

Temperature Dependence of Critical Fiber Length for Glass Fiber-Reinforced Thermosetting Resins

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Synopsis

In discontinuous fiber-reinforced composites, the critical fiber length plays an essential role in determining the mechanical properties. A method was devised to accurately determine the critical fiber length and the temperature dependence of the critical fiber length was studied for glass fiber-epoxy and glass fiber-unsaturated polyester resin composites. If a continuous glass fiber is embedded in the matrix and the system is subjected to a tensile strain greater than the fiber ultimate tensile strain, the fiber breaks into many pieces. If the average length of these broken pieces (\bar{l}) is measured, the critical fiber length (l_c) is expressed as $l_c = 4/3\bar{l}$. The critical fiber length greatly increases with increasing temperature and the apparent shear strength at the interface, calculated from the critical fiber length, decreases linearly with increasing temperature.

INTRODUCTION

Discontinuous fiber-reinforced resins are isotropic, easily molded. In any discontinuous fiber-reinforced resin, loads working on the composite are transmitted to the fiber through the fiber-matrix interface. Consequently, the strength of the composite is greatly influenced by the shear strength at fiber-matrix interface or by the critical fiber length, which is dependent upon the shear strength. Hence, the shear strength at interface or the critical fiber length plays an essential role in determining the reinforcing effect in discontinuous fiber-reinforced resins.

Studies have been conducted on experimental methods for determining the critical fiber length¹⁻³; however, all of these methods are indirect methods and do not permit accurate measurements, nor give good reproducibility. The properties of resins which form the matrix of composites are sensitive to temperature and it is conceivable that the critical fiber length will be affected by temperature.

In this paper, efforts were made to determine accurately the critical fiber length for fiber-resin composites and the temperature dependence of the critical fiber length was studied for the glass fiber-epoxy and glass fiber-unsaturated polyester resin composites.

MEASUREMENTS

Principle

If a sufficiently long fiber is embedded in the resin matrix and the system is elongated [Fig. 1(a)], the fiber eventually breaks into many pieces [Fig. 1(b)]. Measuring the lengths of the broken pieces, the critical fiber length can be estimated as follows:

If the system is elongated [Fig. 1(a)], the stress transferable to the fiber (σ_x) at a distance (x) from the fiber end is given by the following equation according to Kelly et al.,^{1,4} while assuming that the shear strength (τ) at the fiber-matrix interface is constant:

$$\sigma_x = (4\tau/d)x \tag{1}$$

where d is the diameter of the fiber. When the specimen is elongated further, the tensile stress of the fiber (σ_x) increases until it reaches the ultimate tensile strength of the fiber (σ_f). If the value of x at this particular point is designated as x_0 , the following relationship results:

$$\sigma_f = (4\tau/d)x_0 \tag{2}$$

The fiber breaks at any point between A and B , A or B being away from either end of the fiber by x_0 [Fig. 2(b)]. In case a broken piece of fiber exceeds $2x_0$ in length, the breakage repeats itself by the same mechanism as above. When all the broken pieces are reduced to less than $2x_0$ in length, any further elongation of such pieces does not cause the tensile stress transferable to the fiber (σ_x) to reach the ultimate tensile strength (σ_f) and hence, no further disintegration

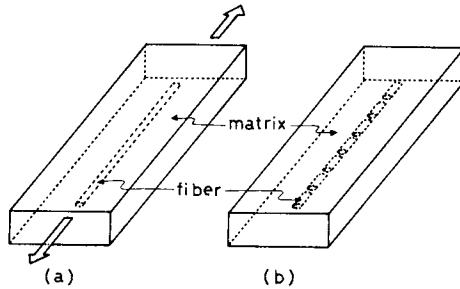


Fig. 1. Schematic diagram of model used in measuring the critical fiber length.

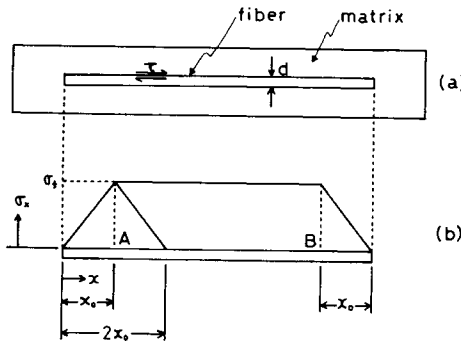


Fig. 2. Schematic diagram of the stress distribution along a fiber.

occurs. The length of broken fiber pieces (l) should be distributed evenly in the range $x_0 \leq l \leq 2x_0$ and the average value (\bar{l}) is given as follows:

$$\bar{l} = \frac{1}{2}(x_0 + 2x_0) = \frac{3}{2}x_0 \quad (3)$$

Since x_0 is the length needed to make the stress of fiber (σ_x) reach the ultimate tensile strength (σ_f), the critical fiber length (l_c) is equal to $2x_0$ or $l_c = 2x_0$. Introduction of this relationship into eq. (3) gives the following expression for the critical fiber length (l_c):

$$l_c = \frac{4}{3}\bar{l} \quad (4)$$

It is therefore possible to obtain the critical fiber length (l_c) from eq. (4) once the average length of the final broken pieces (\bar{l}) is determined. Then, the apparent shear strength at the fiber-matrix interface (τ) is given as follows:

$$\tau = \sigma_f \cdot d/2l_c \quad (5)$$

Preparation of Specimens

The fiber used in this study was 'E' glass fiber (R2220 MA 859X L16, Asahi Fiber Glass, 11.67 μm in diameter) and matrix materials were an epoxy resin (Epikote 828, Mitsubishi Yuka) and an unsaturated polyester resin (Rigolac 2004 WM-2, Showa Kobunshi) (Table I).

The glass fiber was boiled in distilled water for 10 min and dried at 80°C for 8 hr. To prepare specimens differing in bond strength at the interface, the glass fiber was immersed in a 3% toluene solution of a silane coupling agent (KBM 403, Shinetsu Chemical) or in a 6% toluene solution of a release agent (KS 707, Shinetsu Chemical) for 48 hr, air dried and then dried at 70°C for 8 hr.

The epoxy resin was mixed with 10 parts of an amine hardening agent (S-Cure 661, Nihon Kayaku) or the unsaturated polyester resin was mixed with 2 parts of a hardening catalyst MEKPO (Permek N, Nihon Yushi). The mixture was agitated thoroughly and then defoamed in vacuum for approximately 20 min. This mixture was poured into a mold holding a glass fiber in the center and subjected to curing at 65°C for 17 hr and post curing at 140°C for 5 hr. The dimension of the mold resulted in a specimen which measured 0.5 mm in thickness, 20 mm in width and 150 mm in length.

The specimens prepared in this manner were submitted to measurement of the critical fiber length: each specimen was subjected to a tensile strain greater than the fiber ultimate tensile strain at a test rate of 2 mm/min at various temperatures (Table II and Fig. 3). The fiber is broken into a large number of fragments. Figure 4 illustrates typical distribution of fiber fragment length and the epoxy resin used as a matrix. Similar results were obtained for the system using the unsaturated polyester resin. Moreover, the same tendency was noted at various temperatures.

TABLE I
The Properties of Materials

| Materials | Tensile strength (dyn/cm ²), 20°C | Young's modulus (dyn/cm ²), 20°C |
|-----------------------------|--|---|
| Glass fiber | 2.81×10^{10} | 8.75×10^{11} |
| Epoxy resin | 6.81×10^8 | 1.90×10^{10} |
| Unsaturated polyester resin | 4.31×10^8 | 2.41×10^{10} |

TABLE II
Temperature Used in Measuring the Critical Fiber Length

| Matrix | Interface condition | Temperature (°C) |
|-----------------------------|---------------------|---------------------------|
| Epoxy resin | good bonding | 40, 60, 80, 100, 110, 120 |
| | poor bonding | 40, 60, 80, 100, 110, ... |
| Unsaturated polyester resin | good bonding | ..., 60, 70, 80, 90, 100 |
| | poor bonding | 50, 60, 70, 80, 90, ... |



Fig. 3. Typical breaking pattern of a single fiber embedded in the epoxy resin. (Good bonding.)

RESULTS AND DISCUSSION

The relationship between the critical aspect ratio (critical fiber length \div diameter) and the temperature is shown for the epoxy-glass fiber and unsaturated polyester-glass fiber systems (Figure 5). Regardless of the nature of interfacial treatment, the critical aspect ratio increases greatly with increasing temperature. Moreover, for both systems, the specimens treated with the release agent show larger critical aspect ratios than those treated with the coupling agent. The

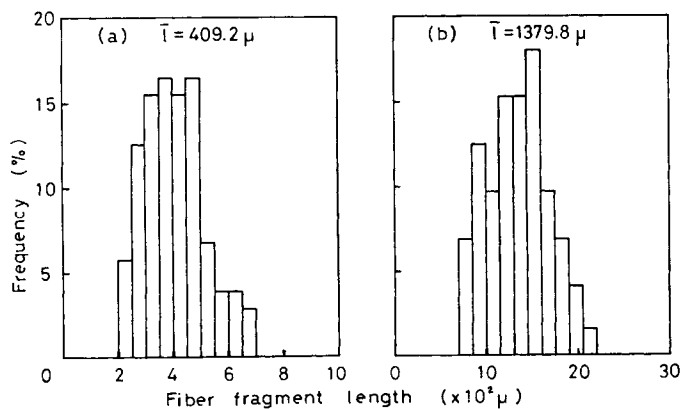


Fig. 4. Distribution of fiber fragment (a) at 40°C, (b) at 100°C (good bonding) (epoxy-glass fiber system).

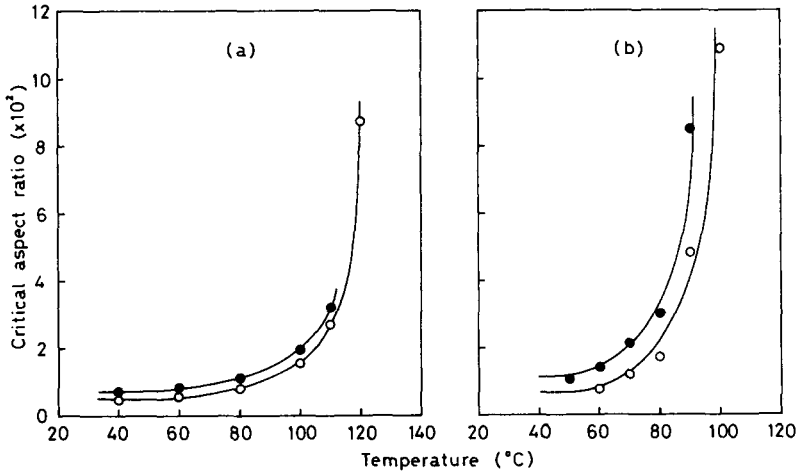


Fig. 5. Relation between temperature and critical aspect ratio. (a) Epoxy-glass fiber system, (b) unsaturated polyester-glass fiber system; (—○—) good bonding; (—●—) poor bonding.

relationship between the apparent shear strength at the interface obtained from the critical aspect ratio l_c/d according to eq. (5) and the temperature is shown in Figure 6. The apparent shear strength at the interface decreases linearly with increasing temperature.

It is conceivable that a thermal stress produced by the difference in thermal expansion coefficient between glass fiber and resin may cause the temperature dependence of the apparent shear strength. Therefore, a study was undertaken on thermal stress produced by embedding a fiber in the resin.

When a fiber is embedded in the resin and the system is allowed to cure, the pressure (P) working perpendicularly on the fiber-resin interface is given by the following equation⁵:

$$P = \frac{(\alpha_m - \alpha_f)E_m \cdot \Delta T}{(1 + \nu_m) + (1 + \nu_f)(E_m/E_f)} \tag{6}$$

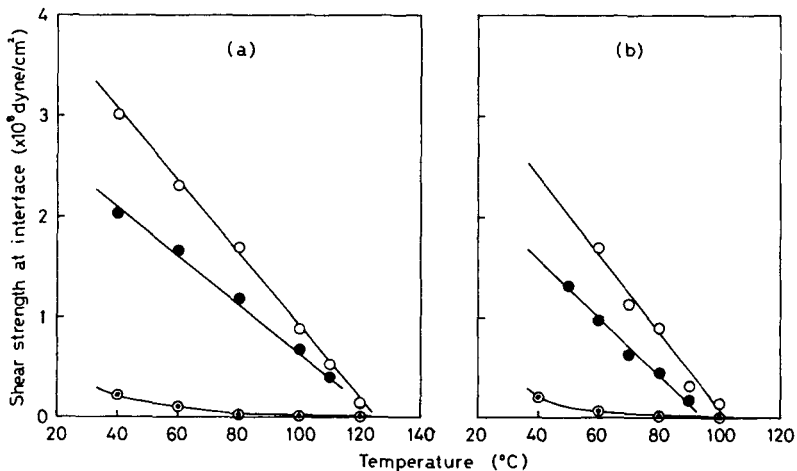


Fig. 6. Relation between temperature and apparent shear strength. (a) Epoxy-glass fiber interface, (b) unsaturated polyester-glass fiber interface; (—○—) good bonding; (—●—) poor bonding; (—○—) frictional force.

where α is the thermal expansion coefficient, E is the Young's modulus, ν is the Poisson's ratio, ΔT is the difference in temperature from the molding temperature and the subscripts m and f denote matrix and fiber, respectively. In glass fiber-resin systems such as used here, $\alpha_m \gg \alpha_f$ and $E_f \gg E_m$. Therefore,

$$P \doteq (\alpha_m \cdot E_m \cdot \Delta T)/(1 + \nu_m) \quad (6')$$

On the assumption that the apparent shear strength at the interface is based only on a physical frictional force produced by this pressure at interface, such a frictional force τ is given as follows using the friction coefficient μ :

$$\tau' = \mu P \doteq (\mu \cdot \alpha_m \cdot E_m \cdot \Delta T)/(1 + \nu_m) \quad (7)$$

In order to determine the pressure at the interface at varying temperatures, the thermal strain and the Young's modulus were determined at varying temperatures. The thermal strain was determined with a strain gauge using a linear expansion tester manufactured on a trial basis in the author's laboratory. The thermal expansion coefficient was obtained for a given temperature from the slope of the curve (Figure 7). The pressure at interface (P) was calculated from the thermal expansion coefficient (α_m) and Young's modulus (E_m) (Fig. 8) and the values of frictional force (τ') obtained from P according to eq. (7) are shown in Figure 6. In this instance, it was assumed that $\nu_m = 0.3$ and $\mu = 0.3$ for both resins.

The frictional force due to the pressure at the interface is numerically always 10% or less of the apparent shear strength at the interface obtained in the experiments on the critical fiber length (Fig. 6). Therefore, the phenomenon that

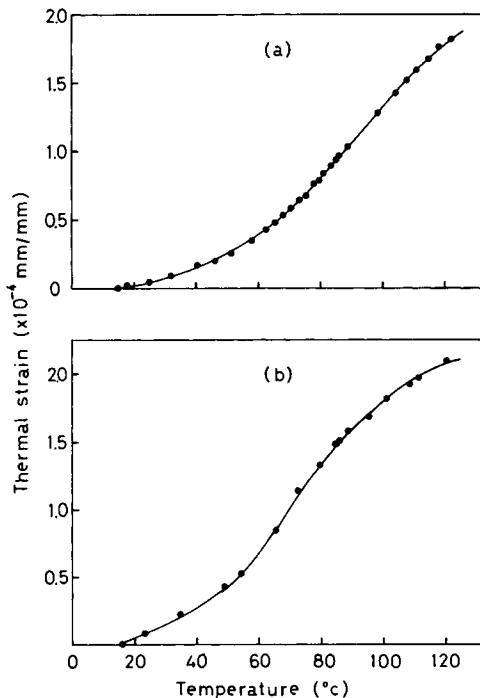


Fig. 7. Relation between temperature and thermal strain. (a) Epoxy resin, (b) unsaturated polyester resin.

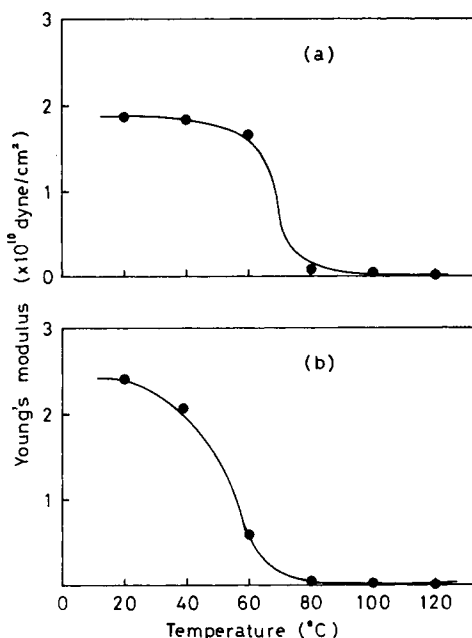


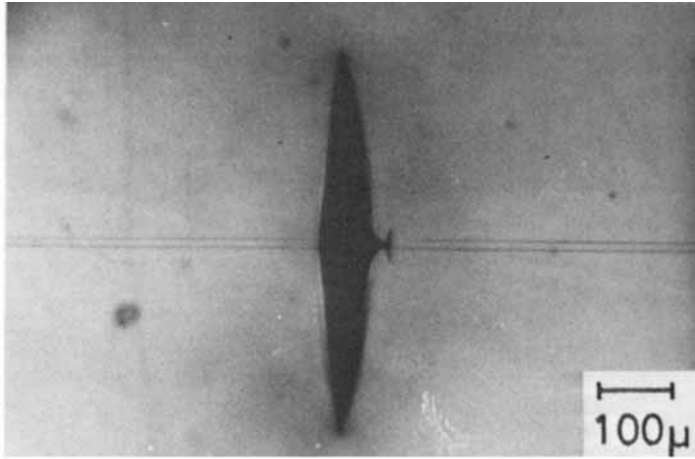
Fig. 8. Relation between temperature and Young's modulus. (a) Epoxy resin, (b) unsaturated polyester resin.

the apparent shear strength at the interface decreases with increasing temperature in the manner shown in Figure 6 cannot be explained solely by relaxation of the pressure at the interface due to thermal expansion of the matrix. This would hold true even if the friction coefficient were much larger, for example, $\mu = 1$.

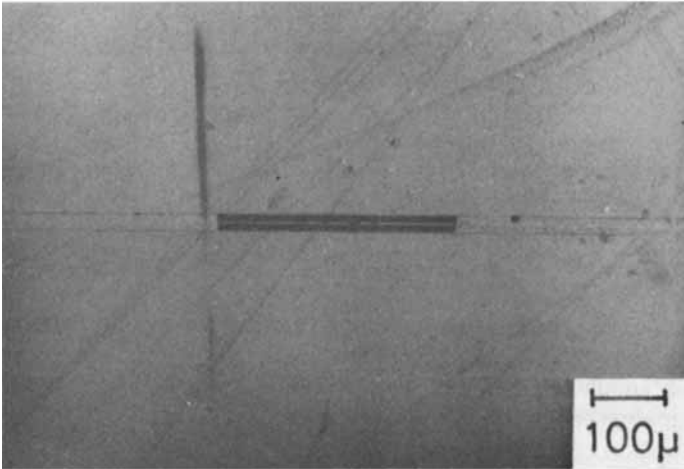
The effect of the compression stress working in the axial direction of fiber on the critical fiber length is conceivable. However, this compression stress is considerably smaller than σ_f and the temperature dependence of the critical fiber length (namely, the apparent shear strength at interface) cannot be accounted for by relaxation of compression stress.

It is also conceivable that the secondary bond existing at the glass fiber-matrix interface undergoes relaxation with increasing temperature and, as a result, the apparent shear strength at the interface decreases. It is further suggested that the temperature dependence of the apparent shear strength at the interface is not brought about by a decrease in bond strength at the fiber-matrix interface, but by a decrease in shear strength of the matrix with increasing temperature. That is, if the bond strength of the fiber-matrix is sufficiently great and greater than the shear strength of the matrix, the apparent shear strength (τ) obtained on the critical fiber length is not the shear strength at the interface, but the shear strength of the matrix.

It is possible to determine which mechanism is responsible for the temperature dependence of the apparent shear strength (τ) found in this study by observation of the breaking point of a fiber in the specimen and the fracture surface of the actual composite. For example, Figure 9 shows examples of optical photomicrographs of the breaking point of a fiber used in the experiments on the critical fiber length. The specimen treated with the coupling agent in Figure 9(a) reveals a generation of cracks in the direction perpendicular to the fiber at the breaking



(a)



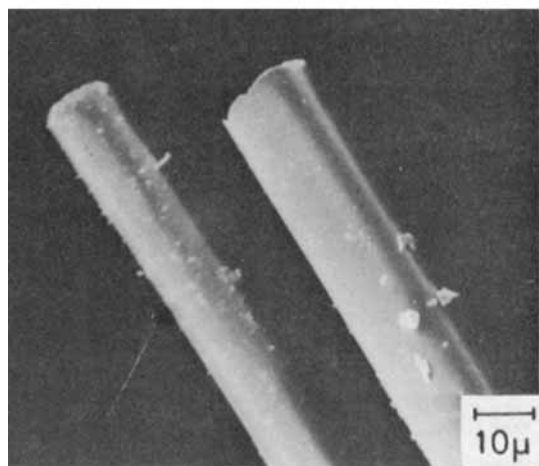
(b)

Fig. 9. Optical photomicrographs of the breaking point of a single glass fiber embedded in the epoxy resin. (a) Good bonding at 80°C, (b) poor bonding at 80°C.

point. On the other hand, the specimen treated with the release agent [Fig. 9(b)] does not reveal cracks in the matrix and debonding of the fiber is noted along the fiber-matrix interface. Figure 10 shows examples of scanning electron photomicrographs of the fracture surface of the discontinuous glass fiber-reinforced epoxy resin composite. The specimen treated with the coupling agent shows adhesion of the resin phase over nearly the entire surface of the fiber, which indicates that the resin phase failed by cohesion when the fiber was removed. In contrast, in the specimen treated with the release agent, the fiber is practically free from the resin phase, which indicates that it was debonded from the interface when removed. Figures 9 and 10 show the system in which the epoxy resin is used as matrix. Similar results were obtained for the system using the unsaturated polyester resin. Moreover, the same tendency was noted at various temperatures.



(a)



(b)

Fig. 10. Electron scanning photomicrographs of the fracture surface of the epoxy-short glass fiber composite. (a) Good bonding, (b) poor bonding at 100°C.

It is, therefore, inferred that the temperature dependence of the apparent shear strength at the interface for the specimen treated with the coupling agent is primarily attributed to a decrease in shear strength of the matrix with increasing temperature; while that of the specimen treated with the release agent is mainly due to a decrease in shear strength of the matrix, and relaxation of thermal stress and secondary bonding at the fiber-matrix interface with increasing temperature.

CONCLUSIONS

Attempts were made to accurately measure the critical fiber length, which plays an essential role in determining the reinforcing effect in discontinuous fiber-reinforced resins. The temperature dependence of the critical aspect ratio was examined for glass fiber-epoxy and glass fiber-unsaturated polyester resin composites. The findings are as follows:

If a continuous glass fiber is embedded in the matrix and the system is subjected to a tensile strain greater than the fiber ultimate tensile strain, the fiber breaks into many fragments. If the average length of such broken fragments (\bar{l}) is measured, the critical fiber length (l_c) is expressed as $l_c = 4/3\bar{l}$.

The critical aspect ratio greatly increases with increasing temperature and the apparent shear strength at the interface, calculated from the critical aspect ratio, decreases linearly with increasing temperature within the experimental range. This decrease in apparent shear strength at the interface may be accounted for mainly by: (1) A decrease in shear strength of the matrix with increasing temperature when the surface of glass fibers is treated with the silane coupling agent, and (2) a decrease in shear strength of the matrix and relaxation of thermal stress and secondary bonding at the interface with increasing temperature when the surface of glass fibers is treated with the release agent.

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Received April 8, 1977