JOURNAL OF MATERIALS SCIENCE 23 (1988) 1182–1188

Excellent flexural properties of aminimide-cured epoxy resin as a matrix for mica-dispersed polymer composites

S. INUBUSHI, T. IKEDA, S. TAZUKE*
Research Laboratory of Resources Utilization, Tokyo Institute of Technology, 4259 Nagatsuta, Midori-ku, Yokohama 227, Japan

T. SATOH, Y. TERADA
NTN Toyo Bearing Co. Ltd, 1380 Higashikaizuka, Iwata, Shizuoka 438, Japan

Y. KUMAGAI
Industrial Products Research Institute, 1-1-4 Yatabe-machi, Higashi, Tsukuba-gun, Ibaraki 305, Japan

The flexural behaviour of mica-dispersed epoxy resin composites has been examined. The flexural strength and flexural modulus have been determined as a function of the volume fraction of mica flakes (V_f) for both aminimide-cured epoxy resin matrix and a conventional epoxy resin reference matrix. On the basis of microscopic observation of fractured surfaces, the effect of improving the particle–matrix interface has been analysed using the modulus reduction factor (MRF) in a modified form. It is found that there is a steady increase in the flexural modulus with the volume fraction of mica flake for the aminimide-cured epoxy resin matrix. In contrast, the increase in flexural modulus levels off at a high content of filler for the reference samples. It is noteworthy that the intact mica flakes without surface treatment exhibit a substantial reinforcing effect on the flexural strength in the case of aminimide-cured epoxy resin composites. A further surprise is the difference among the curing agents used. The reference epoxy resins behave just like conventional matrix resins, exhibiting 30 to 40% reduction in the flexural strength when a small fraction of mica is added. These superior properties of the matrix resin for the composites are ascribed to the characteristics of aminimide-cured epoxy resins such as hardness, toughness, and excellent adhesivity.

1. Introduction

The need for new materials has been a drive for developing better polymer composites. Although lightweight high-performance engineering plastics are taking over from metals and ceramics in many applications, compositing technology is expected to improve further the performance of presently available polymeric materials. Particle-reinforced plastics have a number of advantages over fibre-reinforced plastics such as excellent processability, low cost of filler, and versatility of the shape of product. A serious drawback is, however, the fact that a reinforcing effect of the filler on fracture strength is hardly ever obtained. While the Young's modulus of neat resin is increased by compositing with inorganic fillers such as glass beads, mica flakes, talc powder and so forth, the fracture strength is generally less than that of the neat resin. There are a few reports on trials of improving the fracture strength by means of heavy surface treatment of the filler [1, 2]. A heavy dose of expensive silane couplers is a tentative solution to the problem.

In principle, a good reinforcing effect can be expected if the adhesion between filler surface and matrix resin is strong enough so that failure at the filler–matrix boundary is avoided, while the matrix resin should be tough enough to allow the dissipation of mechanical stress to release stress concentrations. To achieve this target, the required properties for the matrix resin are hardness, toughness and high adhesive force.

Recently we developed a new type of one-part epoxy resin using aminimide compounds as latent curing agents which release tertiary amines and isocyanates above 130° C. The epoxy resin (Epikote 828) cured with various aminimides exhibited peculiar but attractive mechanical properties such as high fracture energy, high elongation and good adhesion, all of which are suitable for the present purpose [3]. We have already used this material as a binder of solid lubricant and succeeded in demonstrating composite materials having a very low friction coefficient and low rate of abrasion [4]. These excellent properties were attributed to the toughness and good adhesivity of this specific epoxy resin binding the lubricant particles tightly. Reference experiments with conventional epoxy resins gave generally poor results. The merits

*To whom correspondence should be addressed.

0022–2461/88 $03.00 + .12

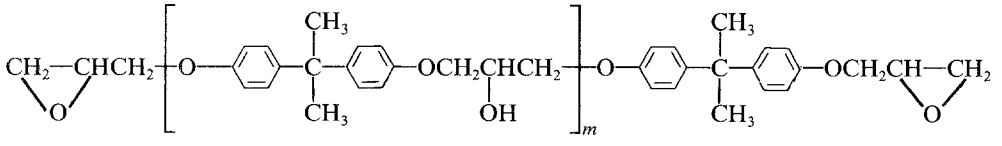
of tough and strong epoxy resin can claim wide applicability to composite materials in general.

In this paper, we report the mechanical properties of epoxy resin–mica composites in which enhancement of both fracture strength and flexural modulus is observed without any surface treatment of the filler.

2. Experimental procedure

2.1. Materials

Epikote 828 (Yuka Shell Epoxy, Tokyo) was used as an epoxy prepolymer. Approximate compositions (degree of polymerization) were determined by gel permeation chromatography (GPC) (Toyo Soda, Tokyo, HCL-802; column, GMH6 × 2 + G4000H8 + G2500H8; eluent, chloroform). GPC indicated that Epikote 828 was composed of $m = 0$ (86%, molecular weight 340) and $m = 1$ (14%, molecular weight 620) components. The structure is shown below:



The structures of the aminimide compounds (*1* and *2*) used in this study as latent curing agents for epoxide are shown in Fig. 1. These aminimide compounds were synthesized as reported previously [5]. Boron trifluoride monoethylamine complex (Hashimoto Kasei, Tokyo) (*3*) and p,p′-diaminodiphenylmethane (Tokyo Kasei, Tokyo) (*4*) used as reference curing agents were the purest grade commercially available and were used without further purification.

2.2 Methods

2.2.1. Preparation of samples

The curing conditions of each sample are described in the captions to Figs 2 to 5 below. Epoxy resin–mica composites were prepared from Epikote 828 (10 mol), curing agents (1 mol for *1*,*2*, *3*; 5.8 mol for *4*) and mica

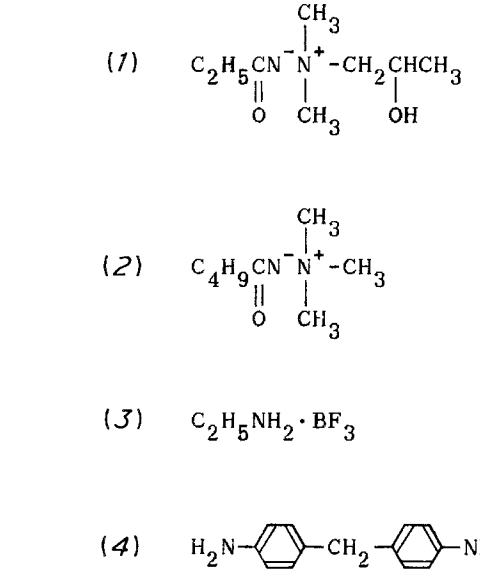


Figure 1 Structure of curing agents for epoxide: (*1*, *2*) aminimide compounds; (*3*, *4*) reference curing agents.

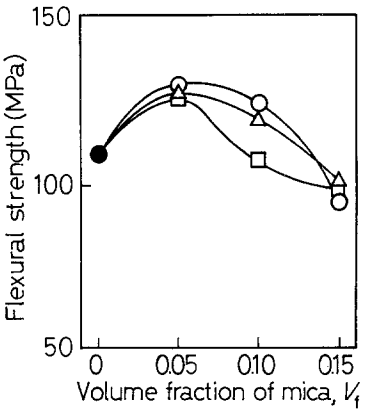


Figure 2 Flexural strength dependence on volume fraction of mica flake for aminimide (*1*)-cured epoxy resin composites. (●) Neat resin: Epikote 828 (10 mol) + *1* (1 mol), 150° C (2 h) + 110° C (5 min). Mica flakes: (○) $D = 16\,\mu m$, $\alpha = 35$; (△) $D = 60\,\mu m$, $\alpha = 55$; (□) $D = 190\,\mu m$, $\alpha = 80$.

flake (phlogopite S-60, S-200, S-325, Canada Mica, Okayama). Characteristics of these mica flakes: particle size distribution, average particle size (D) and average thickness and average aspect ratio (α), are listed in Table I. Volume fractions of the mica flake (V_f) ranged from 0 to 0.15. The curing agents and the mica flake were thoroughly dried before mixing. The mixtures were degassed under reduced pressure at 70° C for 2 h. After degassing, the mixtures were poured into moulds and cured at 80 to 150° C, cooled slowly to room temperature and finally annealed at 110° C for 5 min to give the composite materials.

2.2.2. Flexural measurements

The specimen size for the flexural measurements was 40 mm length × 5 mm width × 1 mm thickness. The flexural properties were evaluated in three-point flexural loading (span length, 20 mm; crosshead speed, 4 mm min^{-1}) using an Instron tester according to the JIS-K7203 procedure. Maximum flexural strength

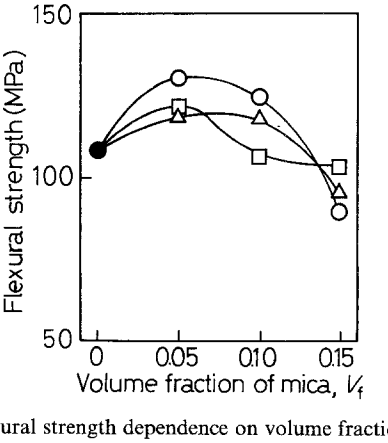


Figure 3 Flexural strength dependence on volume fraction of mica flake for aminimide (*2*)-cured epoxy resin composites. (●) Neat resin: Epikote 828 (10 mol) + *2* (1 mol), 150° C (4 h) + 110° C (5 min). Mica flakes: (○, △, □) as in Fig. 2.

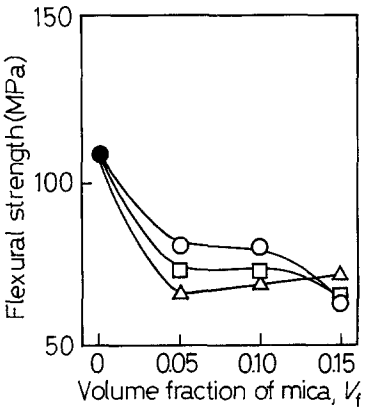


Figure 4 Flexural strength dependence on volume fraction of mica flake for epoxy resin composites cured with *3*. (●) Neat resin: Epikote 828 (10 mol) + *3* (1 mol), 150° C (80 min) + 110° C (5 min). Mica flakes: (○, △, □) as in Fig. 2.

and flexural modulus were determined from stress–strain curves. In all cases, 6 to 7 specimens per sample were tested and the measured values were averaged.

2.2.3. Microscopic observation

In order to examine the distribution and orientation of mica flakes in the moulding, microscopic photographs were obtained for cross-sectioned specimens. The cross-sectioned surface was ground and polished and then examined in reflected light in an optical microscope (OM) (Vanox, Olympus). The fracture surfaces of the specimens, as obtained from the flexural measurements, were examined in a scanning electron microscope (SEM) (Alpha-10, Akashi). Prior to examination the surface was coated with a thin sputtered layer of gold in order to avoid charging effects.

3. Results and discussion

3.1. Flexural strength and flexural modulus

The results of flexural measurements are shown in Figs 2 to 9. Figs 2, 3, 6 and 7 indicate the flexural strength and flexural modulus dependence on the volume fraction of mica flake for aminimide-cured epoxy resin composites. In Figs 4, 5, 8 and 9 are shown those for the reference epoxy resin composites.

It is noteworthy that the intact mica flakes without surface treatment exhibit a substantial reinforcing effect on the flexural strength in the case of aminimide-cured epoxy resin composites (Figs 2 and 3). A further surprise is the difference among the curing agents (*1* to *4*) used. Reference epoxy resins cured with either *3* or *4* behave just like conventional matrix resins. The reduction in flexural strength amounts to 30 to 40% when a small fraction of mica is added (Figs 4 and 5). The results indicate a rather poor adhesion of epoxy resin to the mica surface. The curing conditions (150° C, 80 min for *3* and 80° C, 2 h + 150° C, 2 h for *4*) are recommended for these curing agents. Also the flexural strength of the neat resins without filler falls in the range reported in the literature [6].

It is a general consequence that inorganic fillers enhance the modulus of a resin. In addition to this general consequence, the aminimide composites (Figs 6 and 7) and the reference epoxy composites (Figs 8 and 9) show considerable differences, particularly in flexural strength. This phenomenal difference will be attributed to the superior adhesiveness and toughness of the aminimide-cured resins.

The flexural strength and modulus of neat resins is not much dependent on the kind of curing agent. The specific behaviours of aminimide-cured systems are attributed to the mechanism of curing [3, 5, 7]. At elevated temperatures, aminimides decompose to yield tertiary amines and isocyanates, both of which participate in curing reactions. As we discussed in previous articles [3, 5, 7], comparatively slow curing with tertiary amines (polymerization catalysts) at high temperatures above T_g would provide a stress-free matrix.

TABLE I Characteristics of mica flakes (phlogopite)

Property	Mica		
	S-60	S-200	S-325
Particle size distribution (%)			
+840 μm	Trace	–	–
+420 μm	3	Trace	–
+210 μm	40	1	–
+105 μm	81	15	–
+ 63 μm	91	45	Trace
+ 44 μm	96	65	3
− 44 μm	4	35	97
Average particle size, D (μm)	190	60	16
Average thickness (μm)	3.3	1.5	0.7
Average aspect ratio, α	80	55	35

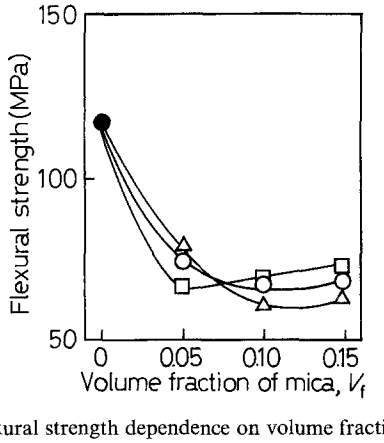


Figure 5 Flexural strength dependence on volume fraction of mica flake for epoxy resin composites cured with *4*. (●) Neat resin: Epikote 828 (10 mol) + *4* (5.8 mol), 80° C (2 h) + 150° C (2 h) + 110° C (5 min). Mica flakes: (○, △, □) as in Fig. 2.

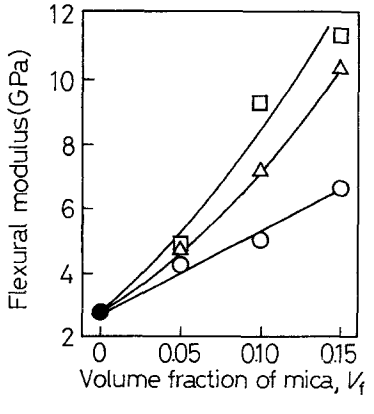


Figure 6 Flexural modulus dependence on volume fraction of mica flake for aminimide (*1*)-cured epoxy resin composites. (●, ○, △, □) as in Fig. 2.

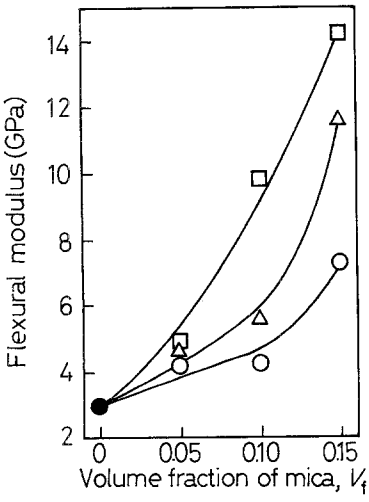


Figure 7 Flexural modulus dependence on volume fraction of mica flake for aminimide (*2*)-cured epoxy resin composites. (●, ○, △, □) as in Fig. 3.

Furthermore, the participation of isocyanate either via urethane or via oxazolidone formation would also modify the properties. The maximum fracture elongation reaches 15%, which is incredibly high for an epoxy resin [3].

An effect of the characteristics of mica flakes, average particle size (D) or average aspect ratio (α), on the flexural strength could not be clearly seen in Figs 2 to 5. In the case of aminimide-cured epoxy resin composites (Figs 2 and 3), however, it seems that in the region of V_f = 0.05 to 0.10, fine particles with a small aspect ratio (S-325: D = 16 μm, α = 35) have higher flexural strength values than the others. With increasing V_f, the flexural strength decreases and at the highest V_f examined (0.15), the particle with the highest aspect ratio (S-60: D = 190 μm, α = 80) exhibits the strongest reinforcing effect. The decrease in flexural strength in the region of V_f = 0.10 to 0.15 is well suppressed when the filler with the highest aspect ratio (α = 80) is dispersed.

On the other hand, the effect of the characteristics of mica flakes on the flexural modulus can be clearly seen in Figs 6 to 9. With increasing aspect ratio of the flakes, the flexural modulus increases in all the regions of V_f. In particular, the mica flakes with the highest aspect ratio (α = 80) dispersed in the aminimide-cured composites increase the flexural modulus of the neat resins by a factor of 4 to 5 at high contents of the filler (Figs 6 and 7). The reinforcing effect of this filler

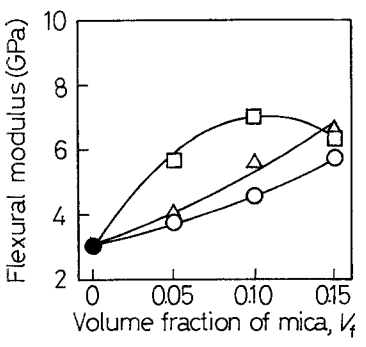


Figure 8 Flexural modulus dependence on volume fraction of mica flake for epoxy resin composites cured with *3*. (●, ○, △, □) as in Fig. 4.

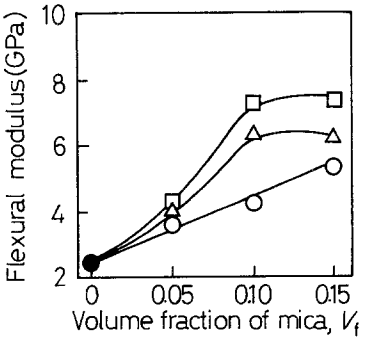


Figure 9 Flexural modulus dependence on volume fraction of mica flake for epoxy resin composites cured with *4*. (●, ○, △, □) as in Fig. 5.

with the highest aspect ratio (α = 80) is not so large in the reference composites (Figs 8 and 9). The flexural modulus levels off at a high content of the filler.

The flexural strength values are quite sensitive to the void contents and other imperfections in the test specimens. In Figs 2 and 3, the flexural strength does not increase in the region of V_f = 0.05 to 0.15, but does not decrease as drastically as in the reference composites. The flexural modulus (Figs 6 and 7), however, increases continuously to the highest volume fraction (V_f = 0.15). These results also clearly indicate the strong adhesiveness between the filler surface and the aminimide-cured epoxy resin matrix. These properties of the matrix resin for the composite materials are ascribed to the features of aminimide-cured epoxy resins, such as hardness and toughness with outstanding elongation at breakage.

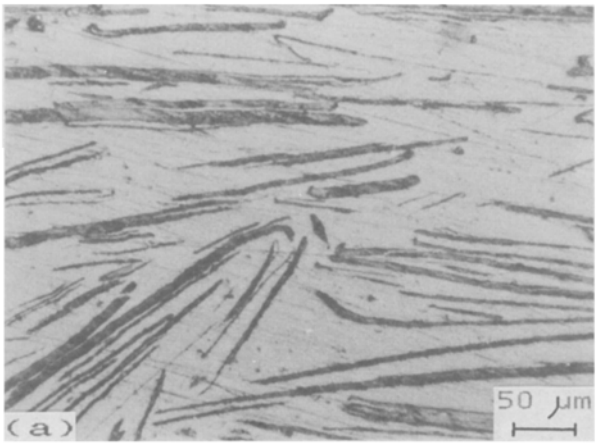
3.2. Microscopic observation of the composites

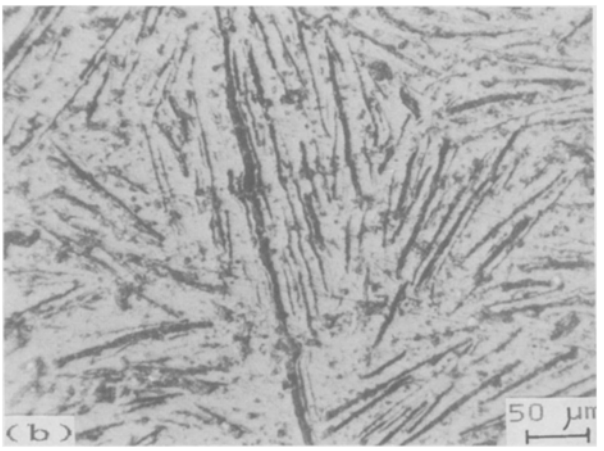
Fig. 10 shows optical micrographs of the polished section of the aminimid (*2*)-cured composite specimens containing mica flakes with different particle sizes. Similar photographs were obtained for other reference epoxy resin composites. There seems to be no significant difference in the distribution of the flakes among the present composite specimens. Overlap and contact of flakes are clearly seen, while the alignment and distribution of flakes in the matrix are random. An orientational effect of the filler giving rise to anisotropic moduli was therefore not considered.

Scanning electron micrographs of the fracture surface of the aminimide (*2*)-cured composite and that of the reference epoxy resin composite cured with *4* are shown in Figs 11a and b, respectively. Many smooth flake surfaces were observed in the fracture surface of the reference composite (Fig. 11b), indicating fracture at the mica–matrix resin boundary. In contrast, residual resin on the mica flakes (thread-like white substance in between flakes) as shown in Fig. 11a is clearly discernible for the aminimide-cured system, indicating the good adhesivity of the resin to the filler. These SEM results are in accordance with the excellent mechanical properties of the present composites.

3.3. Theoretical treatment of the flexural modulus

Since it was difficult to decide from the SEM





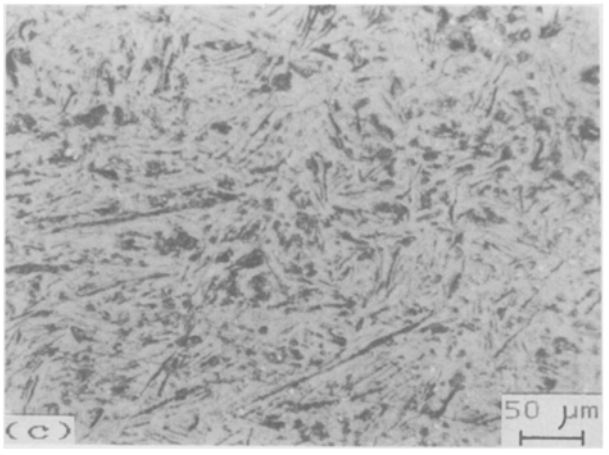


Figure 10 Optical micrographs of polished sections of the composites. Matrix: Epikote 828 + 2. Mica flakes: $V_f = 0.15$. (a) $D = 190\,\mu m$, $\alpha = 80$; (b) $D = 60\,\mu m$, $\alpha = 55$; (c) $D = 16\,\mu m$, $\alpha = 35$.

observation of fracture surfaces after destructive testing whether the mode of failure was flake pull-out or flake fracture, the role of the flake tensile strength in the ultimate strength of the composite could not be ascertained. Mica flakes have a known tendency to shear easily along their cleavage planes, thereby introducing a third mode of failure which has not previously been given theoretical consideration. Padawer and Beecher [2] have summarized the theories of flake and film reinforcement and have reported data for composites containing glass flakes, aluminium boride (AlB_2) platelets, and silicon carbide (SiC) platelets. The results of flexural modulus measurements were compared with the modified Rule of Mixtures relationship

$$E_c = V_f E_f(\mathrm{MRF}) + (1 - V_f)E_m \qquad (1)$$

where E_c, E_f and E_m are the flexural moduli of composite, flake and matrix, respectively, and V_f is the volume fraction of the flakes. The modulus reduction factor (MRF) as modified for flakes by Padawer and Beecher is given by

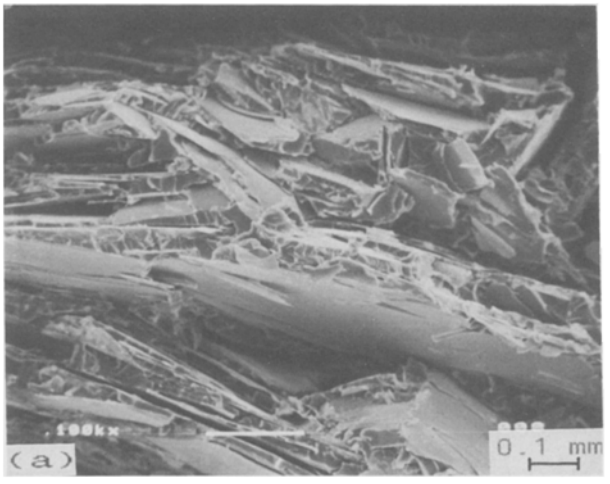
$$\mathrm{MRF} = 1 - \frac{\tanh u}{u} \qquad (2)$$

where

$$u = \alpha\left(\frac{G_m V_f}{E_f(1 - V_f)}\right)^{1/2} \qquad (3)$$

α is the flake aspect ratio and G_m is the matrix shear modulus determined by torsion pendulum measurements. The values used in this treatment for E_f, E_m and G_m are listed in Table II. This derivation is based on a shear-lag analysis representing the case for an isolated flake, neglecting flake–flake interactions. The virtual work analysis of Riley [8] for fibre composites takes fibre interactions into account. This treatment has been modified for a simple lap joint (rectangular flakes) with the result that

$$\mathrm{MRF} = 1 - \frac{\ln(u + 1)}{u} \qquad (4)$$



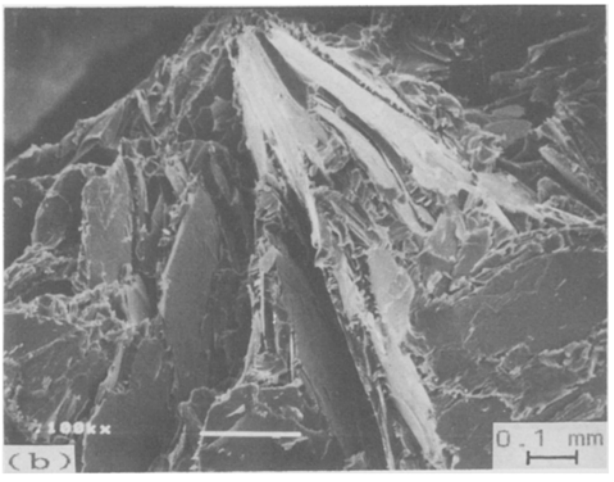


Figure 11 Scanning electron micrographs of the fracture surface. Mica flake: $D = 190\,\mu m$, $\alpha = 80$, $V_f = 0.15$. Matrix (a) Epikote 828 + 2; (b) Epikote 828 + 4.

TABLE II Flexural moduli of flakes (E_f) and matrix (E_m), and matrix shear modulus (G_m)

Modulus	Material	Modulus value (GPa)
E_f*	Mica flake (phlogopite-S)	190
E_m†	Epikote 828 + *1*	2.78
	Epikote 828 + *2*	2.90
	Epikote 828 + *3*	3.02
	Epikote 828 + *4*	2.35
G_m‡	Epikote 828 + *1*	1.276
	Epikote 828 + *2*	1.277
	Epikote 828 + *3*	1.600
	Epikote 828 + *4*	1.093

*From technical data of Canada Mica Co. Ltd.
†Measured in flexural test of neat resin.
‡Measured in torsion pendulum test of neat resin.

TABLE III K values in Equation 5 determined by the curve-fitting treatment.

Material	V_f		
	0.05	0.10	0.15
Epikote 828 + *1*	0.529 (1.00)*	0.488 (0.92)	0.387 (0.73)
Epikote 828 + *2*	0.427 (1.00)	0.400 (0.94)	0.524 (1.23)
Epikote 828 + *3*	0.387 (1.00)	0.240 (0.62)	0.137 (0.35)
Epikote 828 + *4*	0.442 (1.00)	0.400 (0.90)	0.210 (0.48)

*( ) Normalized values.

where the MRF includes the effect of flake–flake interactions.

Overlapping of mica flakes was observed in OM and SEM observation; therefore, Riley's equation (Equation 4) which includes flake–flake interactions is chosen to compare the experimental results with the theoretical values. The comparison of experimentally determined values of the MRF (Equation 1) for various aspect ratios with their theoretical curves (Equations 4 and 3) gave generally poor agreement. The experimental values were below the theoretical relationships derived by Riley and by Padawer and Beecher (Equations 4 and 3). Since both of these theories assume rectangular-shaped flakes with uniform thickness and parallel arrangement, they do not accurately represent the case for mica flakes which can be highly irregular in both diameter and thickness. Moreover, the alignment, overlap and distribution of flakes is irregular. Resin-rich regions, voids and flake imperfections contribute to the lowering of strength and modulus values so that the predictions of Riley broadly represent the upper limit for an ideal case.

Equation 3 was modified by the efficiency factor ($K \leqq 1$) [2] on the basis of the microscopic observations. u values can be described by the equation

$$u = K\alpha\left(\frac{G_m V_f}{E_f(1 - V_f)}\right)^{1/2} \quad (5)$$

K includes the origins of weakening due to flake misalignment and non-uniformities of flake shape, size and distribution in the matrix, and also depends on the degree of adhesion between filler and matrix. Equation 5 reduces to Equation 3 for an ideal system ($K = 1$).

Applications of Equations 4 and 5 to the present results are shown in Figs 12 to 15. K values which give the best-fit curves with their normalized values are listed in Table III.

It can be seen that the MRF values of the aminimide-cured composites (Figs 12 and 13) are larger than those of the reference epoxy resin composites (Figs 14 and 15). In Fig. 13, especially, the aminimide (*2*)-cured composite at a high content of mica flake ($V_f = 0.15$) exhibited high MRF values without any surface treatment for the flakes. This reinforcing effect is almost comparable to that of surface-treated mica flake dispersed in polyester resin matrix, in which the mica flake is encapsulated with a copolymer of styrene, acrylic acid and 2-sulphoethyl methacrylate [1].

K values of the aminimide (*1* and *2*)-cured composites do not change so remarkably with the volume fraction (Table III). In contrast, K values of the reference epoxy resin composites cured with *3* and *4* decrease to a greater extent with increasing volume fraction. This may be attributable to a difference in the degree of dispersion of mica flakes in the matrix. If the

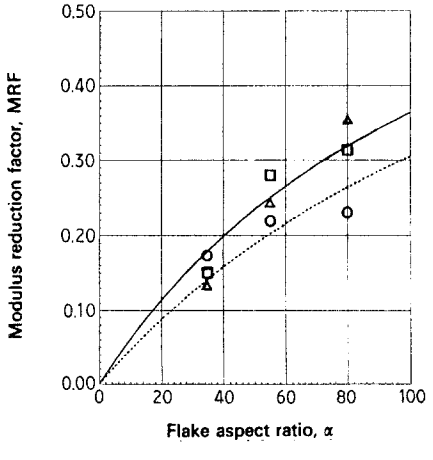


Figure 12 Curve-fitting treatment by Equations 4 and 5 with experimental plots. Matrix: Epikote 828 + *1*. Experimental values: V_f = (○) 0.05, (△) 0.10, (□) 0.15. Fitted curves: V_f = (····) 0.05, (– – –) 0.10, (——) 0.15.

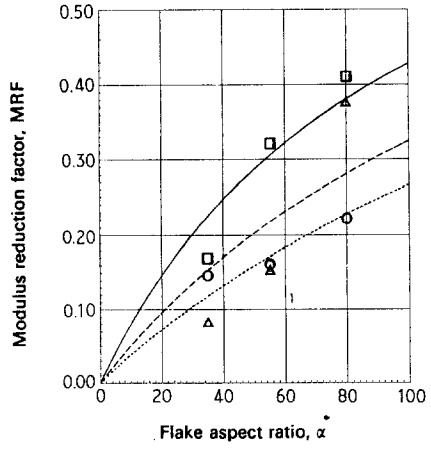


Figure 13 Curve-fitting treatment by Equations 4 and 5 with experimental plots. Matrix: Epikote 828 + *2*. V_f = (○) 0.05, (△) 0.10, (□) 0.15. Fitted curves: V_f = (····) 0.05, (– – –) 0.10, (——) 0.15.

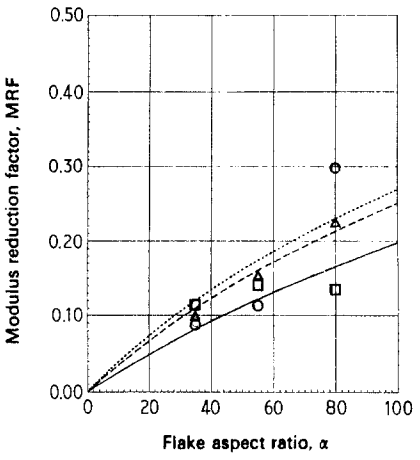


Figure 14 Curve-fitting treatment by Equations 4 and 5 with experimental plots. Matrix: Epikote 828 + *3*. Symbols and curves as in Fig. 13.

aminimides, particularly for *2*, have a good hydrophilic–lipophilic balance (effective in dispersing the mica flakes into the matrix), it is reasonable that the dispersivity of mica flakes and therefore the K values of the aminimide composites do not change so remarkably with the volume fractions of mica flakes.

The present results, therefore, are understandable from our previous study on the mechanical properties of the aminimide-cured epoxy resins. We have found that (i) these epoxy resins show good adhesion properties, and (ii) they are tough and strong, showing an elongation as much as 15% at breakage.

Another factor providing favourable properties for *1* and *2* is the strongly hydrophilic nature of aminimides. These compounds are freely soluble in water and highly hygroscopic. Consequently, the affinity of epoxy resins cured with aminimides to mica flake is expected to be much better than that of the other epoxy resins. Indeed, aminimide compounds act as surfactants [9, 10], and epoxy prepolymers with aminimides are effective surface-treating agents for mica when mica flakes are blended with polyolefins [11, 12]. A substantial reinforcing effect was observed both on strength and on modulus for polyolefin–mica flake composites [11, 12]. Another example is the pretreatment of mica flakes for chemical plating [13]. Mica flakes thus treated show a good receptivity to chemical plating by various metals for the purpose of preparing electromagnetic shielding materials. All pieces of evidence point towards the uniqueness of epoxy prepolymers with aminimides having a good affinity towards the mica surface. Aminimides are thus not only effective as curing agents but also play the role of substituting silane couplers.

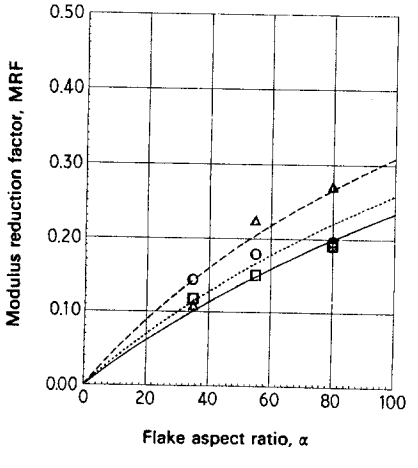


Figure 15 Curve-fitting treatment by Equations 4 and 5 with experimental plots. Matrix: Epikote 828 + *4*. Symbols and curves as in Fig. 13.

The present results do not seem to represent the best performance of the new resin. Since the specific gravities of filler and resin are considerably different, precipitation of mica during the curing process was observed. Although we tried to obtain a homogeneous distribution of filler particles by turning over the sample several times in the course of curing, it was not sufficient to ensure homogeneous dispersion. Recently we constructed a special curing system in which the sample compartment rotates and tumbles in an oven so that a pseudo-zero gravity state is achieved [14]. Using this system, we are preparing composite materials in which filler particles are randomly oriented and homogeneously dispersed, so as to achieve the maximum performance of the present resins.

Acknowledgements

This research was carried out in the course of a joint research scheme between T.I.T and NTN Toyo Bearing Co. Ltd. We would like to express our sincere gratitude to Dr H. Muro, Mr A. Yanagisawa and Mr S. Oshima of NTN Toyo Bearing Co. Ltd. for continuous support and encouragement.

References

1. J. LUSIS, R. T. WOODHAMS and M. XANTHOS, *Polym. Eng. Sci.* **13** (2) (1973) 139.
2. G. E. PADAWER and N. BEECHER, *ibid.* **10** (3) (1970) 185.
3. S. INUBUSHI, T. IKEDA, S. TAZUKE, T. SATOH and Y. KUMAGAI, *J. Polym. Sci., Polym. Chem. Edn* (1988) in press.
4. M. YOSHIKAWA, T. SATOH, S. INUBUSHI, T. IKEDA and S. TAZUKE, *J. Mater. Sci. Lett.* **5** (1986) 1239.
5. S. INUBUSHI, T. IKEDA, S. TAZUKE and T. SATOH, *J. Polym. Sci., Polym. Chem. Edn* **25** (1987) 137.
6. H. KAKIUCHI, "New Epoxy Resins" (Shokodo, Tokyo, 1985) Ch. 5, p. 164.
7. S. INUBUSHI, T. IKEDA, S. TAZUKE and T. SATOH, *J. Polym. Sci., Polym. Chem. Edn* **25** (1987) 1363.
8. V. R. RILEY, Interaction Effects in Fibre Composites, Polymer Conference Series, University of Utah, June 1970.
9. W. J. McKILLIP, E. A. SEDOR, B. M. CULBERTSON and S. WAWZONEK, *Chem. Rev.* **73** (3) (1973) 255.
10. S. WAWZONEK, *Ind. Eng. Chem. Prod. Res. Dev.* **19** (1980) 338.
11. S. INUBUSHI, T. IKEDA, S. TAZUKE, Y. KUMAGAI, M. YOSHIKAWA and T. SATOH, in Preprints of the 2nd Japan–US Polymer Symposium, Kyoto, October 1985 (Society of Polymer Science, Kyoto, Japan, 1985) p. 165.
12. S. INUBUSHI, T. IKEDA, S. TAZUKE, T. SATOH and Y. KUMAGAI, *J. Mater. Sci.* **23** (1988) 535.
13. I. TODOROKI, T. KANBE and Y. KUMAGAI, submitted for Japanese Patent.
14. Y. KUMAGAI and S. TAZUKE, to be published.

Received 14 November 1986
and accepted 18 June 1987