Time and temperature dependence of fracture in a unidirectional glass-reinforced epoxy

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(Received 21 August 1978; revised 19 November 1978)

The results of a study on the time- and temperature-dependent behaviour of unidirectional glass fibre-reinforced epoxy are described and analysed. The fracture parameters examined are the fracture strength, the work of fracture and the apparent fracture toughness. It is shown that the fracture strength decreases with increasing temperature and decreasing loading rate; the work of fracture exhibits a sharp minimum in the vicinity of room temperature, and the fibre pull-out length increases by a factor of 4 at 76K as compared with the room temperature length; the fracture toughness is found to be independent of the crack length and only dependent on the fracture strength; thus its trend with loading rate and temperature follow those of the fracture strength.

INTRODUCTION

Fracture in glassy amorphous polymers, commonly used as matrices for composites, is strongly influenced by the rate of loading and by the temperature at which loading is performed. Regarding the rate effect, it is generally reported that the ultimate load necessary to initiate failure increases as the rate of loading is increased[1]. The fracture toughness (K_C) is expected to exhibit a similar dependence, according to the relationship $K_C \propto \dot{c}^n$, where $\dot{c}$ is the crack speed, and n is a measure of time dependence and is related to the loss tangent of the polymer[2]. In glassy crosslinked polymers such as epoxies, whose viscoelastic behaviour is more moderate, the fracture toughness of initiation is expected to be rate insensitive, or even to decrease with the strain rate[3,9]. A qualitative explanation for this is that in a viscoelastic material subjected to an external load, the flaws may grow in a subcritical mode before reaching an effective critical size corresponding to a lower fracture initiation stress. In crosslinked resins this process is less likely, resulting in rate insensitivity.

The effect of temperature on the fracture of polymeric materials has recently been studied quite extensively, and has been shown to vary with the type of polymer and to depend on the specimen geometry (see for example refs 4–7). The fracture toughness of initiation is mostly reported to increase with increasing temperature. In the case of epoxy resins the fracture energy and the fracture toughness are reported to decrease as the temperature is raised from 77[8] or 220K[9] to room temperature, and thereafter to increase rapidly up to the T_g[9].

The effect of time and temperature on fracture of composite materials is more complex than for homogeneous materials since additional fracture mechanisms are involved. The fracture behaviour of the composite derives from the viscoelastic nature of the matrix and from unique fracture processes, such as fibre debonding or pull-out, typical of the composite system. In addition to this complexity, the application of fracture mechanics parameters such as the fracture toughness to composites is in itself controversial. The present study, which is a continuation of a previous preliminary research[10], has been undertaken in order to answer some of the above questions. A unidirectional glass fibre-reinforced epoxy material has been chosen for the composite system, and since fracture in this system is governed by debonding and pull-out, the emphasis of this study has been put on examining the time and temperature dependence of these processes.

EXPERIMENTAL

Materials

Plates of epoxy resin (Araldite F/HT972) were cast in an open mould, gelled for 12 h at room temperature, cured for 2 h at 120°C and post-cured for 1 h at 180°C. Composite plates were prepared from prepregs manufactured by the following technique: Silane-treated E-glass fibres EC14-300-K937 (Vetrotex Ltd) in the form of single end-rovings containing 816 strands were wound on a winding machine drum and impregnated by an acetone solution of the resin. After the evaporation of the acetone and A-staging of the resin, the prepregs were removed from the drum, cut to size and laid in a mould. Plates were formed under a pressure of 0.7–2.5 MPa at 170°C for 15 min and post-cured for 1 h at 180°C. The thickness of the plates was controlled by spacers,

0032–3861/79/050653–06$02.00

Table 1 Results of the ultimate strength, Young modulus and the work of fracture

		$V_f = 0.37$			$V_f = 0.60$		
T (K)	Loading rate (m/sec)	σ_F (MPa)	E (GPa)	γ_F (kJm^{-2})	σ_F (MPa)	E (GPa)	γ_F (kJm^{-2})
76	8.3×10^{-5}	1,178	25.3	173.9	–	–	–
295	8.3×10^{-6}	–	19.1	38.8	615	31.2	62.7
295	8.3×10^{-5}	–	19.3	43.7	729	30.7	83.5
295	8.3×10^{-4}	–	18.5	48.3	747	31.7	81.7
295	1.02	642	–	91.0	791	–	122.1
295	5.08	801	–	100.9	–	–	118.6
335	8.3×10^{-6}	402	17.0	31.7	433	22.6	52.3
335	8.3×10^{-5}	444	18.6	33.8	391	19.4	55.7
335	8.3×10^{-4}	465	16.7	40.6	479	21.2	60.6
335	1.02	580	–	64.4	714	–	96.7
335	5.08	663	–	87.4	814	–	124.2
372	8.3×10^{-6}	281	14.3	41.6	266	18.9	71.7
372	8.3×10^{-5}	324	15.5	37.1	356	23.0	80.5
372	8.3×10^{-4}	355	15.2	40.5	418	19.3	74.5
372	1.02	455	–	59.9	488	–	70.5
372	5.08	481	–	89.3	609	–	115.1
411	8.3×10^{-6}	166	12.0	71.0	126	11.5	48.7
411	8.3×10^{-5}	214	12.8	68.7	164	12.7	54.1
411	8.3×10^{-4}	232	12.1	60.1	195	16.3	66.8
411	1.02	287	–	61.4	287	–	51.6
411	5.08	394	–	97.6	365	–	78.7

and the volume fraction of the fibres (V_f) in each moulding was determined by the number of prepregs introduced into the mould and by the amount of resin squeezed out during the course of moulding.

Test specimens in the form of $5.0 \times 5.0 \times 0.5$ cm^3 bars were cut from the plates. A 0.1 cm wide by 0.2 cm deep notch was introduced at the centre of each test specimen designated for the measurement of the work of fracture. In composite specimens both the notch and the loading mode were translaminar in order to promote debonding and pull-out rather than delamination.

Three-point bending experiments at crosshead speeds of 8.3×10^{-6} 8.3×10^{-5} and 8.3×10^{-4} m/sec were carried out with an Instron Universal Testing machine. Three-point bending impact experiments at striker velocities of 1.02 and 5.08 m/sec were carried out with a Dynatup instrumented impact system. The testing temperature was observed either by performing the test within a temperature cabinet attached to the Instron, or by heating the specimens to the required temperature in an oven prior to their testing in impact. In the second case, the specimen temperature was affected by the time elapsed from the instant of the removal of the specimen from the oven until the time of impact, and by the rate of cooling of the specimen. These factors were determined by simulating the procedure with a dummy specimen in which a thermocouple was embedded.

The mechanical parameters were determined as follows, repeating each test condition a minimum of three times. The flexural Young modulus (E) and fracture strength (σ_F) were calculated using linear elastic beam theory from the unnotched specimen data. The results from the notched specimens were used to calculate the work of fracture (γ_F) from the ratio of the integrated fracture energy to the total new fracture surface area.

RESULTS AND DISCUSSION

The experimental results for σ_F, E and γ_F are listed as a function of the loading rates and temperatures in *Table 1*. The typical scatter of the results is ±10%. The values of the fracture toughness (K_{IC}) and of the fracture surface energy (γ_I) were also calculated from the experimental data by the method given in ref 11; these results, however, will be discussed later.

Temperature effect

The results showing the dependence of the modulus and of the fracture strength on temperature are presented in *Figures 1* and *2*, respectively. The values of E and σ are seen to decrease linearly with temperature.

In contrast with the monotonic trend of the results of σ_F and E, the results for the work of fracture exhibit a more complex behaviour as demonstrated most clearly by the results for $V_f = 0.37$ in *Figure 3a*. An initial sharp decrease in γ_F with temperature is observed in the range 76–320K, followed by an increase which seems to reach a maximum between 370–390K (roughly the T_g of the resin). The very high values of the work of fracture at low temperatures may be explained on the basis of the very high pull-out length of the fibres at these temperatures. It is thought that due to the big difference in the coefficient of thermal expansion of the resin and the fibres, the high shear stresses which develop at the interface when the material is cooled down to 76K cause debonding, resulting in an increased pull-out length. Thus the pull-out work, being the dominant contribution to the work of fracture, increases dramatically at the low temperature. Microscopic examination of the fracture surfaces (see *Figure 4*) shows that the mean pull-out length at 76K is as much as 4 times higher than the value of approximately 0.6 mm measured at room temperature[12], in agreement with the respective 4-fold increase in γ_F. Between 320K and T_g, γ_F is controlled by the work of plastic deformation of the resin rather than by the shorter pull-out length, the former becoming increasingly more dominant until T_g is reached. Above T_g, the resin is rubbery and the amount of plastic deformation decreases. The results for $V_f = 0.60$ shown in *Figure 3b* generally fit in with this trend with the exception of the results for the two highest rates of test.

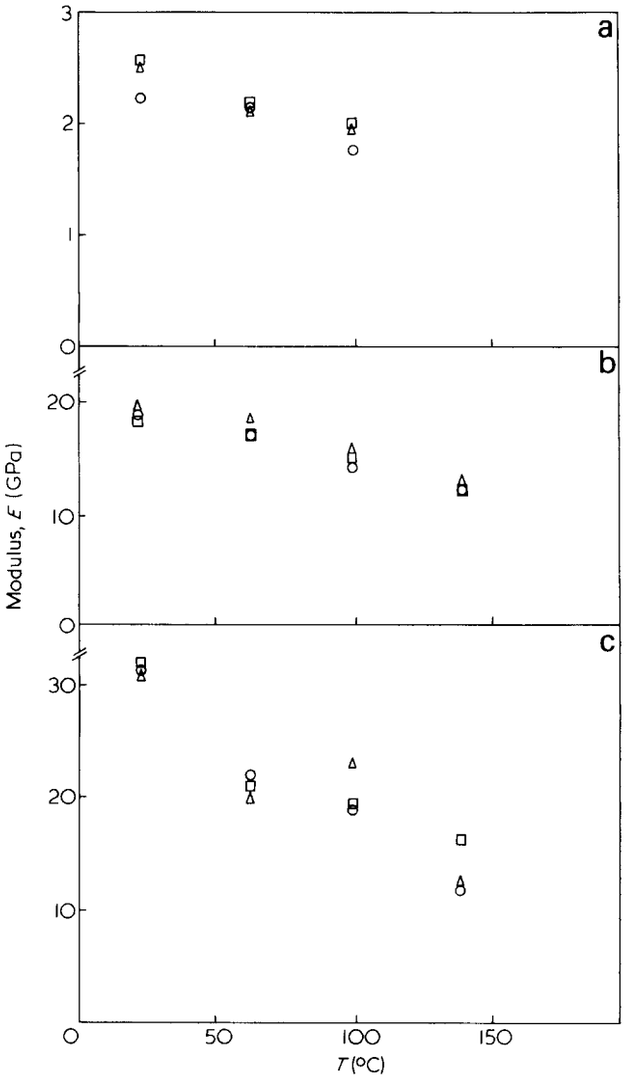

Figure 1 Dependence of the modulus on the testing temperature: (a) pure epoxy ○, 8.3 x 10^{-6}, △, 8.3 x 10^{-5}, □, 8.3 x 10^{-4}; (b) V_f = 0.37, (c) V_f = 0.60 (b) and (c) (□) 8.3 x 10^{-6}, (△) 8.3 x 10^{-5}, (○) 8.3 x 10^{-4}

Although a precise model for predicting the effect of temperature cannot be proposed at this stage, it seems that such a model should include two separate mechanisms: the first, which is pull-out controlled, active at the lower temperature range, and the second, which is resin viscoelasticity governed, dominating the process below T_g.

Rate effect

The results of the effect of the loading rate on the fracture parameters are an extension of a preliminary work described in ref 10. Whereas in the preliminary study various volume fractions were examined, in the present work the emphasis was put on a wider rate range. Thus, in contrast to the observed exponential relationship between σ_F and strain-rate shown in ref 10, the increase in σ_F with loading rate is now believed to be best represented by a power law ($\sigma_F \alpha v^n$, where v is the loading rate). The results showing the effect of the loading rate on the fracture strength are presented in *Figure 5*. The representation by a power law of the dependence of σ_F on loading rate corresponds with the relationship $K_C \alpha \dot{c}^n$ proposed by Williams[2] for pure polymers, and explained on the basis of a slow stable crack growth occurring prior to final fracture.

The results for the work of fracture as a function of loading rate are presented in *Figure 6*. Here also the typical scatter was about ±10%. At the lower temperatures, γ_F increases monotonically with increasing loading rate. This may be explained as before[10] on the basis of the dependence of the pull-out energy on the fibre–matrix friction stress, itself a decreasing function of the speed. The trend at the highest temperatures is less clear, suggesting that the rate sensitivity of γ_F decreases with increasing temperature. As indicated previously, the decrease in the rate sensitivity of γ_F might result from a change in fracture mechanism, i.e. at

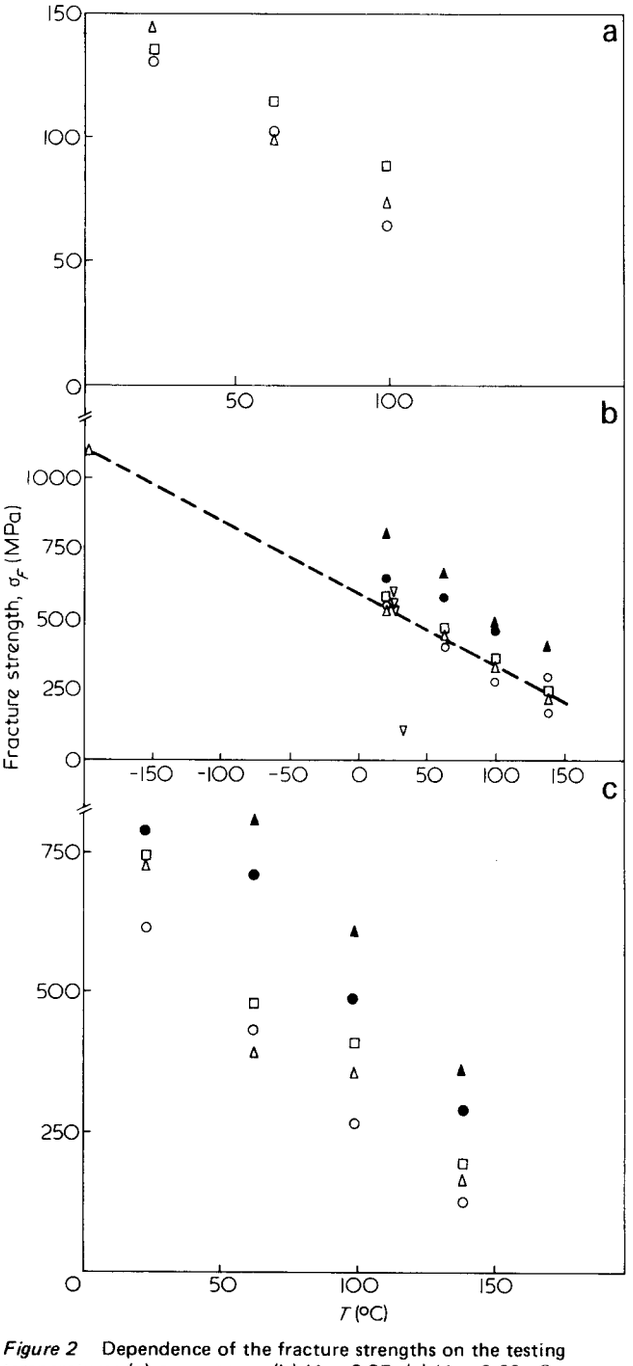

Figure 2 Dependence of the fracture strengths on the testing temperature: (a) pure epoxy; (b) V_f = 0.37; (c) V_f = 0.60. ○, 8.3 x 10^{-6}; △, 8.3 x 10^{-5}; □, 8.3 x 10^{-4}; ●, 1.02; ▲, 5.08 ms^{-1}

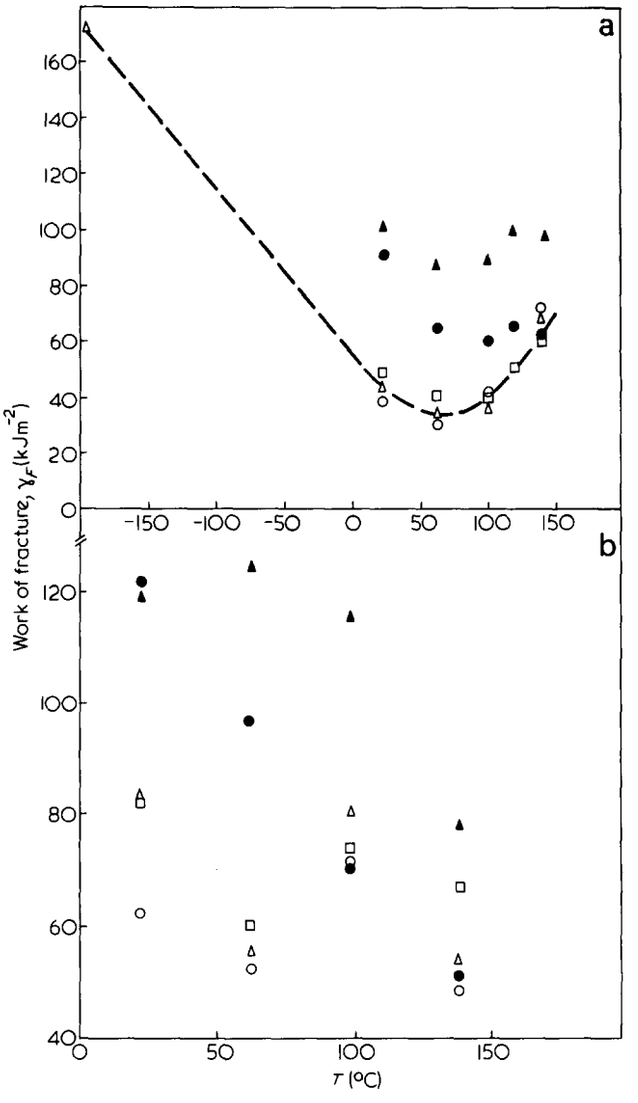

Figure 3 Work of fracture as a function of the testing temperature: (a) $V_f = 0.37$; (b) $V_f = 0.60$. Key symbol as in Figure 2

higher temperature, fracture is governed by the viscoelastic nature of the resin and not by pull-out.

Fracture toughness and fracture surface energy

It has been pointed out by many investigators (see for example ref 13) that, due to the complexity of the role of the fibres and the fibre–matrix interface in the failure process, the fracture toughness determined for composite materials by conventional methods is not necessarily a valid fundamental material parameter. In recognition of this fact, the term 'apparent fracture toughness' (K'_{IC}) is used[14]; and K'_{IC} and the fracture surface energy (γ_I) are generally accepted for materials comparison purposes. In contrast to that it is strongly argued that LEFM might indeed be applicable to composites, provided the testing conditions and the measured parameters are defined precisely[15]. For example, to include the diameter of the debonding zone in the calculation of K[16], or to use an initial sharp crack. Furthermore, a recent study[17] on identical glass–epoxy composites (carried out at room-temperature, crosshead speed of 0.05 cm/min and employing a wide range of notch depths) showed that K_{IC} obtained directly from the Griffith equation was very close to values of K'_{IC} calculated with the use of Srawley–Brown K calibration curves.

The results for K'_{IC} and γ_I are presented in *Table 2* as a function of fibre volume fraction, temperature and loading rate. The values were determined from the notched specimen data by the method described before[11]:

$$K'_{IC} = \frac{1.5 P_{max} l}{bd^2} Y(c) c^{1/2}$$

and

$$\gamma_I = \frac{(K'_{IC})^2}{2E}$$

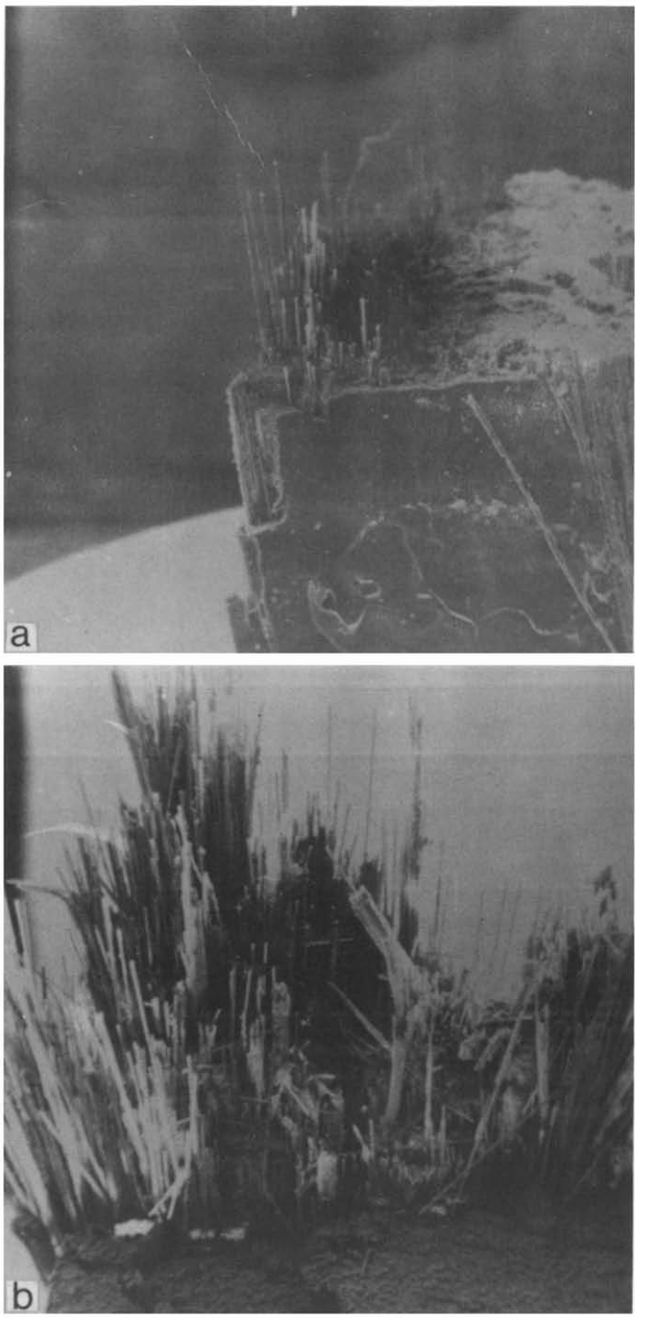

Figure 4 Scanning electron micrographs of the pull-out regions of specimens of $V_f = 0.37$; (a) 295K (x27); (b) 76K (x22)

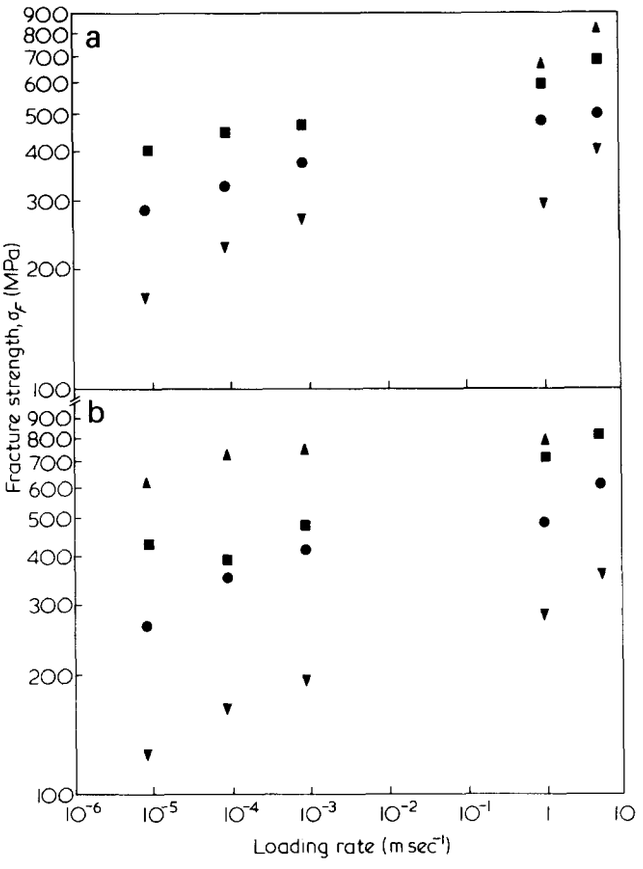

Figure 5 Effect of loading rate on fracture strength: (a) $V_f = 0.37$; (b) $V_f = 0.60$. ▲, 295K; ■, 335K; ●, 372K; ▼, 411K

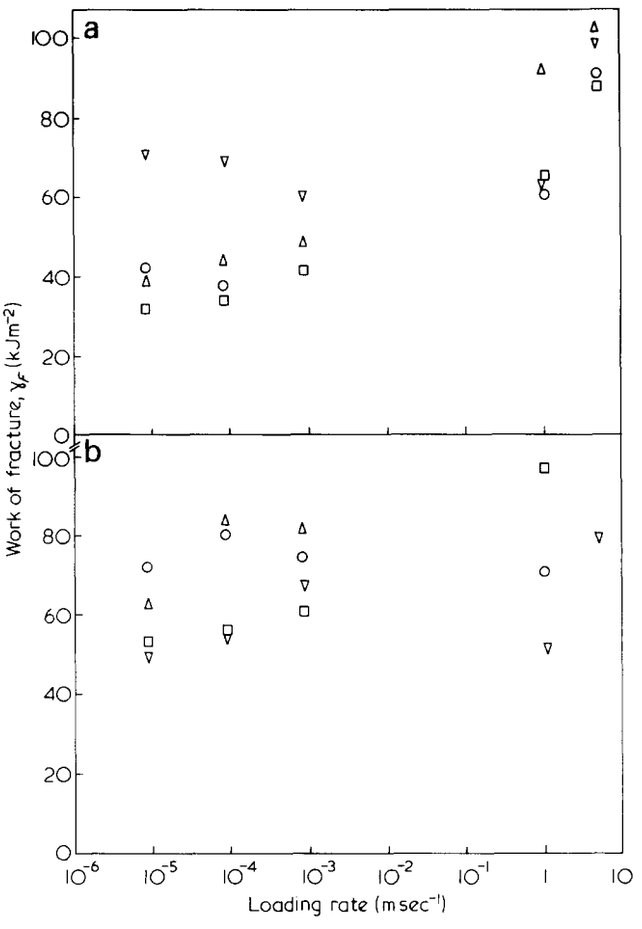

Figure 6 Work of fracture as a function of the loading rate: (a) $V_f = 0.37$, △, 295K; □, 335K; ○, 372K; ▽, 411K; (b) $V_f = 0.60$, △, 245K; □, 335K; ○, 372K; ▽, 411K

where P_{max} is the maximum load; l, b and d are the specimen length, width and depth, respectively; $Y(c)$ is the compliance coefficient; and c is the notch depth.

The results show that K'_{IC} and γ_I increase with increasing loading rate and decrease with increasing temperature. As pointed out previously[10] it seems that the increase in fracture toughness with the strain-rate corresponds with the increase in the fracture strength. In addition to that, it is now apparent that the decrease in fracture toughness with increasing temperature simply reflects the fall of σ_F with increasing temperature. We think that this situation, whereby changes in fracture toughness relate to changes in fracture strength, is essential in composite materials. Clearly, the volume fraction of the fibres is just another typical variable which has a similar effect on the fracture toughness and on the fracture strength. When the volume fraction is increased both σ_F and K_C increase, and *vice versa*.

CONCLUSIONS

The results of this study on glass fibre-reinforced epoxy composites show the following:

(1) the fracture strength decreases with increasing temperature and decreasing load rate;

(2) the work of fracture exhibits a sharp minimum temperature in the vicinity of 60°C. The pull-out length increases by a factor of four at 76K as compared with the room temperature length. It appears that at low temperatures the pull-out mechanism is dominant, while at temperatures

Table 2 Results for the effective fracture toughness and the fracture surface energy (0.2 cm deep notch)

		$V_f = 0.37$		$V_f = 0.60$	
T (K)	Loading rate (m/sec)	K'_{IC} (MPa.m$^{1/2}$)	γ_I (kJ/m^2)	K'_{IC} (MPa.m$^{1/2}$)	γ_I (kJ/m^2)
76	8.3×10^{-5}	47.1	46.0	—	—
295	8.3×10^{-6}	25.8	17.5	31.5	15.9
295	8.3×10^{-5}	28.4	21.3	34.7	19.5
295	8.3×10^{-4}	29.2	22.5	36.6	21.5
295	1.02	34.8	31.9	44.5	31.8
295	5.08	38.6	39.2	43.3	30.4
335	8.3×10^{-6}	17.7	8.9	21.1	10.7
335	8.3×10^{-5}	19.6	10.9	22.2	11.8
335	8.3×10^{-4}	21.6	13.2	23.0	12.7
335	1.02	27.2	21.0	33.7	27.4
335	5.08	28.6	23.1	36.6	32.3
372	8.3×10^{-6}	14.6	6.9	18.1	8.1
372	8.3×10^{-5}	16.5	8.6	21.7	11.7
372	8.3×10^{-4}	17.2	9.5	21.9	11.8
372	1.02	21.9	15.5	27.0	17.9
372	5.08	25.1	20.1	31.4	24.3
411	8.3×10^{-6}	8.9	3.1	8.0	2.4
411	8.3×10^{-5}	11.3	5.0	9.7	3.5
411	8.3×10^{-4}	9.7	4.0	12.7	6.2
411	1.02	17.8	12.3	14.4	7.8
411	5.08	13.9	8.1	17.4	11.2

closer to T_g the viscoelastic behaviour of the resin controls the work of fracture;

(3) the fracture toughness is found to reflect the behaviour of the fracture strength, i.e. to decrease with increasing temperature, and to increase with increasing loading rate.

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