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Critical fibre length and tensile strength for carbon fibre–epoxy composites

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The tensile strength of epoxy resin reinforced with a random-planar orientation of short carbon fibres decreases with increasing temperature. This decrease may be estimated by the strain rate and temperature dependence of both the yield shear strength at the fibre–matrix interphase and the critical fibre length obtained by taking the distribution of fibre strength into consideration. The experimental value at room temperature is smaller than the calculated value. It is inferred that this result is attributed to the stress concentration caused by ineffective fibres produced during preparation which were shorter than the critical fibre length.

1. Introduction

As is well known, loads working in the short-fibre-reinforced composites are transmitted to the fibre through the fibre–matrix interface. Consequently, mechanical properties of the composites are greatly influenced by the shear strength at the fibre–matrix interface or by the critical fibre length, which is dependent upon the shear strength. Hence, in discontinuous fibre–reinforced resins, precise determination of shear strength at the interface or critical fibre length is of great importance, and many methods to determine them have been proposed [1].

We reported a method for directly measuring critical fibre length or yield shear strength at the fibre–matrix interphase. Namely, if a sufficiently long fibre is embedded in the resin matrix and the system is elongated, the fibre eventually breaks into many pieces. By measuring the lengths of the broken pieces, the critical fibre length for the system can be estimated in both cases where the strength of fibres is assumed to be uniform [2] and where it is assumed to be variable [3, 4]. According to the latter method, we were able to explain clearly the strain rate and temperature dependence of both the yield shear strength at the interphase and the critical fibre length for glass fibre–thermoplastic resin systems [4], glass fibre–thermosetting resin systems [5], and a carbon fibre–epoxy resin system [6].

Strain rate and temperature dependence of mechanical properties, e.g. strength [7–10] and creep [11–16], of fibre-reinforced resins based on mechanical properties of the resin serving as a matrix, have also been reported.

As mentioned above, mechanical properties of composites reinforced by short fibres are strongly governed by the critical fibre length and the shear strength at the fibre–matrix interface. It therefore appears appropriate to study the strain rate and temperature dependence of the tensile strength of the composites in conjunction with the strain rate and temperature dependence of both the critical fibre length and the shear strength. There are virtually no studies of the tensile strength of composites reinforced by short fibres based on this approach.

In a preceding paper [17], we studied the tensile strength temperature dependence for glass fibre–epoxy and glass fibre–unsaturated polyester resin composites taking the temperature dependence of critical fibre length into consideration. In this work, epoxy resins reinforced with a random-planar orientation of short carbon fibres were prepared, and the tensile strength temperature dependence of the composites was investigated. Furthermore, the strain rate and temperature dependence of both the critical fibre length and the yield shear strength at the fibre–matrix interphase, as previously studied [6], was applied to the composites.

2. Experimental procedure

The carbon fibres and resins used in the preparation of the specimens were the same as those used in the previous experiment to measure critical fibre length [6].

The composites reinforced with a random-planar orientation of short fibres were prepared as reported previously [3, 17], i.e. the fibre used was a carbon fibre (Pyrofil T1, 8.83 μm diameter, Mitsubishi Rayon), and the matrix material was an epoxy resin (Epikote 828, Yuka Shell Epoxy). The surface of the carbon fibres was treated with surface treatment and sizing agents.

Next, a mat of short carbon fibres oriented random planarly was prepared as reported previously [3, 17], i.e. fibres in roving form were bundled with the use of a dilute PVA solution, dried, and cut into short fibres of uniform length by a constant-length cutter. After cutting, the PVA was dissolved and removed in water, and the short fibres were suspended in a large amount of distilled water and then allowed to settle on a filter paper placed at the bottom of the vessel. The water

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was removed by pressing at a suitable pressure. The fibres were then dried at 80 °C for 24 h. In this manner it was possible to obtain a mat in which the short fibres were oriented random-planarly and yet distributed uniformly.

The distribution of fibre length in carbon fibres randomly extracted from a mat which had received several treatments after cutting, is shown in Fig. 1. The carbon fibres had been cut sufficiently uniformly and suffered virtually no damage in the subsequent treatments. The mean fibre length, $\bar{L}$, was 0.93 mm.

The resin mixtures were prepared under the same conditions as reported in previous papers [2, 3, 5, 6, 17]. Epoxy resin, 100 parts, was mixed with 10 parts amine curing agent (S-Cure 661, Kayaku Nuri). The mixture was agitated thoroughly and then defoamed under vacuum at 30 °C for about 20 min.

A random mat was then introduced into an impregnating apparatus which was evacuated thoroughly to remove the air entrapped in the fibre mats, and the resin mixture was poured into the apparatus so that mats could be impregnated fully. Thereafter, atmospheric pressure was gradually applied to promote the resin mixture. Finally, the impregnated mats were cured at 65 °C for 17 h and then post-cured at 140 °C for 5 h. The composite was then allowed to cool to room temperature at a cooling rate of about 0.5 °C min^{-1}.

This procedure allowed preparation of bubble-free resins reinforced with a random-planar orientation of short fibres. The volume fraction of the carbon fibres could be controlled by pressing the mat before impregnation. In this experiment, the volume fraction was set at 9.9%.

The dumb-bell-shaped test specimens were cut from these composites in accordance with JIS K 7113 and subjected to tensile tests at a test strain rate of 0.03 min^{-1} with the aid of a Tensilon UTM-I-2500 (Orientec). Measurements were made at intervals of 20 °C from 20–140 °C. For each temperature level tested, 15–20 measurements were taken.

3. Results and discussion

The strength distribution of the carbon fibres used in the experiments is shown in Fig. 2.

The tensile strength of brittle fibres, such as carbon fibre used in this experiment, was variable with fibre length. As reported previously [3], the tensile strength of such fibres was generally represented by a chain model and Weibull distribution function. In this chain model, it was assumed that a fibre is constituted by a chain of n pieces of equal links, and the strength within the link is uniform.

The probability, $g(\sigma)$, that a fibre will break at a stress, σ, can be expressed as

$$g(\sigma) = n m \sigma_o^{-1} \left(\frac{\sigma - \sigma_p}{\sigma_o}\right)^{m-1} \times \exp\left[-n\left(\frac{\sigma - \sigma_p}{\sigma_o}\right)^m\right] \quad (1)$$

where σ_o, σ_p, and m are the Weibull parameters determined for the materials. The solid lines in Fig. 2 represent values calculated by substituting Weibull parameters σ_o, σ_p, m, and the number of links n given (Table I) into Equation 1. The length of the link (gauge length/number of links) is 3.00 mm. The mean strength, $\bar{\sigma}_{f,l}$, of the links is found to be 3.36 GPa by using the following equation [3]

$$\bar{\sigma}_{f,l} = \sigma_p + \sigma_o \Gamma\left(\frac{m+1}{m}\right) \quad (2)$$

where Γ is the complete gamma function. Other properties of carbon fibre and epoxy resin used in this experiment are shown in Table II.

The relationship between tensile strength and temperature is shown in Fig. 3. For comparison, the tensile strength of epoxy resin is also plotted. The strength decreases with increasing temperature.

We showed in the preceding papers [3, 17] that the tensile strength, $(\sigma_{c,s})_T$, of a composite in which short fibres are oriented random-planarly depends strongly on the yield shear strength, τ, at the fibre–matrix interphase and is written as

$$[\sigma_{c,s}]_T = \frac{2\tau}{\pi}\left\{2 + \ln\left[\frac{(1 - l_c/2L)\sigma_f \sigma_m V_f + \sigma_m \sigma'_m V_m}{\tau^2}\right]\right\} - [\sigma_r]_T \quad L \geqq l_c \quad (3a)$$

$$[\sigma_{c,s}]_T = \frac{2\tau}{\pi}\left\{2 + \ln\left[\frac{(L/d)\sigma_m V_f + \sigma_m^2 V_m}{\tau^2}\right]\right\} - [\sigma_r]_T \quad L < l_c \quad (3b)$$

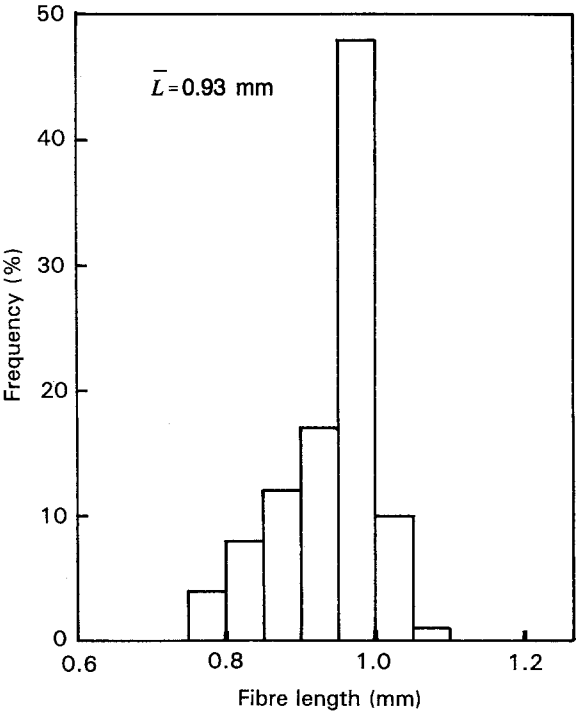

Figure 1 Frequency distribution of short carbon fibres.

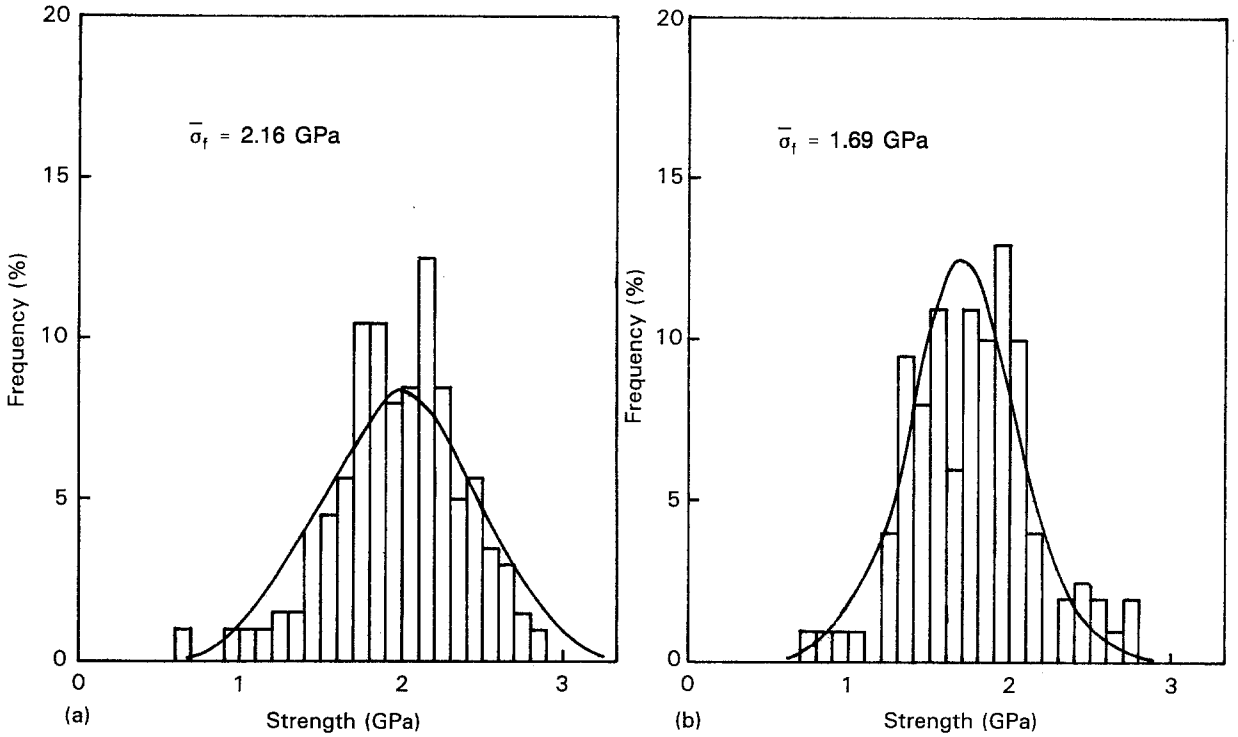

Figure 2 Strength distribution of carbon fibres: (a) test length 25 mm; (b) test length 100 mm; (—) curve calculated from Equation 1.

TABLE I Statistical values of strength for carbon fibres

Test length (mm)	Mean value, $\bar{\sigma}_{f,l}$ (GPa)	Weibull parameters			Number of links, n
		σ_o (GPa)	σ_p (GPa)	m	
25	2.16				8.3
100	1.69	2.88	0.77	3.4	33.0
Link	3.36				1.0

TABLE II Properties of materials (at 20 °C)

Materials	Tensile strength (GPa)	Young's modulus (GPa)	Thermal expansion coefficient ($°C^{-1}$)
Carbon fibre	1.69[a]	216.7[a]	9.5×10^{-6} [b]
Epoxy resin	0.0667	1.90	5.9×10^{-5}

[a] Test length = 100 mm.
[b] Perpendicular direction to fibre axis.

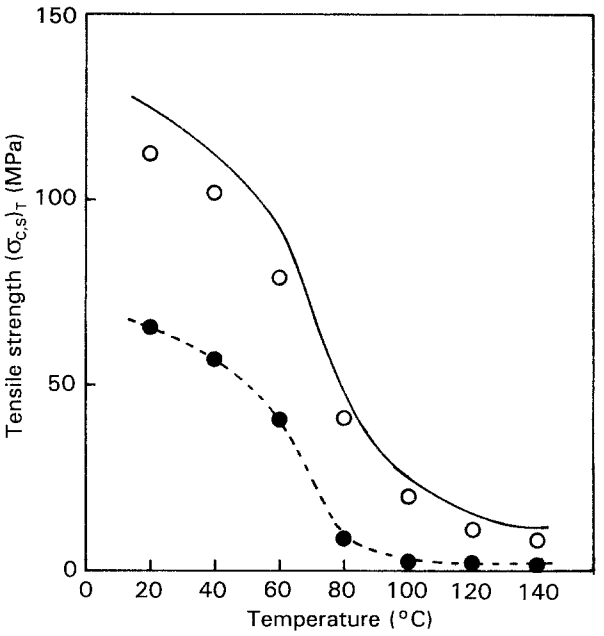

Figure 3 Relationship between temperature and tensile strength for epoxy resin and epoxy–short carbon fibre composites: (○) composite, (– – ●– –) resin, (—) calculated value from Equation 3.

where L is the fibre length, d the fibre diameter, l_c the critical fibre length, σ_f the tensile strength of the fibre, σ_m the tensile strength of the matrix, V_f the fibre volume fraction, V_m the matrix volume fraction, and σ'_m the matrix stress at the fracture strain of the fibre. $[\sigma_r]_T$ is the thermal stress produced during moulding of the composite by the difference in the thermal expansion coefficient between fibre and matrix resin, and is given by

$$[\sigma_r]_T = \frac{2(\alpha_m - \alpha_f) E_m \Delta T}{(1 + \nu_m) + (1 + \nu_f)(E_m/E_f)} \quad (4)$$

where α is the thermal expansion coefficient, E is Young's modulus, ν is Poisson's ratio, ΔT is the difference in temperature from the moulding temperature, and subscripts m and f represent matrix and fibre, respectively.

From Equations 3a and b, it is clear that we must obtain the yield shear strength, τ, at the fibre–matrix interphase and the critical fibre length, l_c, to calculate the tensile strength, $[\sigma_{c,s}]_T$.

In the preceding papers [5, 6], we discussed the strain rate and temperature dependence of both the yield shear strength and the critical fibre length. For

the carbon fibre–epoxy resin system, the yield shear strength, $\tau_{\dot{\varepsilon},T}$, at the fibre–matrix interphase at given strain rate, $\dot{\varepsilon}$, and temperature, T, was given as

$$\tau_{\dot{\varepsilon},T} = K_1 + K_2 \log(\dot{\varepsilon}\, a_T) \quad (5)$$

where a_T is the shift factor, K_1 and K_2 are constants depending upon the materials constituting the composites, the adhesive state at the fibre–matrix interface, and the reference temperature. In this experimental system, K_1 is 41.1 MPa, K_2 is 2.42 MPa [6].

Mean critical fibre length, $(\bar{l}_c)_{\dot{\varepsilon},T}$, under the same conditions, using $\tau_{\dot{\varepsilon},T}$ given by Equation 5, was given as

$$(\bar{l}_c)_{\dot{\varepsilon},T} = \frac{\bar{\sigma}_{f,l}\, d}{2\tau_{\dot{\varepsilon},T}} \quad (6)$$

where d is the fibre diameter, $\bar{\sigma}_{f,l}$ is the mean value of strength of the links constituting the fibre and calculated from Equation 2.

$\tau_{\dot{\varepsilon},T}$ and $(\bar{l}_c)_{\dot{\varepsilon},T}$ obtained from Equations 5 and 6, respectively, at the strain rate ($\dot{\varepsilon} = 0.03\ \text{min}^{-1}$) in this experiment, are shown in Fig. 4. As mentioned above, the reinforcing fibre mean length, $\bar{L}$, is 0.93 mm. Therefore, experiments in which the reinforcing fibre length is shorter than the critical fibre length below 100 °C, and experiments utilizing reverse conditions above 120 °C, can be performed.

The solid lines in Fig. 3 represent calculated values of $[\sigma_{c,s}]_T$ obtained from Equations 3 and 4 by substituting the yield shear strength, $\tau_{\dot{\varepsilon},T}$, and the mean critical fibre length, $(\bar{l}_c)_{\dot{\varepsilon},T}$ (Fig. 4), into Equation 3 for τ and l_c. In this calculation the values of σ_m are shown in Fig. 3, experimental values of σ'_m, E_m, α_m and ν_m at various temperatures are shown in Figs 4 and 5. We employed the mean fibre length, $\bar{L}$, for L, the mean strength $\bar{\sigma}_{f,l}$ (= 3.36 GPa), of the links for σ_f, because the reinforcing fibre length used in this experiment was 0.93 mm. We also assumed that $\bar{\sigma}_{f,l}$ was constant in the experimental temperature range. Then, we employed Young's modulus, E_f, the thermal expansion coefficient, α_f, and Poisson's ratio, ν_f (= 0.23), as shown in Table II, and assumed these values were also constant in the temperature range of this study. The experimental values approximately agreed with the calculated values. The difference in tensile strength between the experimental value of composites and the calculated value, which takes the strain rate and temperature dependence of τ and l_c into consideration, was smaller than that for glass–epoxy composites [17], in which the temperature dependence of τ and l_c only was considered. It was therefore possible to estimate the tensile strength of the composites using Equations 3 and 4, which utilized the strain rate and temperature dependence of both the yield shear strength and the critical fibre length obtained by taking the distribution of fibre strength into account for τ, which is the most important factor of Equation 3.

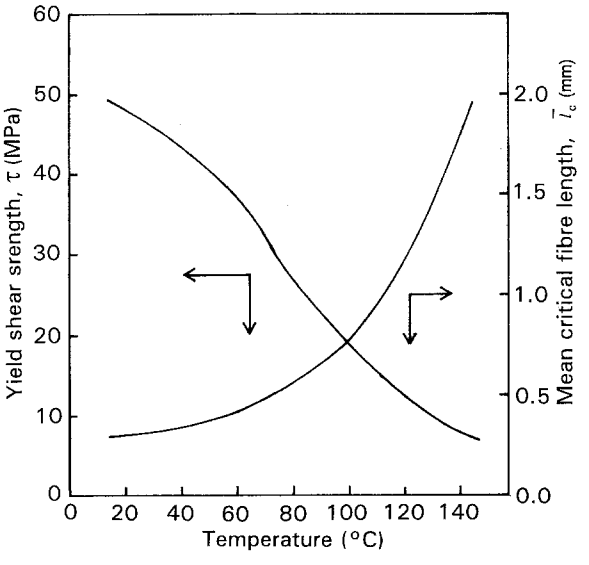

Figure 4 Temperature dependence of yield shear strength and mean critical fibre length calculated from Equations 5 and 6.

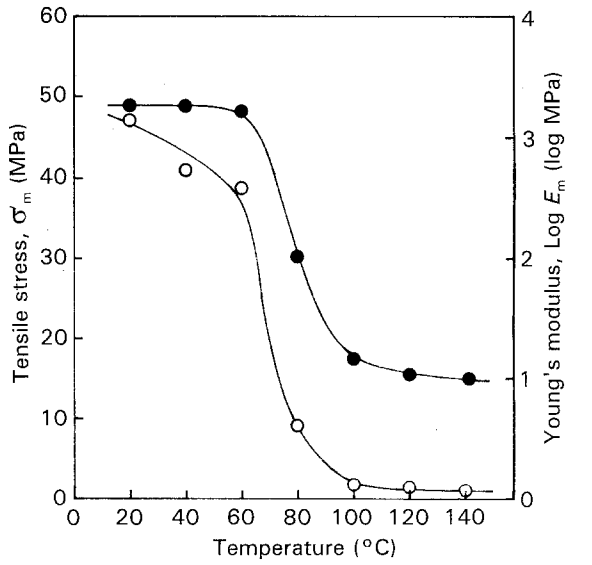

Figure 5 Temperature dependence of (○) the tensile stress of the matrix at the fracture strain of the composite and (●) the Young's modulus of the matrix.

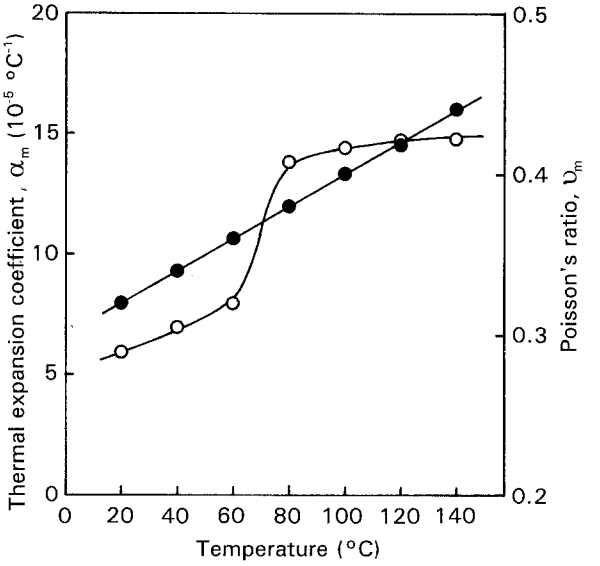

Figure 6 Temperature dependence of (○) the thermal expansion coefficient and (●) Poisson's ratio of the matrix.

The tensile strength of the composites at room temperature was smaller than the calculated value. A possible cause for this was stress concentration produced by the pores at the side of the specimen formed during preparation. The dumb-bell-shaped test specimens were cut in accordance with JIS. The fibres embedded in the neighbourhood of the side of the specimen were cut and resulted in a length which was shorter than the critical fibre length. Hence, these fibres did not play any role in reinforcement. The embedded positions in such fibres acted as pores. The tensile strengths of the composites were influenced by them as if there were many cracks. The effect of the cracks on the tensile strength was larger in brittle materials. Young's modulus of the epoxy resin used in this experiment at room temperature was larger by two figures than that at a higher temperature (Fig. 5), and the fracture strain was smaller. Therefore, epoxy resin exhibited a ductile fracture at a higher temperature, but the brittle fracture at room temperature. As a result, crack sensibility was significant at room temperature. It was inferred that this result could be attributed to the fact that tensile strengths expected from Equations 3 and 4 were not realized.

4. Conclusion

Epoxy resin reinforced with a random-planar orientation of short carbon fibres was prepared, and the temperature dependence of the tensile strength of composites was examined, taking the strain rate and temperature dependence of both the yield shear strength at the fibre–matrix interphase and the critical fibre length into consideration.

The tensile strength of composites decreases with increasing temperature. This decrease may be estimated by the strain rate and temperature dependence of both the yield shear strength at the fibre–matrix interphase and the critical fibre length obtained by a method proposed in the preceding paper [6], taking the distribution of fibre strength into consideration. The experimental value at room temperature is smaller than the calculated value. It is inferred that this result is attributed to the stress concentration caused by ineffective fibres produced during preparation which were shorter than the critical fibre length.

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