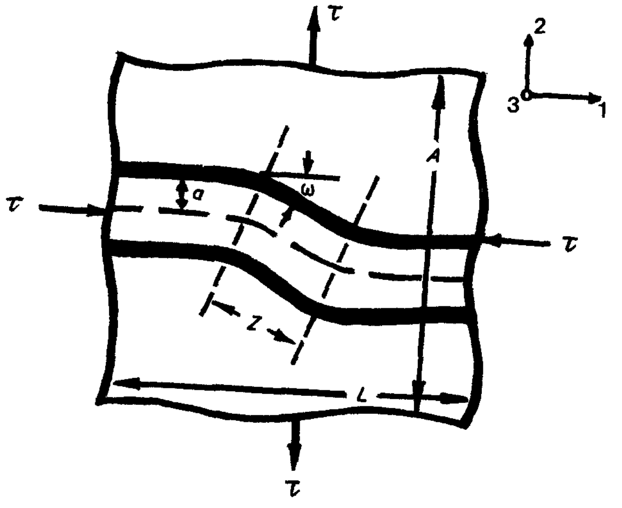


Figure 5 Schematic illustration of the formation of a pair of molecular kinks [after 5–7]. For illustrative purposes, the reverse deformation problem of an initially aligned specimen is considered.

absolute zero [5, 6] is given by

$$\left(\frac{z^*}{a}\right)_{T=0\,\mathrm{K}} = \left[\frac{45}{8(1-\nu)}\right]^{1/6} A^{-1/5} \qquad (6)$$

Equation 4 predicts that plots of $(\tau/G)^{5/6}$ against T/G should be linear and, if ν is taken to be 0.35 for most glassy polymers, it is expected that they should all have a common intercept of $A = 0.169$ as T approaches absolute zero, and all polymers asymptotically reach their ideal shear resistance. In practice, the carbon chain polymers such as polystyrene (PS), polymethymethacrylate (PMMA) and polycarbonate (PC) have been found to have values of $A \simeq 0.17$ while true aromatic and heteroaromatic polyimides have $A \simeq 0.14$.

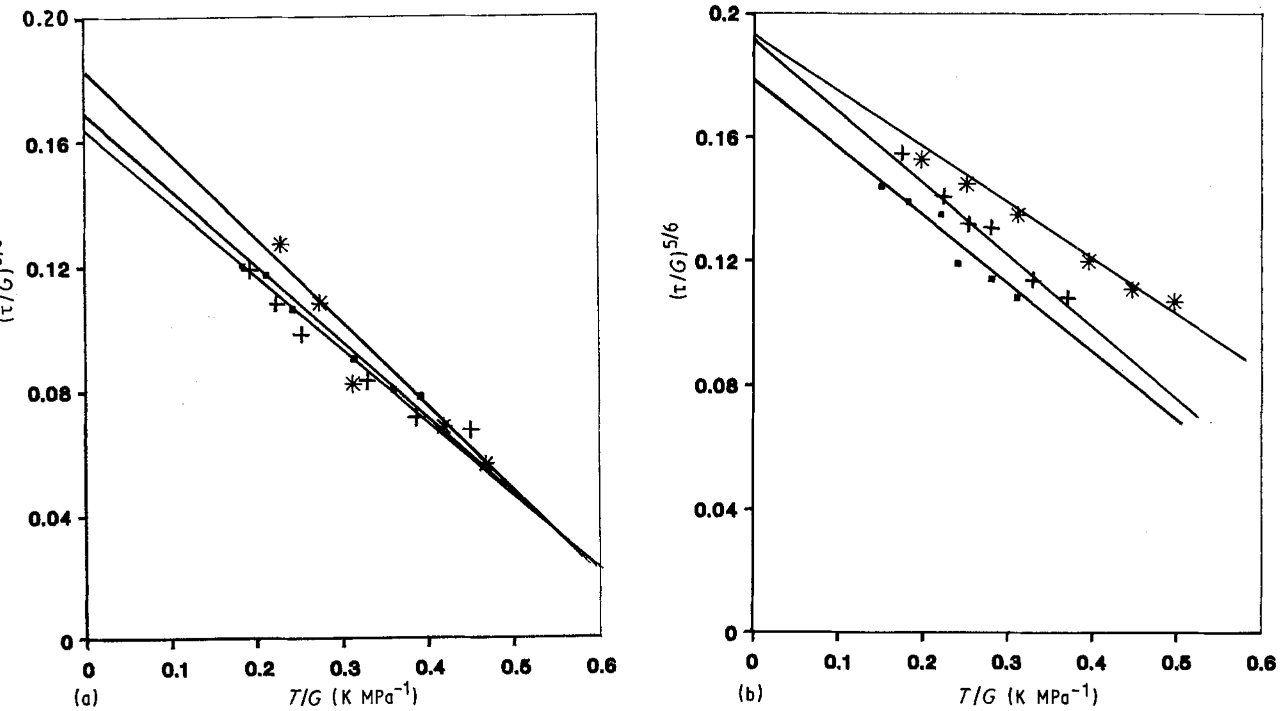
To test the applicability of this theory, it is necessary to convert the flexural moduli results and compression yield stress results into the corresponding shear moduli, G, and shear yield stresses, σ_y, by using the following relationships [5, 6]

$$G(T) = \frac{E(T)}{2(1+\nu)} \qquad (7)$$

$$\tau(T) = \frac{\sigma_y(T)}{3^{\frac{1}{2}}} \qquad (8)$$

taking Poisson's ratio, ν, to be 0.35. Strictly speaking, both $E(T)$ and $\sigma_y(T)$ should be measured at the same strain rate; however, it is not envisaged that significant error will be introduced in determining the various parameters as suggested by Young and co-workers [10, 11].

Fig. 6 shows the variation of $(\tau/G)^{5/6}$ with (T/G) for the various epoxy and rubber-modified epoxy systems. The solid lines are drawn as best fits of the lower temperature data. The experimental values deviate from Equation 2 as temperature approaches the T_{g_E} of the resin and the polymer become leathery, the experimental points tending to become parallel to the (T/G) axis. This deviation was also observed by Argon and Besonov [5–7] on thermoplastics, and Young and co-workers [10, 11] on epoxies. This explanation may



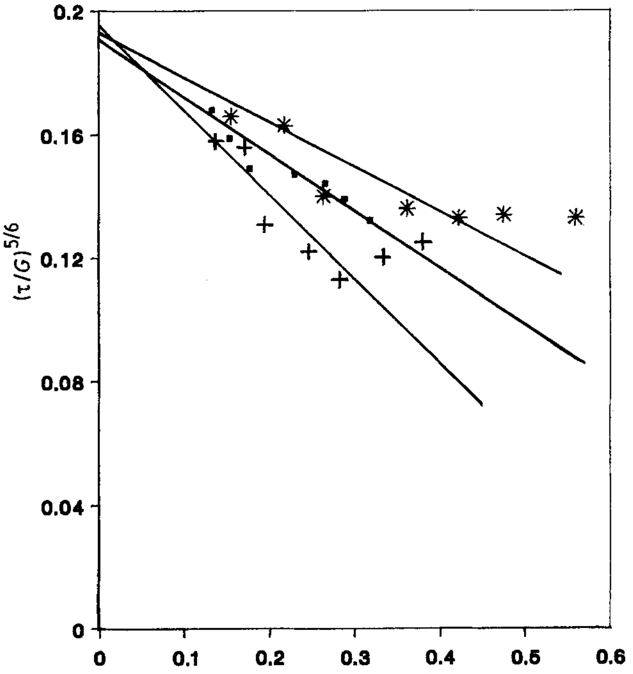


Figure 6 Variation of $(\tau/G)^{5/6}$ with T/G for unmodified and rubber-modified epoxies with (■) 0%, (+) 10% and (*) 20% rubber content. (a) P750-8, (b) D750-13, (c) D720-13.

account for the behaviour of the unmodified and rubber-modified P750 and D720 systems, where the deviation actually occurs at temperatures far below their respective T_{g_E}. However, Yamini and Young [10], on applying the theory to a DGEBA epoxy system with different amounts of curing agent (TETA), found that the temperature at which the deviation occurs is reduced progressively with the amount of hardener, and hence the cross-link density, or T_{g_E}, is increased.

The parameters A and B determined from the intercepts and slopes are given in Table III together with the molecular parameters obtained. For comparison, values determined by other investigators on other glassy polymers are also included.

The values of A for the epoxies are slightly higher than those predicted by Yamini and Young [10], while the values of B are slightly lower; nevertheless, they are in the range of those of other glassy polymers. The values of A for the unmodified epoxy resins increase with increasing cross-link density of the epoxy, while the values of B for both P750 and D750 are almost similar. No systematic variation in A and B values can be observed for the rubber-modified epoxy resin for all the systems.

Because the parameter A is related to the yield strain of the polymer, the increase in yield strain from unmodified P750 to D750 and D720, respectively, would be due to the increase in cross-link density.

The molecular parameters a and z^* did not show any systematic variation with rubber content, nor did the a and z^* values of the respective unmodified resins. This implies that in the rubber-modified resins, the yield process is governed principally by the deformation of the epoxy resin matrix, in the systems investigated.

The mean molecular radius of unmodified P750 epoxy resin is close to 0.47 ± 0.002 nm, while for D750 it is 0.515 ± 0.02 nm and for D720, 0.537 ± 0.033 nm. All the values are higher than those reported by Yamini and Young [10], but are within the range of those for PC and PMMA. Because the DGEBA epoxy resin and PC are both derivatives of bisphenol A, it is expected that the value of a for the epoxy should be similar to that for PC. However as the epoxy resin molecule is less bulky and has a larger number of carbon atoms in the repeat unit, the mean radius of the epoxy resin (0.472–0.537 nm) may be expected to be slightly less than that of PC (0.577 nm).

It is interesting to note that the value of D750 is closer to that for D720 than P750, despite the similar

TABLE III Values of various parameters derived from Argon's and Bowden's theories, of plastic deformation in glassy polymers

Polymers	A	B (MPa K^{-1})	a (nm)	z^* (nm)	$\boldsymbol{b}$ (nm)
P750	0.168	0.245	0.474	0.97	0.578
P750-8(10)	0.163	0.243	0.470	0.97	0.537
P750-8(20)	0.182	0.271	0.470	0.95	0.544
D750	0.181	0.231	0.495	0.10	0.859
D750-13(10)	0.190	0.219	0.512	1.223	0.865
D750-13(20)	0.192	0.194	0.535	1.07	1.000
D720	0.190	0.182	0.545	1.088	1.125
D720-13(10)	0.195	0.290	0.504	1.021	0.799
D720-13(20)	0.192	0.157	0.57	1.14	1.17
PC[a]	0.176	0.129	0.577	1.16	0.425
PET[a]	0.169	0.138	0.564	1.14	0.49
PS[a]	0.180	0.480	0.372	0.744	0.28
PMMA[a]	0.171	0.364	0.408	0.824	0.27
Epoxy-DGEBA[b]					
7.4 p.h.r. TETA	0.120	0.258	0.410	0.897	0.30
9.8 p.h.r. TETA	0.158	0.369	0.398	0.825	0.38
12.3 p.h.r. TETA	0.160	0.289	0.434	0.897	0.42
14.7 p.h.r. TETA	0.162	0.285	0.438	0.903	0.43

[a] A, B, a and z^* values taken from [5–7] and $\boldsymbol{b}$ from [8, 9]

[b] All values of A, B, a z^* and $\boldsymbol{b}$ taken from [10].

epoxy resin used to P750 and D750. The reason for this could be the different curing agents used for P750 and D750 systems. The latter system is cured with diamino diphenyl sulphone (DDS) as curing agent, and this curing agent forms part of the molecular chain; therefore, it is conceivable that the sulphone group from DDS would contribute to the bulkiness of the chain.

The value of critical separation, z^*, depends very much on the stiffness of the molecular chain. Polymers with flexible –C–C– backbones such as PMMA and PS have smaller values of z^* than PC. This is precisely what is found for the values of z^* for the epoxy resins, from 0.95–1.088 nm, which lie between the 0.824 nm of PMMA and 1.16 nm of PC.

It is also found that the value of z^* increases with decrease in cross-link density of the epoxy resin. It is tempting to explain this observation in terms of increasing stiffness of the molecular chain with increasing cross-link density. However, Yamini and Young [10] have pointed out that the degree of cross-link density has little effect on the values of a and z^* and that the yield process conceived in Argon's theory is controlled by the stiffness of the main chain. Therefore, the variations in z^* observed are probably due to the spacing of benzene rings, the presence of sulphone groups and, for the D720 system, the bulkiness of the TGDDM molecules.

3.4. Bowden's theory of yielding

An alternative model to predict the yield behaviour of glassy polymers was proposed by Bowden and coworkers [8, 9]. They suggested that the critical step in the yield process was nucleation caused by thermal fluctuations of small disc-shaped sheared regions in the polymer. The strain fields of the sheared regions were assumed to be analogous to those of dislocation loops are shown in Fig. 7 with a Burger's vector equal to the shear displacement and the radius equal to the radius of the sheared regions. It was stressed that the dislocation concept was only an analogy and at no time was it suggested that dislocations are present in glassy polymers. The concept was introduced because solutions for the energies and stresses in dislocation loops are readily available.

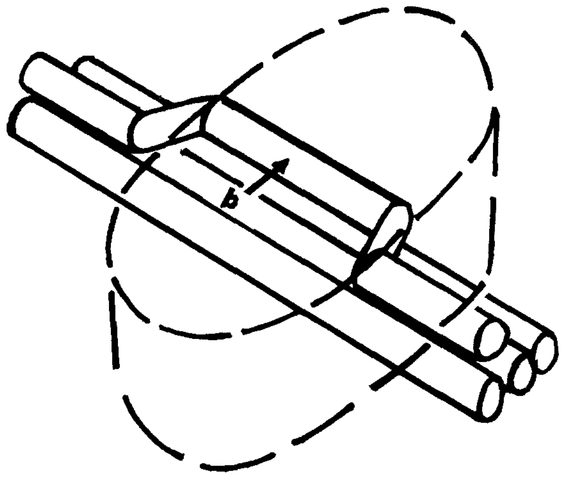


Figure 7 Schematic illustration of the disc-shaped sheared region, indicated by Bowden and co-workers [8, 9] with Burger's vector, $\boldsymbol{b}$.

Under an applied shear stress, the energy of a dislocation loop with Burger's vector $\boldsymbol{b}$, and radius, R, in a solid with shear modulus, G, is given by

$$U = (2\pi R)\left(\frac{G\boldsymbol{b}^2}{4\pi}\right)\ln\left(\frac{2R}{r_0}\right) - \pi R^2 \tau \boldsymbol{b} \qquad (9)$$

where r is the core radius of the dislocation. As the loop of radius, R, increases, the energy, U, increases until it reaches a value of U_c at R_c which can be found by differentiating Equation 9 to give

$$U_c = \left(\frac{G\boldsymbol{b}^2 R_c}{4}\right)\left[\ln\left(\frac{2R_c}{r_0}\right) - 1\right] \qquad (10a)$$

and

$$R_c = \frac{Gb}{4\pi\tau}\left[\ln\left(\frac{2R_c}{r_0}\right) + 1\right] \qquad (10b)$$

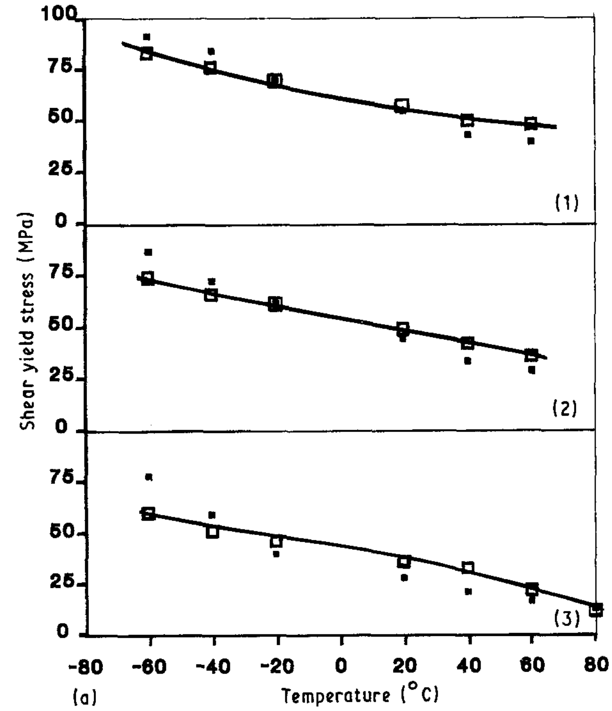
By assuming yield will occur at absolute zero when the critical value of $= G/\pi 3^{1/2}$ [8, 9], this leads to a value of $r_0 = b3^{1/2}/e$ ($\approx b$). Bowden assumed that yielding will occur when U_c reaches an energy of 50 KT (where K is Boltzmann's constant and T is the absolute temperature), which is the energy available from thermal fluctuations. By using this value of U_c, Equation 10b can be solved by choosing suitable values of $\boldsymbol{b}$ and experimental values of G to predict the corresponding value of R_a with temperature. Alternatively, Burger's vector, $\boldsymbol{b}$, can be calculated using experimental values of τ and G.

Flexural moduli and compressive yield stresses, σ_y, were converted to the corresponding shear moduli and shear yield stresses using Equations 7 and 8. The factors G/τ and U_c/Gb^3 were then evaluated from Equation 10b by taking values of $R_c = 2b, 3b, 4b, \ldots, nb$. This theoretical dependence of G/R_c on U_c/Gb^3 allows the values of U_c/Gb^3 corresponding to experimental G/τ values to be obtained. By substituting the experimental G values and taking $U_c = 50\ KT$, values of Burger's vector $\boldsymbol{b}$ were obtained. The average values of $\boldsymbol{b}$ were then used to reobtain the values of U_c/Gb^3 at different temperatures, and the corresponding theoretical G/τ values. These values enabled the theoretical τ to be estimated using experimental G values.

The theoretical τ values estimated by the above procedures were compared with experimental τ at different temperatures and are shown in Fig. 8a–c. The solid lines are the results calculated from Bowden's theory. The values of $\boldsymbol{b}$ chosen to achieve the best fit between the theoretical predictions and the experimental data are given in Table III.

The theoretical τ shows reasonable agreement with experimental τ, as shown in Fig. 8a–c. With increasing cross-link density of the epoxy, the value of $\boldsymbol{b}$ increases, but no systematic variation of $\boldsymbol{b}$ values with rubber content can be seen.

Because $\boldsymbol{b}$ is related to the yield strain of the polymer (in a similar way to A in Argon's parameter), increase in cross-link density, will increase the yield strain and therefore the Burger's vector. According to the model, the magnitude of $\boldsymbol{b}$ is related to the geometry and dimensions of the molecules making up the polymer. Therefore, polymers with large side groups (such as PS), or large units making up the molecular chain, such as PC or PET, have larger Burgers vectors



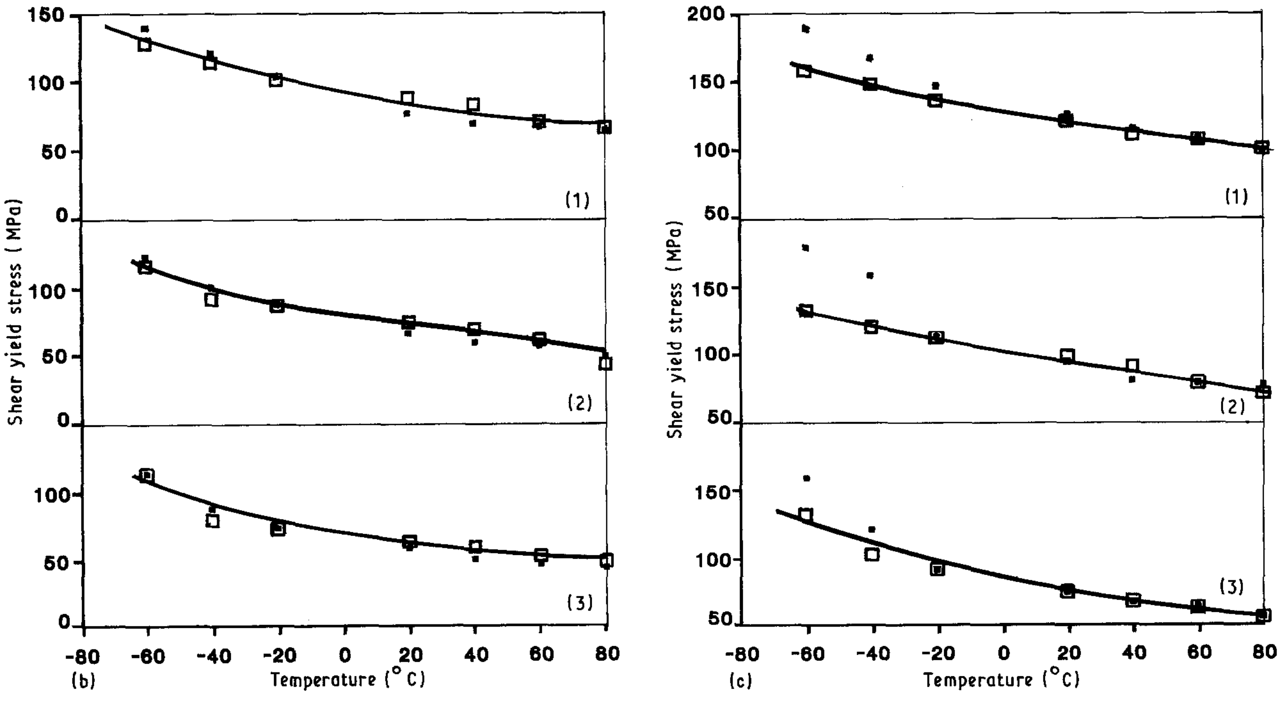
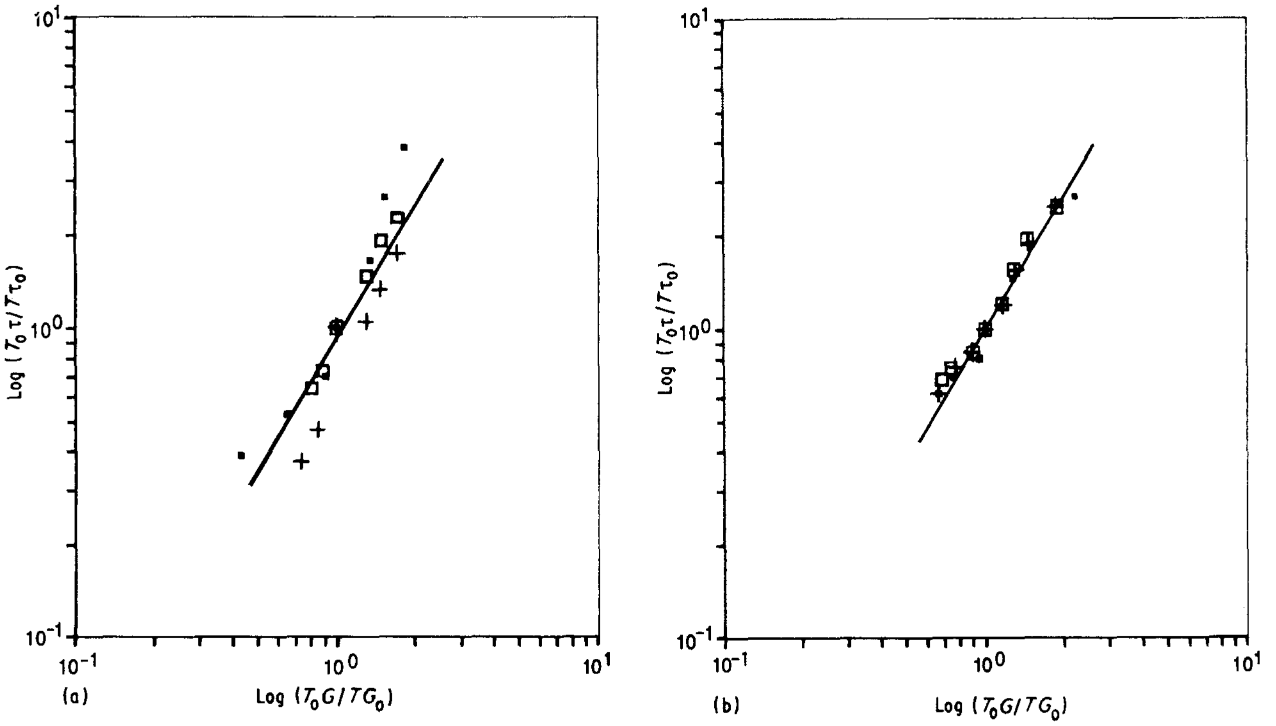


Figure 8 Variation of τ with testing temperature, T, for unmodified and rubber modified epoxies, with (1) 0%, (2) 10% and (3) 20% rubber content. (a) P750-8, (b) D750-13, (c) D720-13. The solid lines were predicted from Bowden's theory with values of $\boldsymbol{b}$ given in Table III.



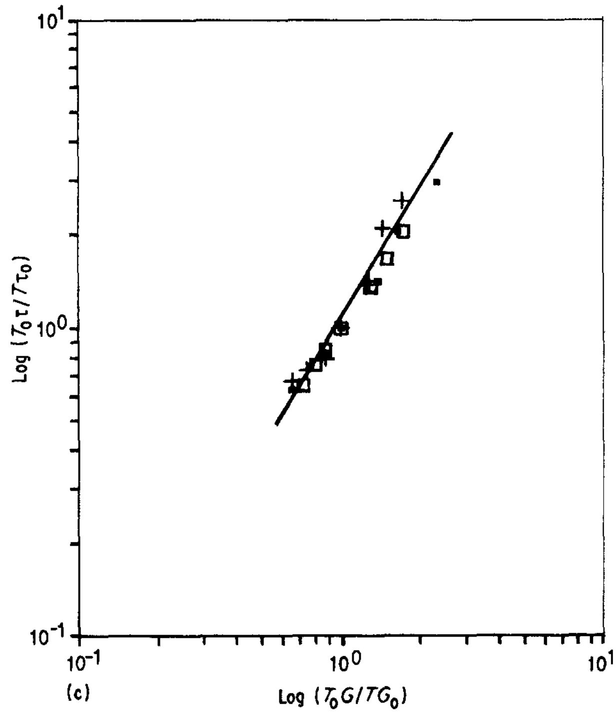


Figure 9 Plots of log ($T_0\tau/T\tau_0$) versus log ($T_0/G/TG_0$) for the unmodified and rubber-modified epoxies, with (□) 0%, (+) 10% and (■) 20% rubber content. (a) P750-8, (b) D750-13, (c) D720-13.

than polymers with smaller side groups, or smaller repeat units. The Burgers vectors found in this work are much higher than those that have been reported previously for epoxies and other polymers. The reason for this is not understood.

The relationship between G and τ in the Bowden's theory was expanded by Kitagawa [15] in a power-law relationship given as

$$\left(\frac{T_0\tau}{T\tau_0}\right) = \left(\frac{T_0G}{TG_0}\right)^n \qquad (11)$$

where T_0 is the reference temperature, τ_0 and G_0 are, respectively, the shear yield stress and the shear modulus at T_0, and n is a temperature-independent exponent.

It was shown that this relationship holds over a wide range of temperature for most polymers and that the value of n is 1.63 for amorphous glassy polymers without exception and about 0.7–0.9 for semicrystalline polymers. Young and co-workers [10, 11] have also found similar values of $n = 1.63$ for DGEBA epoxy containing various amounts of hardeners and also for rubber-modified epoxy filled with various amounts of glass particles.

The log–log plots according to Equation 11 for all the systems are shown in Fig. 9a–c. All the points fall close to the solid line which has a slope of 1.63. This suggests that the degree of cross-linking and the presence of phase-separated rubber particles have no influence on the values of n.

4. Conclusions

Plastic deformation in several epoxy resin systems with different degrees of cross-link density and rubber content has been studied. The experimental results were interpreted by Argon's and Bowden's theories. The molecular parameters determined from the theories were related to the molecular structures of the resin systems and are not influenced by the presence of cross-links and rubbers. These findings suggest that the mechanisms of deformation, in the temperature range studied, are similar to those in other amorphous polymers.

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Received 7 December 1989
and accepted 14 August 1990