

Deformation characteristics of reinforced epoxy resin

Part 1 *The mechanical properties*

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Specimens, cut from a commercially produced epoxide resin reinforced by layers of glass cloth, were tested in tension at various strain-rates and temperatures, and at various orientations of the fibres to the tensile axis. Results are presented for elastic moduli, yield and fracture stresses, and elongations. These results give ample evidence of a decrease in the effectiveness of the reinforcement as deformation proceeds. Strain-rate-change tests show that the rate sensitivity of the composite rises rapidly as the strain increases beyond a critical value, which also indicates that the behaviour is increasingly governed by the properties of the matrix.

1. Introduction

The fact that the addition of a second phase may be used to strengthen materials is well known. For many years this phenomenon has been technically used and theoretically investigated in metal alloys, where suitable thermal treatments can produce an optimum distribution of precipitates. The strengthening of polymers with fibres of a different material type (e.g. glass or metal) can provide even more spectacular results due to the large difference between the properties of the two phases. Such strengthening has been treated theoretically under idealized assumptions by several investigators in terms of the theory of elasticity, but few have tried to understand the thermal or dynamic properties. In a recent review [1], for example, only a few references to work on this subject were quoted. But it is known that polymeric materials show a considerable rate and temperature dependence in their strength properties, so that similar effects are likely to occur in reinforced polymers. The use of such materials in applications involving dynamic loading therefore depends on the study of the rate and temperature dependence of their strength and fracture behaviour.

2. Material and testing procedure

This report describes an investigation on com-

mercially available epoxide resin with bi-directional woven glass fibres, a system which has been rarely dealt with in the literature, and only with respect to properties at room temperature and at conventional low strain-rates [2]. The material was supplied by Permalit Limited, Gloucester, in the form of sheets 0.9 mm thick. It is specified as "Permaglass 22 FE", and it is a laminate of five layers of Marglass 116 S close-weave glass cloth and an epoxide resin (Araldite MY 753 resin and HY 951 hardener). At room temperature the matrix is in the brittle state, with a glass transition at approximately 150°C. A separate sheet of this matrix without glass reinforcement was also tested for comparison.

Samples were cut out at different angles to the fibre directions. The original sheets were rectangles ($1.9 \times 0.9 \text{ m}^2$) with sides parallel to these directions, and the orientation of $\theta = 0^\circ$ was arbitrarily defined as parallel to the longer side of these sheets. The microstructure of the composite is shown in Fig. 1.

Tensile tests at room temperature have been carried out over a wide range of strain-rates using an Instron floor model test machine. For samples of about 25 mm gauge length, the minimum strain-rate was $3.3 \times 10^{-5} \text{ sec}^{-1}$. Tests at rates up to 15 sec^{-1} have been carried out using a hydraulic machine which has been

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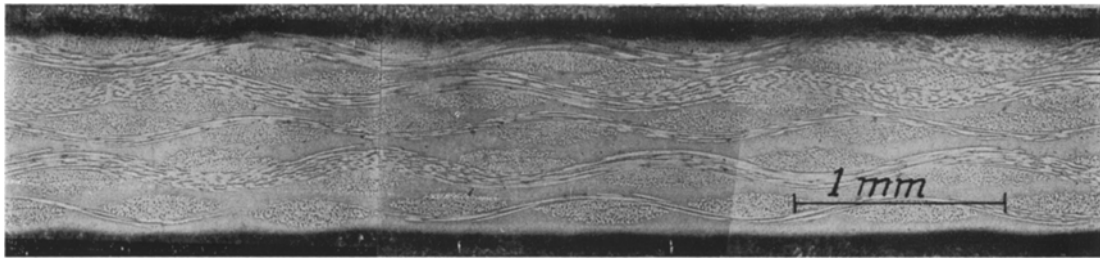


Figure 1 Longitudinal section through "Permaglass 22 FE" sheet.

described in a previous publication [3]. Sub-ambient temperatures were obtained in a temperature-controlled chamber by spraying with liquid nitrogen. Temperatures above room temperature were achieved by a heated-air stream.

It was necessary to use samples with steel shims attached with araldite (see inset in Fig. 4) and specially designed grips to avoid errors arising from the deformation of the specimen ends in conventional grips [4].

The upper temperature limit was determined by the strength of the araldite which was used to attach the steel parts to the samples, and this limit was about 120°C. The total temperature range covered was therefore 77 to 380 K. More details about testing methods have been given in a previous report [4].

3. Discussion of experimental results

3.1. General observations

For all orientations, the stress-strain curves for the composite were linear during an initial portion, then they continuously deviated from linearity and decreased in slope. The failure in samples of 0 or 90° orientation always occurred rapidly and the macroscopic fracture surface was perpendicular to the tensile axis. The crack went straight across the specimen, in places passing through bundles of fibres which were in its path. For 45° specimens the crack again followed the direction of the fibres so that it created local fractures which were inclined at $\pm 45^\circ$ to the tensile axis. At a maximum or ultimate load the crack started at one side of the sample and propagated successively over the entire cross-section, while a drop in the applied load could sometimes be recorded. In the areas immediately adjacent to the fracture and along the whole gauge length, the structure exhibited an effect which is typical of a material which has undergone some deformation (this was seen for

45° as well as for 0° orientations). The individual fibres in each of the fibre bundles did not remain intact, as in the undeformed state, but were broken (Fig. 2). Voids were visible around them and in parts of the immediately neighbouring matrix. Since this appearance was not exclusively limited to the failure site, it could be traced back to the general deformation prior to fracture, during which voids could have been formed within the plastically strained matrix around the undeformable glass fibres. As a consequence of this, the fibres must have lost their ability to act as reinforcement. Afterwards, during the preparation for microscopic examination, polishing could have damaged the fibres, since they were no longer supported by the surrounding matrix.

During the deformation, a process could be seen which has elsewhere been called "multiple fracture" [5]. More and more small cracks developed mainly normal to the fibres. These "micro-cracks" were only resolvable by optical magnification, and towards the end of the deformation they were so concentrated in several regions that the sample lost its transparent appearance: the deformation was not uniform (Fig. 3). This behaviour was not influenced

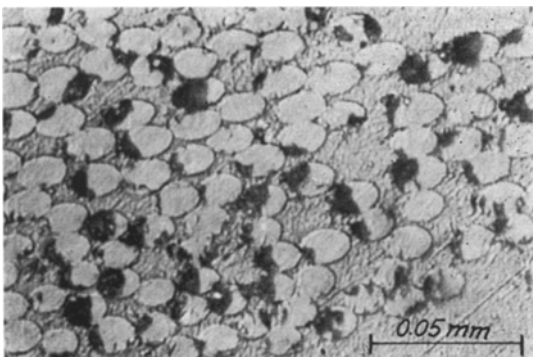


Figure 2 Detail of a strained material in 45° orientation (the cross-sections of the fibres are elliptical).

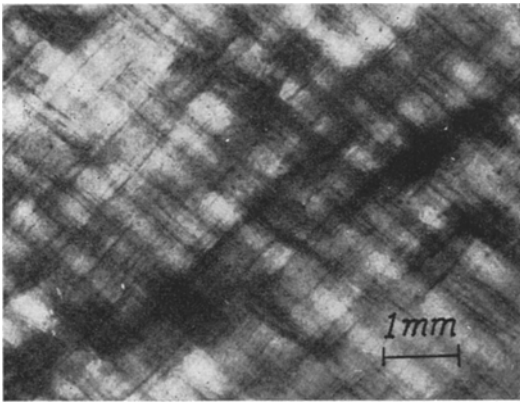


Figure 3 Areas of high localized deformation.

directly by the test temperature. Only inasmuch as the fracture elongation increased with increasing temperature were the oblique "deformation bands" more pronounced. These features were totally absent in the deformation of unreinforced epoxy.

3.2. The orientation dependence of some properties

Stress-strain curves, determined with an Instron extensometer on samples cut out in different

orientations θ as defined earlier, are shown in Fig. 4. In Fig. 5, the orientation dependences of several material characteristics are plotted. Young's modulus E , fracture stress σ_f and yield stress σ_y (defined as the stress at the first deviation from linearity) seem to change continuously*, yet the curve of the fracture elongation ϵ_f

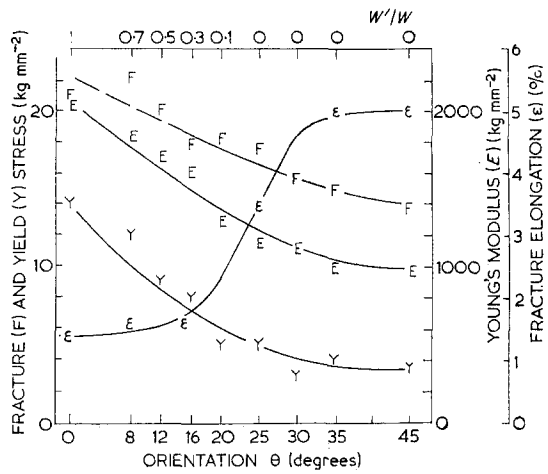


Figure 5 Characteristics for different fibre orientations at room temperature.

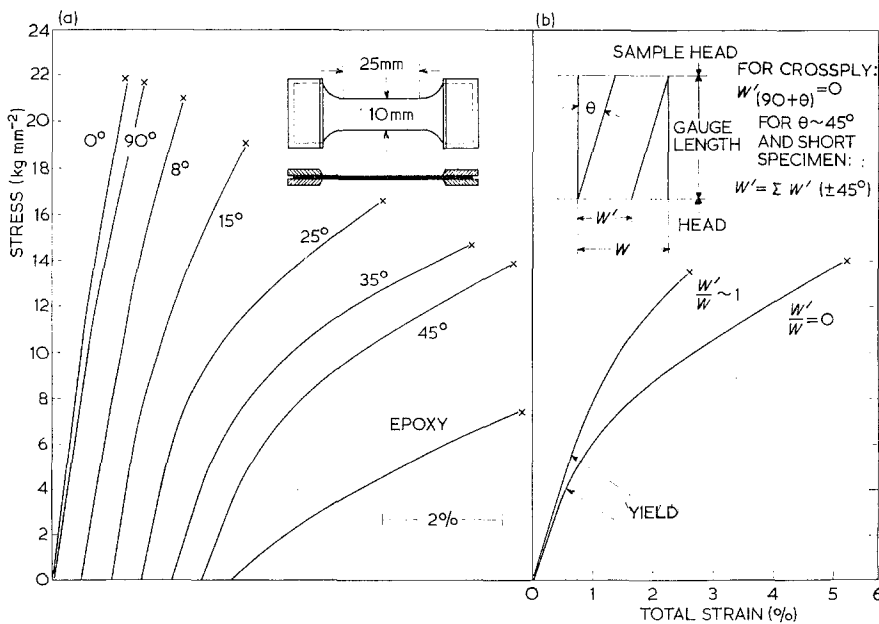


Figure 4 (a) Stress-strain curves of specimens with different fibre orientations ($w'/w = 0$, except for 0° and 90° orientations) at room temperature. (b) Stress-strain curves of specimens with 45° orientation and different w'/w ratios. The inserts give sample shape and the definition of the w'/w ratio.

*From the theory of anisotropic elasticity it can be shown that $1/E$ should vary linearly with $\cos^2 2\theta$. This is indeed obeyed [4].

exhibits a rather pronounced step. This may indicate that some change in the testing conditions occurs at medium angles. The inset in Fig. 4b illustrates the foundation for such reasoning. It is clear that both upper and lower ends of continuous fibres are, for a given gauge length, only attached to both end pieces of the specimen when the angle of orientation is small. The ratio w'/w (defined in Fig. 4b), apparently of some importance, is therefore added for the various orientations in Fig. 5.

It appeared of interest to vary the ratio w'/w further by changing sample length and width, and to examine its influence on the material properties. First, we measured the effect of the two extreme conditions $w'/w = 0$ and 1 on the shape of the stress-strain curve of a 45° composite (Fig. 4b). No significant influence was exerted on Young's modulus* or on the fracture stress. The yield stress increased slightly with decreasing sample length. Only the total elongation was considerably reduced by about one half for a short sample where the ratio w'/w approached unity. With this information the abrupt change of the ϵ_f curve in Fig. 5 can be explained. When the value of w'/w changes from zero to unity, ϵ_f is reduced. This change is superimposed on the inherent change which comes from the variation of θ alone: the total reduction of ϵ_f with decreasing angle of orientation and at constant gauge length is the sum of the two contributions. The variation of ϵ_f will be more pronounced than that of the other characteristics since ϵ_f is, according to Fig. 4b, more affected than σ_f , σ_y or E .

A more detailed description of shape effects on elastic properties is given in Fig. 6. Sample length and width were varied so that the resulting w'/w values were for all orientations within the limits zero and unity. The variation of E was minor for samples with 45° orientations, in accordance with the result of Fig. 4b, but it became severe for low values of θ .

3.3. The strain-rate dependence of some properties at room temperature

Fig. 7 shows strength and ductility values for all orientations in their dependence on strain-rate, and they are compared with the properties of unreinforced epoxy. The strength properties increased at first slightly, but there is a more pronounced increase at high strain-rates. The

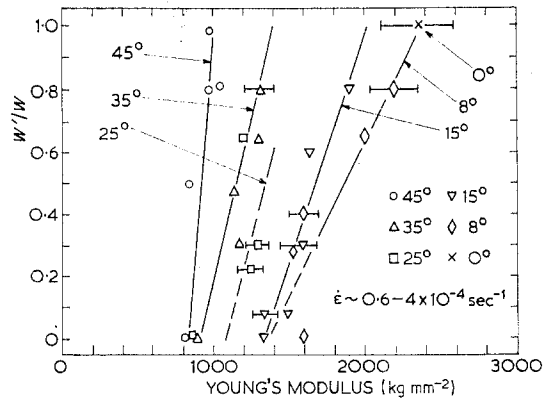


Figure 6 The dependence of Young's modulus on w'/w .

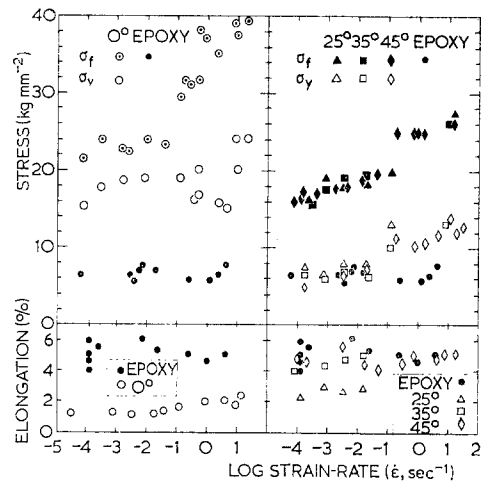


Figure 7 The strain-rate dependence of strength and fracture elongation for various orientations at room temperature.

ductility was rather insensitive to the applied variations.

Samples with fibre orientations of 0° showed little ductility and so it is not surprising that the σ_f values for 0° composites scatter considerably. Misalignments, due to unavoidable inaccuracies in the sample preparation, caused local stress concentrations and these could not be relieved by local straining. The equally large scattering of the yield stress has, however, other reasons, and is to be explained by the difficulties of detecting the first deviation from the initial straight line. In agreement with the previous findings of Fig. 5, there was not much difference between the fracture and yield stresses for 25° ,

*Since the Instron extensometer's use is limited to a certain sample width and length, the curves of Fig. 4b had to be determined from load-time charts of the Instron machine. The accuracy is reduced.

35° and 45° composites. Scattering was small since the ductility was good. It is noteworthy that the elongation did not decrease when the fracture stress was at its largest at relatively high strain-rates, as often happens under such conditions in other materials.

3.4. The temperature dependence of some strength characteristics

The following tests were performed at a strain-rate of $\sim 3.3 \times 10^{-4} \text{ sec}^{-1}$. The temperature dependence of Young's modulus is given in Fig. 8. The values increase rapidly with decreasing temperature, and they are highest for low angles of orientation. In comparison, both the slope and the absolute values of the moduli are much lower for unreinforced epoxy. The accuracy of these results is not good owing to the fact that the measurement again had to be derived from the corrected Instron load-time recordings.

The fracture stresses of composites and of epoxy are plotted in Fig. 9. Scattering was severe for specimens of 0° orientation due to reasons already mentioned. The fracture stresses decreased as the angle between fibre orientation and the tensile axis approached 45°, and at the

same time the scatter band became narrower. Fracture stresses for epoxy were considerably lower, and they were again strongly affected by the brittle behaviour.

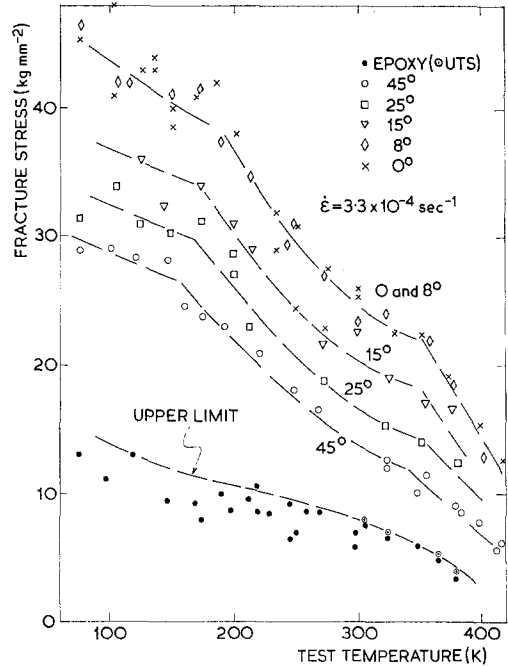


Figure 9 The temperature dependence of the fracture stress.

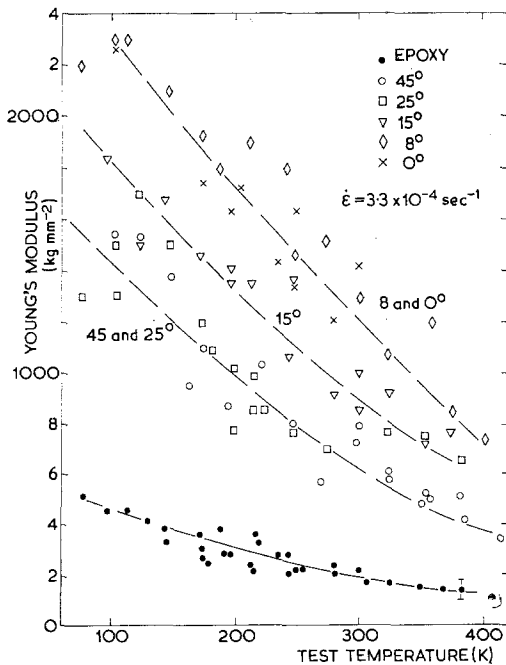


Figure 8 The temperature dependence of Young's modulus.

Fig. 10 shows the total elongation at the point of fracture, which is the sum of elastic, reversible plastic, and irreversible plastic elongation [4]. The elongation was largest near the glass-transition temperature, decreased to a minimum at about room temperature, and increased again slightly with falling test temperature. The result for unreinforced epoxy differed: the fracture elongations, which between 350 and 400 K were comparable to those of a 45° composite, decreased and at 100 K were only 1/4 to 1/3 as great. Below 200 K, the ductility (as indicated by the elongation to fracture) was markedly improved by the reinforcement, except for orientations near 0°.

3.5. Limits to the effectiveness of the reinforcement

In Section 3.1 it was briefly mentioned that the reinforcement loses its importance in the course of the deformation. This was concluded from the microscopic observation of voids in the fibres

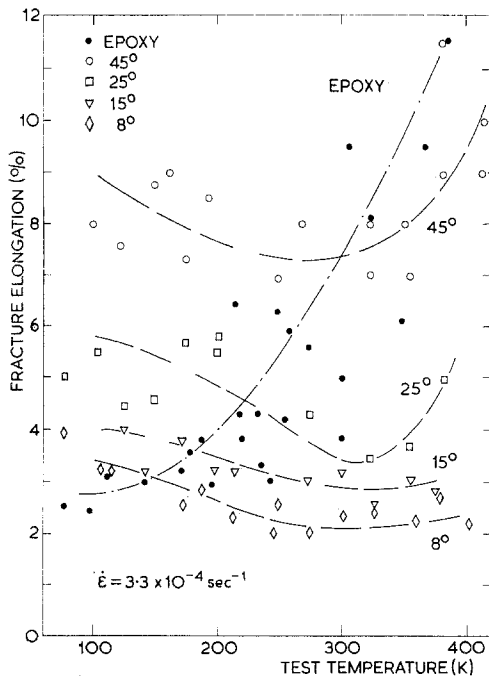


Figure 10 The temperature dependence of the fracture elongation.

and the surrounding matrix (Fig. 3). If the reinforcement were effective until the fracture occurred, the stress-strain curve would be absolutely linear – which it is not (cf. Fig. 4). Theoretical models for the stress-strain behaviour of reinforced materials are generally based on gross simplifications: unidirectional fibres are considered. Besides, they are assumed to be straight and independent of each other, and not undulated and interwoven as in our real material. What in this investigation was measured as Young's modulus, therefore, certainly is not describable by any of the existing theories. The maximum properties of a glass-epoxy composite can never be exploited in our material. Deviations from linearity in the stress-strain behaviour, observed even for 0° orientations, can only be understood by the straightening tendency of the undulated fibres. In contrast, in samples with unidirectional straight and continuous fibres ("Permaglass XE 6") of 0° orientation, where no such straightening effect should be expected, completely linear stress-strain relations with high elastic moduli ($E = 2900 \text{ kg mm}^{-2}$) and fracture stresses ($\sigma_f = 35 \text{ to } 40 \text{ kg mm}^{-2}$) could indeed be measured.

* λ , which is of special physical significance, is considered further in Part 2.

The deviations from the initial straight line in materials of the type shown in Fig. 1 have to be interpreted as being due to the increasing destruction of bonds between fibres and matrix. We have shown that the onset of this deviation depends on the ratio of sample width and length (or on w'/w , cf. Fig. 4b). This means that in short specimens (where $w'/w \rightarrow 1$) the fibres are more effective in preventing the matrix from yielding initially.

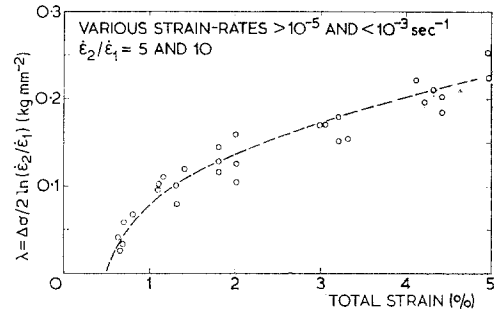


Figure 11 The strain dependence of the strain-rate sensitivity for a 45° composite, measured at room temperature.

The limiting strain at which the fibres begin to lose their effectiveness as reinforcement can be estimated by systematic observation of the strain-rate sensitivity*

$$\lambda = \Delta\sigma/2 \ln(\dot{\epsilon}_2/\dot{\epsilon}_1),$$

where $\Delta\sigma$ is the increment of stress measured when the strain-rate is increased from $\dot{\epsilon}_1$ to $\dot{\epsilon}_2$ during the course of a test. Fig. 11 shows the observed strain dependence of λ , and it is seen that λ does not increase continuously. Whether the rate sensitivity is indeed zero at strains less than about 0.5% cannot be determined because of the experimental difficulty of measuring very small stress increments. However, it is clear that λ increases rapidly at strains above 0.5%, and we may conclude that the low or even zero strain-rate sensitivity at strains below 0.5% reflects the behaviour of the glass fibres rather than that of the rate-sensitive matrix. At a given strain above 0.5%, the rate sensitivity varies with temperature, which further indicates the influence of the matrix (see Part 2).

Curves equivalent to those for the 45° composite in Fig. 11 were obtained for all orientations (ϵ_0 is nearly independent of orientation) [4]. In our opinion, these curves suggest

that the strain ϵ_0 and the corresponding stress (which equals σ_y) represent the limiting values for a good bond between fibres and matrix. From this point onward the deformation will increasingly be controlled by the matrix alone. Unreinforced epoxy can never be loaded up to stress levels as high as these because crazing occurs after some increase of stress and strain. But the fibres in reinforced epoxy prevent initial straining and stop catastrophic crack propagation and the epoxy can be studied at stresses beyond the usual fracture limit. Only after the bond between fibres and matrix has failed, the deformation continues, controlled by the matrix, indicating that the strain, not the stress, is the factor which primarily determines the failure behaviour in epoxy.

Similar conclusions concerning the fact that the matrix is rate-controlling in the later stages of deformation have been deduced from comparisons of the final strain-hardening coefficients of epoxy and composites [4].

4. Conclusions

Reinforcement clearly strengthens the epoxy resin. Increasing the strain-rate or decreasing the test temperature causes, in addition, a pronounced improvement of yield and fracture stresses. No general statement concerning the effect of temperature or strain-rate on the fracture elongation can be made, since fibre orientation and sample shape affect the results considerably. At room temperature, specimens with fibre orientations near or equal to 45° , in which the fibres do not connect the grips, have the same fracture elongation as the epoxy itself, but the elongation decreases as the fibre orientation approaches 0° . At temperatures below 200 K, however, reinforcement significantly increases

the elongation to fracture for fibre orientations near 45° . Young's modulus for the composite is strongly affected by temperature and fibre orientation, as well as by the fixity of the fibres in the grips.

It appears that initially the deformation is successfully controlled by the fibres. At stresses which exceed the fracture strength of the epoxy the crack propagation is stopped before a catastrophic failure can develop. When the bond between the fibres and matrix is weakened the properties are gradually determined by the matrix. This appears to be the basis of the variations which are observed when strength or ductility data of composites are recorded. Thus, the central interesting feature is the correlation between properties of epoxy in the unreinforced and in the reinforced state. The main argument, however, has to be based on the results of temperature and strain-rate changes, which are presented in Part 2.

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