Effect of Coupling Agents on the Mechanical Properties of Mica/Epoxy and Glass Fiber/Mica/Epoxy Composites

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SYNOPSIS

The effect of coupling agents, two silane and one zirconate, on the mechanical properties of mica/epoxy and glass fiber/mica/epoxy composites has been investigated. The results showed that tensile modulus and flexural strength and modulus values were improved by the surface treatment of the coupling agents. The property retention was also found to be better in the case of coupling agent-treated mica/epoxy samples after boiling in water for 2 h. In the case of glass fiber/mica/epoxy composites, the flexural modulus and interlaminar shear strength values improved with increase in mica content, but the effect of coupling agents was not pronounced.

INTRODUCTION

The increasing use of mica as an alternative reinforcing filler for thermoplastics and thermosets has focused attention on the factors that influence the physical and mechanical properties of mica-filled composites.[1,2] Compared with other platelet-type materials, mica offers the advantage of a relatively high modulus of 172 GN/m^2 against 73 GN/m^2 for glass flakes. Hence, the choice of mica as a candidate for two-dimensionally reinforced composites is obvious.

Mica also has excellent chemical and corrosion resistance, good electrical properties, and low coefficient of thermal expansion and causes low abrasion and wear to the processing equipment.

Maine and Shepherd[3] suggested that the most promising area of full utilization of the planar-reinforcing properties of mica is in sheet materials, although other fabrication techniques can be used. Processing, application, and properties of mica and its composites have been reviewed in Ref. 4. The friable nature of the mica flakes frequently leads to partial breakage and delamination of the filler particles during processing, such that the initial dimensions may be altered significantly, thereby influencing the physical and mechanical properties of the composites.

There is extensive literature pertaining to the mechanical behavior of filled polymers.[5-9] In all these studies, it has been demonstrated that the modulus is the easiest mechanical property to estimate, since it is a bulk property that depends primarily on the geometry, modulus, particle-size distribution, and concentration of the filler. The tensile strength of a filled polymer is, however, more difficult to predict because it depends strongly on local polymer–filler interactions in addition to the above.

A number of investigators have shown that two general tensile strength–filler concentration responses are possible on the basis of adhesion between the two materials.[10-13] Strong adhesion or the interfacial bond strength depends on the effectiveness of the coupling agents and the inherent wetting ability of the polymer. The main assumptions for good wetting are related to similar polarities of both phases, as well as to low surface tension and low viscosity of the matrix polymer. Among the polymers, thermosetting resins are generally low viscous liquids and are converted to solids by cross-linking and, also, most of them are polar in nature.

Coupling agents play an important role in enhancing the filler–resin interaction, thus they help in improving the mechanical properties due to better

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stress transfer. Weatherability is also improved due to the more stable interface between the matrix and the reinforcing material. An additional advantage is improved processability even at higher filler loading in a composite. Two classes of coupling agents that are used mainly in composites are silanes and titanates. Application, evaluation, and mechanism of these agents have been reviewed in Ref. 14. It has been observed that mica responds to coupling agents significantly, in terms of both mechanical properties and weatherability.[4,15] In the present study, two silanes, namely, 3-aminopropyltriethoxysilane and 3-glycidoxy propyltrimethoxysilane, and a zirconate, neoalkoxy tri(dioctylpyrophosphato)zirconate, have been used for epoxy/mica composites. Influence of these coupling agents on the mechanical behavior of glass fiber/mica/epoxy laminates has also been studied.

EXPERIMENTAL

Materials

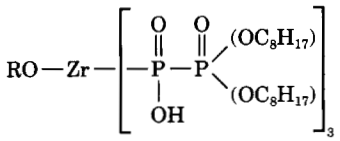
The epoxy resin used was diglycidyl ether of bisphenol-A supplied by Hindustan Ciba Geigy Ltd., India, commercially known as Araldite LY 556, of epoxy equivalent value 189. The cross-linking agent used was 4,4′-diaminodiphenylmethane.

Micronized mica was supplied by Export Linkers, Bihar, India. It is a dry ground product of muscovite type whose particle-size distribution is shown in Figure 1. The loading was from 10, 20, and 30% by weight.

Three coupling agents were used in the present study:

(i) Neoalkoxy tri(dioctyl pyrophosphato)-zirconate, LZ38, a product of Kenrich Petrochemicals, USA:

LZ38

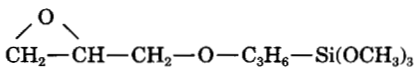


(ii) 3-aminopropyltriethoxysilane:

SiNH $NH_2—C_3H_6—Si(OC_2H_5)_3$

(iii) 3-glycidyloxy propyltrimethoxysilane

SiEP



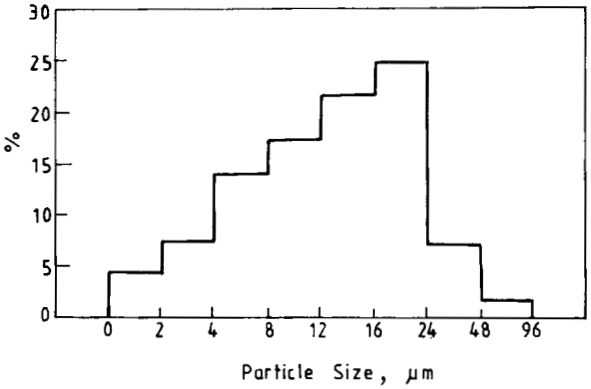


Figure 1 Particle-size distribution of mica.

The glass fabric (300 g/m^2) was of the B type supplied by Pilkington Fibre Glass Ltd., India, with a surface treatment compatible with epoxy resin.

Coating of Mica with Coupling Agent

One percent (on the weight of mica) coupling agents solution was prepared in toluene. Mica powder was first washed with demineralized water and dried at 100°C for 2 h. Mica, 200 gm, was prewetted using about 150 mL of toluene, and the coupling agents solution was added to this with vigorous stirring. The slurry was agitated for 4 h and dried at 100°C for 2 h.

Contact Angle Studies

Mica in the form of sheet (2 × 2 cm) was subjected to coupling agent treatment. Small drops of resin was kept on its surface and the contact angle formed by the resin on the mica surface was measured under an optical microscope. The thermodynamic work of adhesion was calculated using the following relationship[16]:

$$W_a = S(1 + \cos\theta)$$

where W_a = thermodynamic work of adhesion, S = surface tension of epoxy resin, and θ = contact angle formed at the resin and mica.

Preparation of Test Specimens

Both treated and untreated mica were added to hot epoxy resin and stirred with a mechanical stirrer for about 0.5 h. This mixture was degassed under vacuum at 90°C and then added to the molten hardener (in stoichiometric ratio, i.e., 27 parts by weight per

Table I Mechanical Properties of Untreated and Surface-Treated Mica/Epoxy Composites

Sample	Mica (%)	FL STR[a] (MPa)	FL MOD[a] (GPa)	TEN STR[a] (MPa)	TEN MOD[a] (GPa)	ELONG[a] (%)
Untreated	0	128.9	3.03	76.4	1.73	11.48
		(2.46)	(4.77)	(2.28)	(3.43)	(2.18)
	10	132.2	3.57	52.9	2.06	4.60
		(4.07)	(3.52)	(2.91)	(7.53)	(3.34)
	20	90.3	4.60	52.2	2.38	3.69
		(6.83)	(5.68)	(3.13)	(5.26)	(4.26)
	30	84.9	5.22	48.3	2.67	2.77
		(1.08)	(3.08)	(5.29)	(3.23)	(3.02)
LZ38	10	95.1	3.76	55.8	2.36	4.95
		(4.82)	(6.05)	(4.02)	(2.08)	(6.80)
	20	126.4	4.84	50.4	2.45	3.56
		(1.63)	(4.25)	(1.43)	(5.29)	(6.30)
	30	90.7	5.70	46.3	3.08	2.41
		(2.96)	(6.54)	(4.42)	(3.30)	(3.26)
SiNH	10	143.8	4.05	57.2	2.51	3.84
		(1.85)	(1.05)	(6.61)	(5.48)	(6.75)
	20	131.0	4.58	51.7	2.79	3.53
		(2.24)	(3.26)	(3.33)	(7.37)	(5.88)
	30	95.3	5.61	50.0	3.06	2.62
		(3.36)	(3.38)	(3.22)	(4.93)	(3.84)
SiEP	10	137.3	3.45	53.5	2.45	4.55
		(1.07)	(2.47)	(3.44)	(3.31)	(4.11)
	20	114.4	5.40	53.7	2.70	3.45
		(3.31)	(2.71)	(3.23)	(5.32)	(2.06)
	30	102.2	5.78	48.4	3.51	2.58
		(2.53)	(3.04)	(3.87)	(4.18)	(5.72)

Standard deviation (SD) values are given in parentheses in terms of percentage covariance (%CV). %CV = SD × 100/mean.
[a] FL STR = flexural strength; FL MOD = flexural modulus; TEN STR = tensile strength; TEN MOD = tensile modulus; ELONG = elongation.

100 parts of the resin), mixed thoroughly, degassed till the surface became clear, and cast in metal molds with a Teflon spacer of 3 mm thickness. Silicone spray was used as a mold-releasing agent. The curing cycle adopted was 110°C for 2 h, followed by a post-curing of 5 h at 180°C under nitrogen. The sheets thus obtained were cut into strips by a high-speed diamond-edged wheel cutter and the edges of the samples were polished using fine-grade sandpaper to remove the possible surface cracks.

Aging of the Samples

The specimens were subjected to boiling water treatment for 2 h.

Preparation of Glass/Mica/Epoxy Laminates

The epoxy resin/hardener/mica mix was applied uniformly on the glass cloth (20 × 15 cm^2). Sixteen plies were used to obtain a thickness of about 2 mm. The sheets were cured in a hot press at 110°C for 2 h with a pressure of 10 kg/cm^2. The sheets were then postcured at 180°C for 5 h under nitrogen atmosphere. The test specimens were cut as in the previous case. The glass content was estimated by burning off the matrix at about 600°C in a muffle furnace and was found to be 70 ± 1% by weight. The void volume fraction calculated by density measurements was 0.03.

Evaluation of Composites

The tensile and flexural studies were carried out on an Instron machine (Model 1112) as per ASTM D-638 and ASTM D-790, respectively. Fracture surface morphology was studied for the tensile fractured specimens with the help of a Cambridge Stereoscan S4-10 scanning electron microscope.

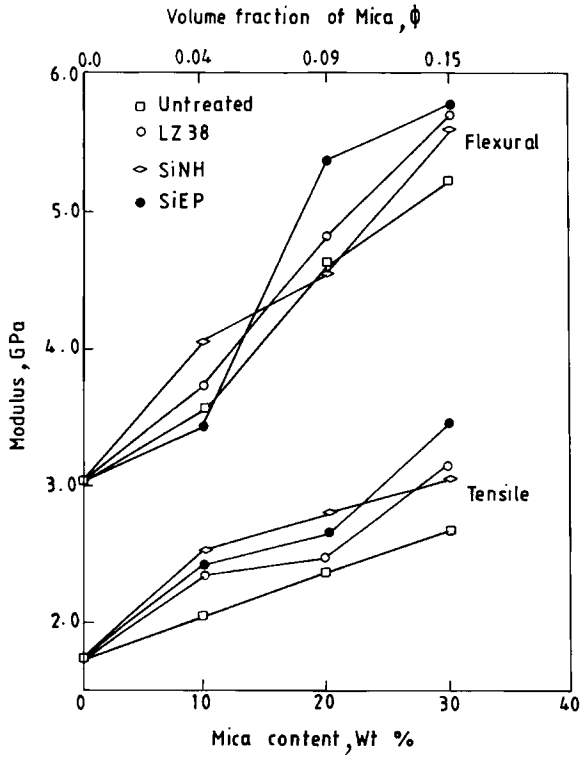


Figure 2 Flexural and tensile modulus vs. mica loading for untreated and surface-treated mica/epoxy composites.

RESULTS AND DISCUSSION

Flexural and Tensile Moduli

The flexural and tensile moduli values for all the epoxy/mica systems are shown in Table I and Figure 2.

On increasing the mica loading, moduli of the composite increase. Enhancement in flexural modulus of the 30% mica-loaded epoxy sample is about 70% with respect to the neat resin sample (3.03–5.22 GPa), while the increase in tensile modulus for the same sample is about 54% (1.73–2.63 GPa).

Modulus is also dependent on the aspect ratio of the filler.[17] Bramuzzo et al.[18] reported that for equal volume fractions of the filler in polypropylene (PP) composites talc (flaky in nature) showed higher modulus values compared to glass beads and calcium carbonate. A similar observation has been made by Jancar[19] for PP/$Mg(OH)_2$ and PP/$CaCO_3$ systems. The difference is attributed to higher specific surface area and anisotropic particle shape. Other factors that influence the strength and modulus properties are orientation, interfacial interaction, and the nature of failure. The performance of mica used in the present case lies in between that of a spherical filler and fiber or high-aspect ratio flakes.

With surface treatment, the modulus values are further increased for mica/epoxy composites. However, glycidoxy silane-coated 30% mica/epoxy composite showed the highest value of both flexural (5.78 GPa) and the tensile modulus (3.51 GPa). Effective surface treatment of mica with glycidoxy silane perhaps changes the free surface energy of mica by interacting physically and/or chemically (through the glycidoxy ring with the OH groups), thereby improving the wettability. Surface treatment also helps in deagglomeration of the fine particles.

In case of the zirconate-coated mica/epoxy samples, the interaction may be purely physical or through hydrogen bonding. The results of contact angle study further substantiate the improved wettability of mica with resin by surface treatment with silanes and zirconate, as shown below:

	Untreated	LZ38	SiNH	SiEP
Contact angle (deg)	26.1	9.9	9.5	12.7
Work of adhesion (J/m^2)	0.911	0.953	0.953	0.948

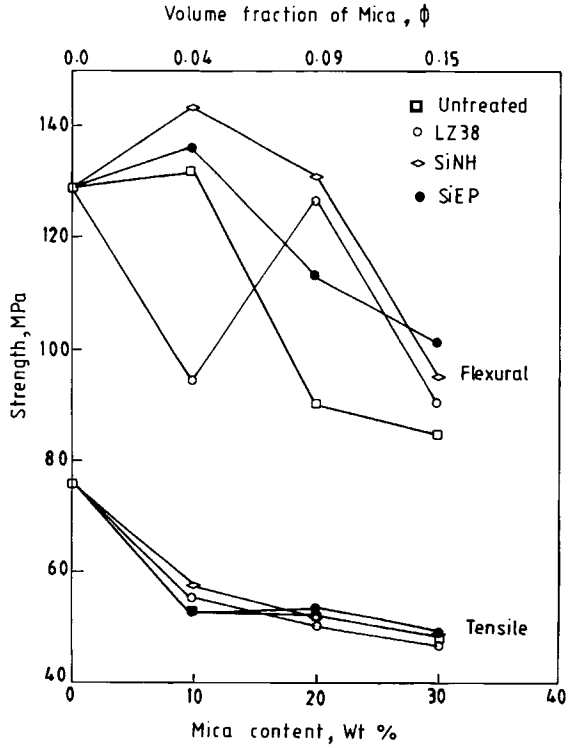


Figure 3 Flexural and tensile strength vs. mica loading for untreated and surface-treated mica/epoxy composites.

Flexural and Tensile Strength of Epoxy/Mica Composites

There is a slight increase in the flexural strength at 10% mica (untreated) loading with respect to neat resin (Fig. 3). But further loading of mica, i.e., 20–30%, decreases the flexural strength by 30%. Tensile strength also drops by about 30–35% with respect to the neat cross-linked epoxy sample.

Improvement in flexural strength at low mica loading was shown by Vu-Kanh and Decharentenay,[13] for PP/mica systems by Xavier and Sharma,[15] for the epoxy mica system by Inubushi et al.[20]

With surface-treated mica/epoxy samples, significant improvement in flexural strength has been observed, particularly for the 10% mica/epoxy sample, in which mica was coated with aminosilane or glycidoxy silane. However, tensile strength values of the surface-treated mica/epoxy sample are nearly the same as that of the uncoated mica/epoxy sample. The difference may be due to the mode of stress experienced by the sample. In the case of flexural testing, it is a combination of compression and tensile, whereas in tensile testing, it is purely tensile. From the mechanical properties, it may be inferred that interfacial adhesion is more effective in compression loading than is tensile loading.

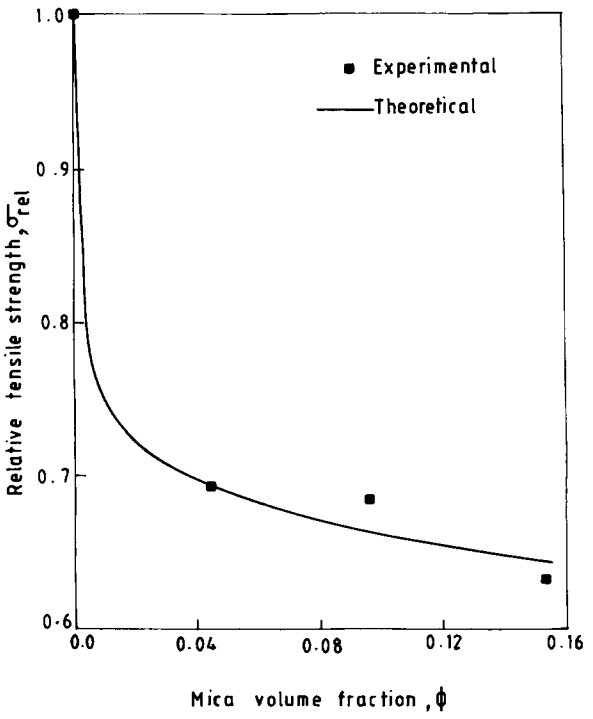


Figure 4 Theoretical prediction of tensile strength using the equation $\sigma_{rel} = (1 - a\varnothing^b)$.

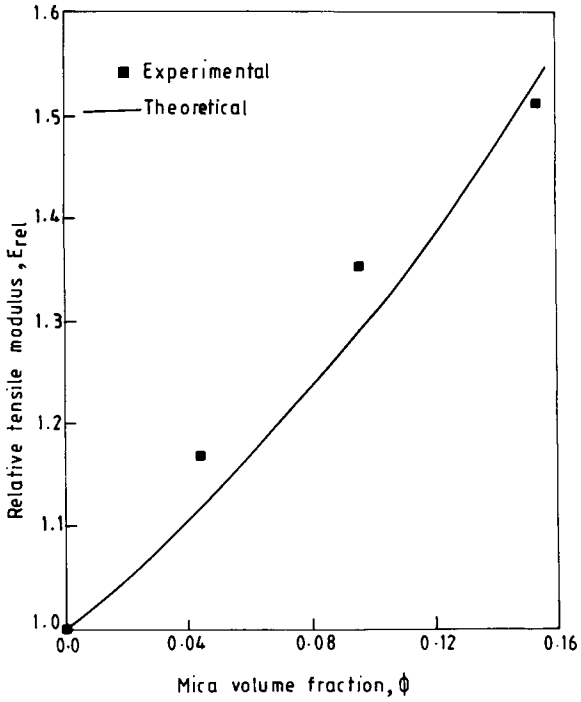


Figure 5 Theoretical prediction of tensile modulus using the equation $E_{rel} = (1 - 0.5K\varnothing)^{-2}$.

Strength reduction in the mica/epoxy sample in relation to the neat resin sample could be attributed to the following factors:

(i) Formation of microcracks in the resin matrix due to the internal stresses developed during curing and the difference in the thermal shrinkage of epoxy resin (8.5×10^{-5} m/m/°C) and mica ($10–20 \times 10^{-6}$).

(ii) Presence of defects like air bubbles due to the addition of filler.

(iii) Ease of delamination of mica particles (spitting energy is 3 J/m^2).

(iv) Mica particles acting as stress concentrators and providing sites for crack growth.

(v) Debonding occurring under tensile stresses due to the poor interfacial adhesion (as shown in the SEM micrograph). The debonded sites can then facilitate the crack growth.

The surface treatment with silanes and zirconate promotes the matrix–mica interaction, thereby improving the flexural strength values in relation to uncoated mica/epoxy samples. At intermediate loading, the improvement in flexural strength in mica/epoxy samples is significant.

Table II Mechanical Properties of Untreated and Surface-Treated Mica/Epoxy Composites after Boiling in Water for 2 h

Sample	Mica (%)	FL STR[a] (MPa)	FL MOD[a] (GPa)	TEN STR[a] (MPa)	TEN MOD[a] (GPa)	ELONG[a] (%)
Neat	0	111.7	2.39	71.3	1.28	12.88
		(7.66)	(3.75)	(2.83)	(5.22)	(8.50)
Untreated	10	122.5	2.77	48.3	1.97	5.51
		(4.82)	(6.81)	(3.02)	(5.28)	(4.89)
	20	81.4	3.15	43.7	2.12	3.74
		(1.50)	(2.56)	(4.42)	(4.59)	(9.92)
	30	76.0	4.12	40.5	2.57	2.97
		(2.39)	(4.40)	(2.02)	(6.06)	(5.45)
LZ38	10	91.1	2.96	54.1	2.26	4.48
		(2.96)	(2.04)	(3.82)	(6.46)	(5.15)
	20	110.4	3.67	45.4	2.41	3.85
		(2.71)	(4.02)	(2.98)	(2.65)	(7.21)
	30	84.7	4.47	41.5	3.00	2.81
		(6.08)	(3.47)	(2.80)	(5.54)	(6.42)
SiNH	10	131.6	3.68	54.6	2.42	4.33
		(3.80)	(1.73)	(4.12)	(6.34)	(6.05)
	20	124.6	4.33	47.3	2.67	3.86
		(2.82)	(3.12)	(3.63)	(2.51)	(5.23)
	30	86.3	4.48	44.3	3.05	2.50
		(4.02)	(3.66)	(3.69)	(6.87)	(8.20)
SiEP	10	121.5	2.85	51.7	2.31	4.85
		(1.22)	(2.53)	(3.68)	(5.03)	(6.38)
	20	104.9	3.87	47.1	2.65	3.70
		(3.28)	(3.52)	(4.57)	(3.57)	(3.88)
	30	95.3	4.18	42.0	3.22	2.90
		(4.08)	(1.21)	(1.87)	(5.72)	(9.55)

[a] See footnote a to Table I.

Elongation at Break

The breaking elongation values decreased considerably (Table I) with an increase in the mica loading. Ten percent loading of mica caused a reduction in elongation-at-break by 60%, but addition of mica up to 30% caused a further reduction of only 15%. Such a significant drop could be due to the ease of delamination of mica that can act as a crack initiation point. In the present situation, the mica particles are randomly oriented and, hence, a large number of particles are subjected to a tensile stress acting perpendicular to the plane.

Theoretical Analysis

Incorporation of a rigid particulate filler in a low modulus matrix results in an increase in modulus and decreases strength values. Prediction of such properties of a filled system is often made complex by factors like filler geometry, size distribution, packing efficiency, orientation, and interfacial interaction.

Padawer and Beecher[21] developed a modified rule of mixtures equation that attempts to predict the composite modulus for short ribbons in a uniform array that takes the flake aspect ratio and the shear modulus of the interface into account. For particulate fillers, various simple approaches have been suggested and are summarized in some recent publications.[11,22] In the present study, an equation developed by Nicolais and Nicodemo[23] has been used to predict the lower-bound tensile strength:

$$\sigma_{rel} = \frac{\sigma_c}{\sigma_m} = (1 - a\varnothing^b) \qquad (1)$$

where σ_{rel} = relative tensile strength; σ_c, σ_m = tensile strength of the composite and matrix, respectively; $\varnothing$ = volume fraction of the filler; and a, b = constants related to the stress concentration and the geometry of the filler, respectively.

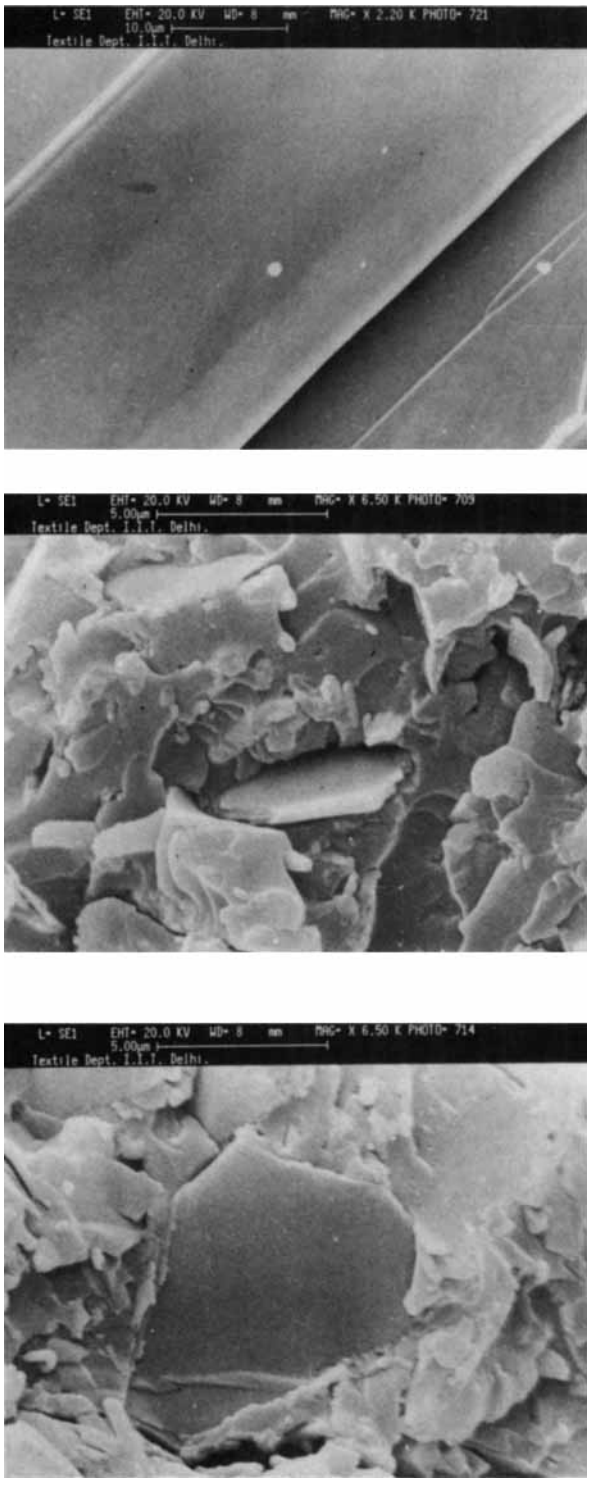


Figure 6 SEM micrographs of tensile-fractured mica/epoxy samples: (a) Fracture surface of unfilled resin showing brittle fracture. (b) Untreated mica/epoxy fracture surface showing a case of debonding followed by failure at the interface. (c) 30% SiEP-treated mica/epoxy showing delamination during crack propagation.

The following equation by Quemada[26] was used for predicting the tensile modulus of the mica/epoxy system:

$$E_{\text{rel}} = \frac{E_c}{E_m} = (1 - 0.5\, K\varnothing)^{-2} \qquad (2)$$

where E_{rel} = relative tensile strength; E_c, E_m = tensile strength of the composite and matrix, respectively; and K = a constant related to the packing efficiency of the filler.

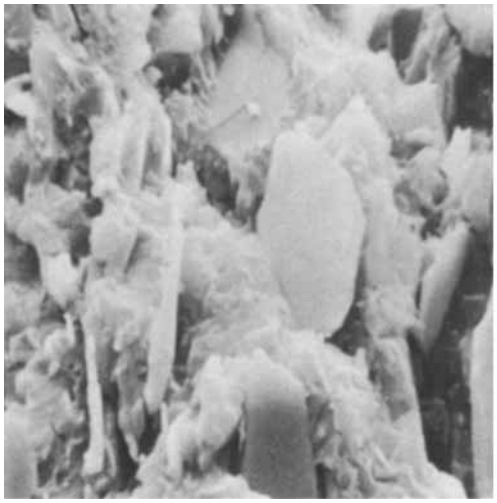
Both the experimental points and theoretically predicted curves for strength and modulus values are shown in Figures 4 and 5, respectively.

Effect of Aging

The strength and modulus values of the samples tested after 2 h boiling water treatment are given in Table II. It has been proposed that water hydrolyzes the interfacial bond and acts as a plasticizing agent.[22] The extent of hydrolysis depends on the thickness of the sample, presence of voids and other surface defects, and length of exposure time and also on the stability of the bonding that, consequently, influences the physical and mechanical properties. It can be seen that the unfilled epoxy and the untreated mica/epoxy samples showed lower retention values compared to that of samples containing surface-treated mica. Among all the cases, the aminosilane-treated mica/epoxy samples showed better retention values. The improved property retention may be due to the hydrophobicity imparted by the coupling agents to the resin–filler interface.

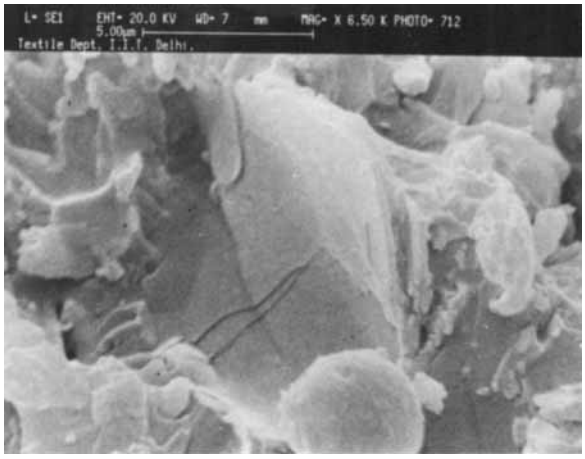
Fracture Morphology

The scanning electron micrographs of the mica powder and the tensile-fractured surfaces of the unfilled and mica-filled epoxy composites are given in Figures 6 and 7. The possible origins of crack initiation in a composite material are air bubbles or voids, resin-rich areas, foreign matter such as dust particles, particle size, and poor particle matrix adhesion.[25] The fractured surface of the unfilled resin [Fig. 6(a)] shows a brittle failure. Debonding at the interface and subsequent flake pullout has been shown in Figure 6(b) for an untreated mica/epoxy sample that may be due to the lack of proper interfacial adhesion. Another possible mode of failure is delamination of the mica particle as shown in Figure 6(c). Since the mica particles are randomly oriented, a large number of them are subjected to

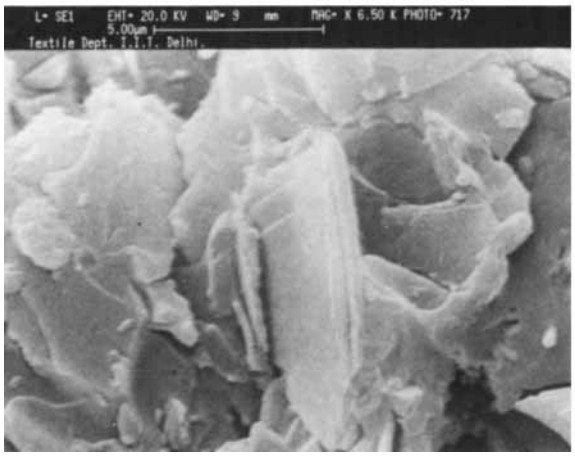


(a) Untr
(x 3000)

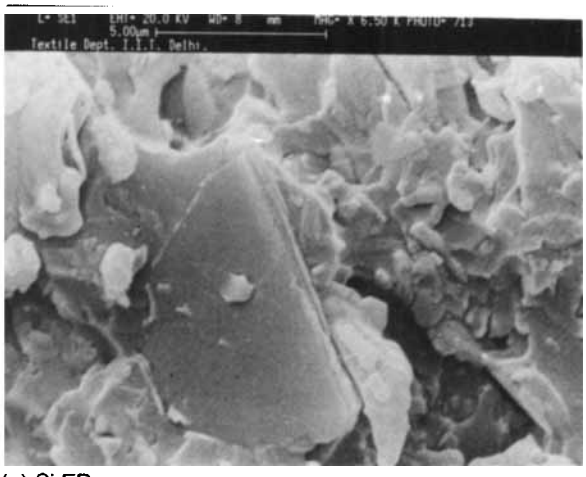
(b) Untr
(x 6000)



(c) LZ 38



(d) Si NH



(e) Si EP

Figure 7 SEM micrographs of tensile-fractured untreated and surface-treated mica/epoxy samples: (a) and (b) Untreated mica/epoxy sample showing debonding and flake pullout. (c) (d), and (e) Surface-treated mica/epoxy samples showing the failure occurring mainly in the matrix and absence of flake pullout.

Table III Mechanical Properties of Glass Fabric/Mica/Epoxy Laminates

Sample	Mica (%) in Resin	FL STR[a] (MPa)	FL MOD[a] (GPa)	ILSS[a] (MPa)
Neat	0	257.1	5.46	18.6
		(2.82)	(4.68)	(2.56)
Untreated	10	255.8	6.63	21.4
		(2.76)	(6.06)	(6.62)
	20	225.4	6.80	20.5
		(2.76)	(4.19)	(4.79)
	30	251.8	7.35	21.9
		(3.73)	(5.31)	(2.63)
LZ38	10	240.1	6.41	23.4
		(2.12)	(2.68)	(7.58)
	20	260.6	7.66	20.7
		(2.80)	(5.42)	(3.33)
	30	224.4	6.57	22.2
		(4.93)	(6.48)	(6.81)
SiNH	10	221.4	5.32	20.4
		(4.50)	(3.57)	(4.65)
	20	213.6	6.12	18.6
		(1.52)	(4.54)	(5.68)
	30	257.2	7.14	23.7
		(2.96)	(6.40)	(6.61)
SiEP	10	209.7	6.46	19.6
		(3.15)	(2.82)	(5.60)
	20	239.6	6.59	23.8
		(3.08)	(7.87)	(7.79)
	30	263.1	7.05	24.2
		(2.53)	(6.85)	(4.94)

[a] FL STR = flexural strength; FL MOD = flexural modulus; ILSS = interlaminar shear strength.

tensile stresses acting perpendicular to the plane and the crack propagation occurs parallel to the plane. Mica undergoes delamination readily since it has a low splitting energy.

Figure 7(a)–(e) shows the fracture surfaces of the untreated and surface-treated mica/epoxy samples. It is clearly seen that the untreated mica/epoxy sample [Fig. 7(a) and (b)] has experienced a high level of debonding and flake pullout. This accounts for the lower strength and modulus values and higher elongation at break. In the case of surface-treated mica/epoxy samples [7(c)–(e)], the failure seems to have occurred mainly in the matrix, which can be explained by the improved interfacial adhesion resulting in better mechanical properties. The absence of flake pullout justifies the reduction in elongation at break in these samples. Fracture of mica particles across the plane was not observed, which may be due to its high tensile strength (about 650 MPa) compared to that of the resin (about 80 MPa). The aspect ratio of mica presently used is below the critical value (< 30) above which the failure might also occur in the reinforcement.

Glass Fabric/Mica/Epoxy Laminates

The mechanical properties of the glass fabric/mica/epoxy laminates are given Table III. The flexural strength was influenced slightly by the mica content but not in a consistent manner. However, the modulus values showed a considerable improvement of about 30% for the laminate samples having a mica : epoxy resin ratio of 30 : 70, the highest value being shown by the glycidoxy silane-treated mica-containing sample. The interlaminar shear strength values were found to improve on the addition of mica as compared to that of unfilled resin/glass fabric samples. The effect of coupling agents on the mechanical properties of glass fabric/mica/epoxy laminates was not pronounced, which may be attributed to the high percentage of the reinforcement that is glass fabric (70%). However, the addition of

mica as a third component in epoxy/glass fiber laminates does offer advantages like better stiffness, machinability, solder resistance, reduced warpage, and improved adhesion to metal films.

CONCLUSIONS

The following conclusions were arrived at from the above study:

(i) Mica used in the present investigation seems to increase the tensile and flexural moduli considerably.

(ii) The flexural strength increases for 10% mica-filled samples, but at higher loadings, it decreases. The tensile strength, however, showed a decreasing trend at all mica loadings.

(iii) The surface treatment of mica with coupling agents improved the flexural strength, modulus, and tensile modulus.

(iv) Aging in boiling water deteriorated both strength and modulus values of all the samples. However, the percentage retention of properties was better in the surface-treated mica/epoxy samples.

(v) Addition of mica to epoxy/glass fabric laminates resulted in better flexural modulus and interlaminar shear-strength values.

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