

Effect of Coupling Agents on Thermal and Electrical Properties of Mica/Epoxy Composites

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SYNOPSIS

The influence of 3-aminopropyltriethoxysilane, 3-glycidyloxypropyltrimethoxysilane, and neoalkoxytri(dioctyl pyrophosphato)zirconate on thermal expansion behavior, dielectric strength, and arc resistance of mica/epoxy composites has been investigated. The addition of mica up to 30% resulted in the reduction of thermal expansion with respect to neat resin. However, the coefficient of linear thermal expansion of 30% mica treated with aminosilane was the least among the various coupling agent-coated filler/epoxy composites. Mica (30%)/epoxy composites showed the highest dielectric strength values (26 kV/mm), but the highest arc resistance was obtained in zirconate-treated mica (30%)/epoxy composite. © 1995 John Wiley & Sons, Inc.

INTRODUCTION

Thermal and electrical properties are some of the important characteristics of materials. In a polymer, such properties are significantly affected when a filler is added to it. Factors like chemical and physical characteristics of the filler and the nature of interaction between the filler and the polymer also influence these properties.

Thermal expansion behavior is a function of the resin, curing agent, modifiers, and the fillers present in a composite. The lowering in the coefficient of thermal expansion is desirable in minimizing the dimensional changes occurring in a polymer or a composite exposed to temperature changes during fabrication or use. A number of research papers have appeared on the thermal expansion behavior of the composites. The degree of cure of epoxy resin using DSC¹⁻³ and mica/epoxy insulation material has been studied using thermomechanical analysis.⁴ Segal⁵ compared the thermal expansion behavior of particulate and fiber-filled nylon 6 above and below the glass transition temperature and correlated it to the aspect ratio of the reinforcement. Shimbio et al.⁶ observed that particle-particle in-

teraction affects the shrinkage of a composite material. Feltham and co-workers,⁷ on the other hand, reported that the particle size of the fillers did not have any effect on thermal expansion. Aramid fiber-reinforced composites have been found to have a negative thermal expansion character.⁸ The thermal expansion of graphite/epoxy composite materials increased significantly when exposed to moisture.⁹

Reduction of internal stress in composites is important for mechanical stability, which may be achieved by lowering the linear thermal expansion coefficient of the resin, the elastic modulus of the resin, or the T_g of the resin. For example, the dispersion of rubber particles in epoxy resin lowers the elastic modulus and, consequently, low thermal stress is developed.¹⁰

Gupta and Brahatheeswaran¹¹ also showed significant reduction in internal stress in mica flake-filled epoxy resin as compared to neat resin. The presence of mica results in the enhancement of the modulus of the filled resin with a small change in T_g . The coefficient of linear thermal expansion also is reduced in mica/epoxy systems.

The reduction of internal stress in mica/epoxy composites is of considerable interest in electrical and electronic devices. The significance of low thermal stress generation in microelectronic device encapsulation has been emphasized.

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A number of equations have been developed to predict the coefficient of thermal expansion of filled systems,¹²⁻¹⁵ but in each case, it is assumed that the filler particles are isotropic and that the expansion coefficient does not depend on the size and shape of the filler particles. According to Kenyon and Duffey,¹⁴ the degree of adhesion at the interface does not affect the stiffness and dimensional stability. Therefore, the coefficient of thermal expansion, which is stiffness-dominated, should not be influenced by the coupling agents.

Gupta and Brahatheeswaran¹¹ predicted the coefficient of linear thermal expansion, CLTE (β), using the rule of mixtures, according to which the coefficient of the linear expansion of the composite, "Compo," is given as

$$\beta_f \text{Compo.} = \beta_f V_f + \beta_r V_r$$

where β_f and β_r are the coefficients of the linear thermal expansion of the filler and the resin, respectively, and V_f and V_r are their volume fractions.

Thermal stability of a cured epoxy resin system depends on the chemical structure, functionality, and crosslink density of the resin. Generally, epoxies containing aromatic ether or ester links are more stable compared to those containing aliphatic links.¹⁶ Substitution of chlorine or bromine in an epoxy resin has led to improved thermal stability. The OH groups present in the cured system are vulnerable to oxidation. Sometimes, the presence of catalytic centers such as tertiary amines may affect the stability. In addition to the above factors, high residual thermal stresses are often induced during processing at the interface between an epoxy resin and the filler such as metal, glass, fiber, or rubber.¹⁷ Hence, many studies on glass-fiber-reinforced epoxy resins,^{18,19} graphite/epoxy composites with electropolymerized interphase,²⁰ and the thermal behavior and mechanism of residual microstress buildup at the matrix interphase in filled epoxy resins²¹ have been conducted.

Attempts are being made to correlate the long-term aging properties with the data obtained from accelerated aging techniques like TGA. Flynn²² showed that the lifetime of polymers can be accurately predicted by employing the factor-jump method.

Thermogravimetry has been employed to evaluate the thermal resistance of glass-mica insulators with epoxy binders²³ and some correlations have been made between the time and temperature of isothermal aging. Unsworth and Li²⁴ also studied the thermal degradation of epoxy/silica composites through a dynamic mechanical analyzer.

Apart from the thermal properties, the study of electrical properties of polymer composites is also of great significance, especially for electrical and electronic appliances.²⁵ Properties like dielectric constant, dissipation factor, dielectric strength, arc resistance, etc., are of importance for a material to be used in such applications.

The use of mica as a filler in the form of flakes or powder results in improved dielectric strength, reduced dielectric loss, and improved heat resistance and hardness of the composite material.²⁶ Mica-flake-integrated sheet/epoxy laminates have been exclusively employed as insulation for high-voltage electrical appliances.²⁷ In the present investigation, the influence of silane and zirconate coupling agents on the thermal and electrical properties of a mica-filled epoxy system is discussed. Cure kinetics of the mica/epoxy system² and its mechanical properties are reported elsewhere.²⁸

EXPERIMENTAL

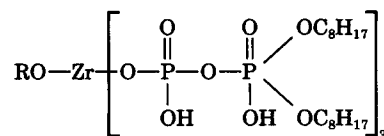
Materials

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 crosslinking agent used was 4,4'-diaminodiphenylmethane.

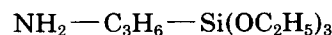
Micronized mica was supplied by Export Linkers, Bihar, India. It is a dry ground product of muscovite type whose particle-size distribution was given in an earlier article.²⁸ The loading was from 10 to 30% by weight.

Three coupling agents were used in the present study:

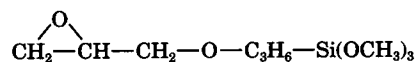
- (i) Neoalkoxy tri(dioctyl pyrophosphato)zirconate, LZ38, a product of Kenrich Petrochemicals, USA:



- (ii) 3-Aminopropyltriethoxysilane:



- (iii) 3-Glycidyloxypropyltrimethoxysilane:



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 g, was prewetted using about 150 mL of toluene, and the coupling agent solution was added to this with vigorous stirring. The slurry was agitated for 4 h and dried at 100°C for 2 h.

Sample Preparation

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 a stoichiometric ratio, i.e., 27 parts by weight per 100 parts of the resin), mixed thoroughly, degassed until the surface became clear, and cast in a metal mold with a Teflon spacer of 1 mm thickness. Silicone spray was used as a mold-releasing agent. The curing was done at 110°C for 2 h, followed by a postcuring for 5 h at 180°C, under nitrogen atmosphere. 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.

Films of about 100 μm thickness were prepared for dielectric measurements by casting mica/epoxy/hardener mixture between the two polished metal plates. Samples containing 60% mica were also prepared by mixing the mica powder with epoxy resin/hardener mixture at room temperature and subjecting them to the cure cycle as mentioned above.

Thermomechanical Analysis

Rectangular bar-shaped samples of dimensions $3 \times 0.1 \times 0.4$ cm were cut from the cast sheets. The thermal expansion was measured with the help of a Stanton Redcroft thermomechanical analyzer Model 691 at a heating rate of 5°C/min in the temperature range from 25 to 250°C. The coefficient of linear thermal expansion (CLTE), β , was calculated as

$$\beta = \frac{\Delta L}{L\Delta T} \quad (1)$$

where L is the initial length; ΔL , the change in length during heating; and ΔT , the change in temperature.

Differential Scanning Calorimetry

Disc-shaped samples of thickness 1 mm and weighing about 20 mg were used for the DSC studies. A Perkin-Elmer Delta Series DSC 7 was used to measure the glass transition temperature of the samples. The scanning was done under a nitrogen atmosphere at a heating rate of 10°C/min. DSC traces were obtained as plots of the rate of heat flow as a function of temperature. The midpoint of the inflection of the curve was taken as the glass transition temperature.

Thermogravimetric Analysis

Thermal degradation of the mica/epoxy samples was carried out with a DuPont 1090 thermogravimetric analyzer, at a heating rate of 10°C/min under nitrogen atmosphere from ambient to 500°C (after which point no change in weight was observed).

The data obtained by TGA were expressed in the form of the following parameters:

- (i) Initial decomposition temperature is the temperature at which the actual decomposition of the sample starts, referred to as T_i .
- (ii) Maximum decomposition temperature, T_p , is the temperature at which the rate of weight loss is maximum.
- (iii) Final decomposition temperature, T_f , at which the major decomposition is completed.
- (iv) Percentage residue is the amount of residue left at 500°C.

Dielectric Measurements

A Hewlett-Packard 4192 A impedance analyzer was used for measuring the dielectric constant and dissipation factor. The measurements of aluminum-coated epoxy samples were carried out at ambient temperature (25°C) in the frequency range of 10 kHz to 10 MHz. The capacitance and dissipation factor ($\tan \delta$) were directly read from the instrument. The dielectric constant (ϵ') was calculated from the capacitance value using the following relationship:

$$\text{Dielectric constant, } \epsilon' = \frac{Cd}{\epsilon_0 \cdot A} \quad (2)$$

where C is the measured capacitance; d , the thickness of the sample; A , the area of the sample; and ϵ_0 , the permittivity of air.

Table I Coefficient of Linear Thermal Expansion for Mica/Epoxy Samples

Sample	Mica (%)	CLTE ($\times 10^5$) (m/m/°C)	
		30–50°C	200–220°C
Neat		8.65	53.70
UT	10	5.89	14.30
	20	5.15	11.70
	30	4.48	10.90
LZ	10	5.66	13.40
	20	5.05	11.30
	30	4.31	10.20
AS	10	5.50	12.90
	20	4.73	11.20
	30	4.20	10.00
GS	10	5.70	13.00
	20	4.83	11.20
	30	4.36	10.10

UT = untreated, LZ = zirconate-treated sample, AS = aminosilane-treated sample, GS = glycidylsilane-treated sample.

Dielectric Strength and Arc Resistance

The dielectric strength was measured on a Shanto high-voltage breakdown tester (capacity 7.5 kV) as per ASTM D149. The voltage at which the puncture occurred was noted and the dielectric strength was calculated as follows:

$$\text{Dielectric strength, } S = \frac{V_a}{d} \quad (3)$$

where V_a is the applied voltage at puncture, and d , the thickness of the sample. Arc resistance was measured using samples of 3 mm thickness at a voltage of 12.5 kV according to ASTM D495.

RESULTS AND DISCUSSION

Thermomechanical Analysis

Values of the coefficient of linear thermal expansion (CLTE) are given in Table I. It may be seen that the addition of 10% of mica reduced the thermal expansion values significantly (Fig. 1). Addition of mica up to 30% resulted in further reduction but to a lesser extent. In the case of the coupling agent-treated mica/epoxy samples, the reduction was slightly more, which may be due to improved dispersion of mica particles as coupling agents to en-

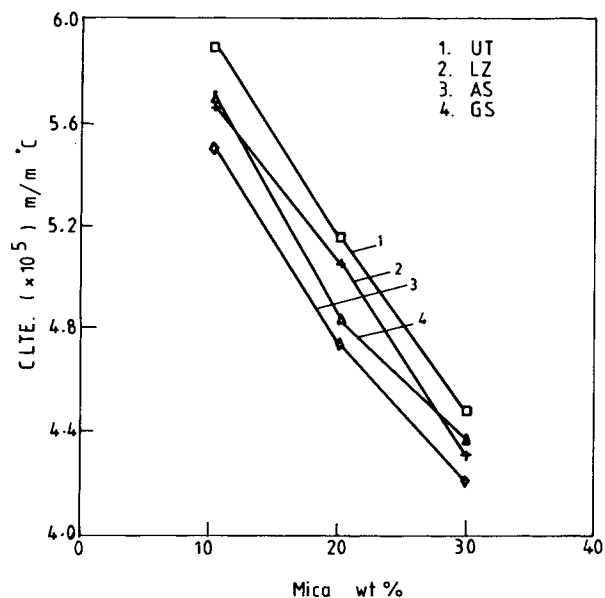


Figure 1 Plots of CLTE vs. mica loading below T_g (30–50°C) for mica/epoxy samples.

hance the wettability of the filler surface, which was confirmed by contact angle studies. The maximum reduction for the 30% mica/epoxy sample was observed for the aminosilane-treated sample followed by zirconate- and glycidyl silane-treated samples. Figure 2 shows the plots of CLTE vs. mica content measured above the T_g , i.e., the rubbery state (200–220°C). Here, the effect of the coupling agent appears to be more prominent, perhaps due

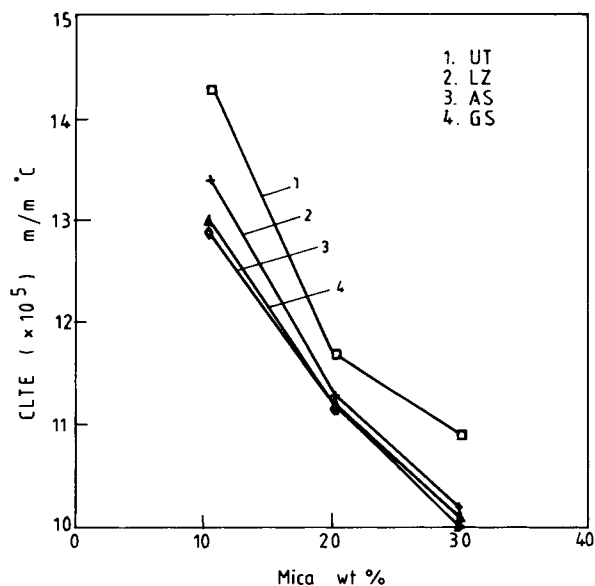


Figure 2 Plots of CLTE vs. mica loading above T_g (200–220°C) for mica/epoxy samples.

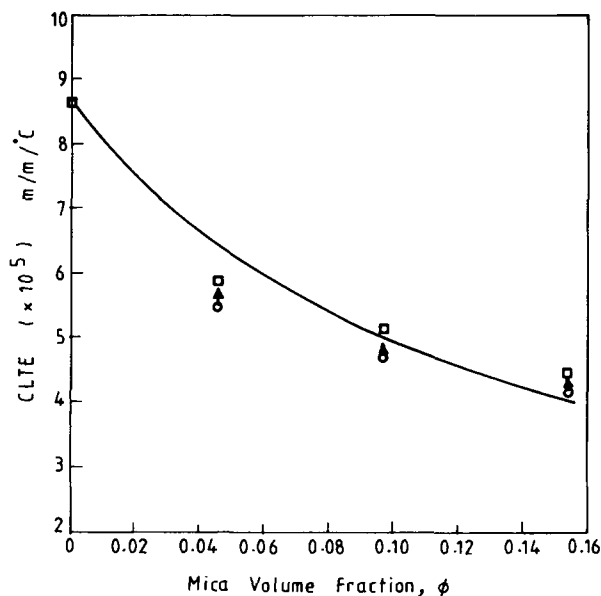


Figure 3 Plots of CLTE vs. mica volume fraction: (points) experimental; (solid line) calculated using Morimoto's equation.

to low internal (curing) stresses developed. Using the Wang and Kwei²⁹ refined rule of mixtures, the coefficient of linear expansion of the mica/epoxy composite was also calculated. The experimental values seem to be closer to the predicted values (Fig. 3).

Differential Scanning Calorimetry

The DSC traces of 60% untreated and surface-treated samples are shown in Figure 4. Both the onset and glass transition temperatures showed a decreasing trend as the mica content increased (Table II). There is a reduction of about 12°C for the 60% mica-filled epoxy sample compared to that of the neat resin. Similar observations have been cited in the literature for the filled epoxy systems.^{30,31} Filyanov³² and Petrovic and Stozakovic³⁰ suggested that reduction in the T_g of an epoxy in the presence of a glass filler might be due to the possible reaction occurring between the filler and the resin, resulting in reduction of the amount of hardener consumed in the curing process. In the present study, the reduction in T_g might be explained as follows: As mica acts as a catalyst for the epoxy cure and helps in increasing the cure rate significantly at the initial stage,² the nature of the network thus formed could be considerably altered due to the relatively high rate of crosslink formation and also the crosslink density.

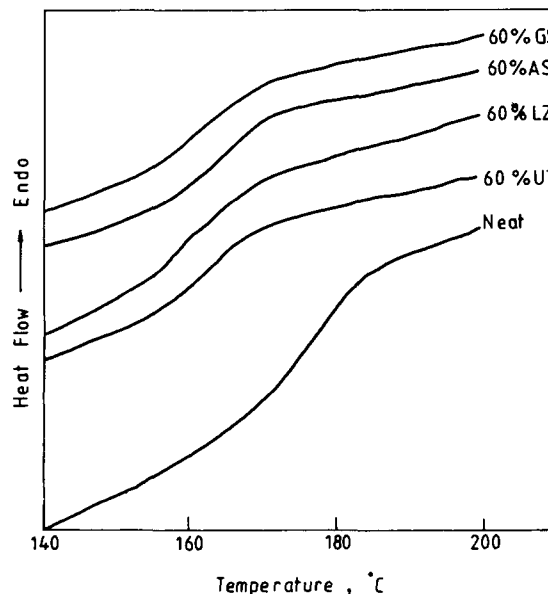


Figure 4 DSC traces of neat resin and 60% untreated and surface-treated mica/epoxy samples.

The surface modification of mica by coupling agents does not show any significant effect on the T_g values. Although all the T_g values corresponding to the coupling agent-treated mica (60%)/epoxy sample were found to be slightly higher as compared to that of untreated mica/epoxy sample, the difference, however, is not significant ($\sim 2^\circ\text{C}$ for the aminosilane-treated mica/epoxy sample) to draw any conclusion.

Thermogravimetric Analysis

The plots of percentage weight loss with respect to heating in nitrogen atmosphere are shown in Figures 5 and 7. It can be observed that all the samples have undergone a single-stage decomposition process with the major weight loss occurring between 300 and 500°C. The initial peak and final decomposition

Table II DSC Data for Mica/Epoxy Samples

Sample	Mica Filler (%)	Onset of T_g ($^\circ\text{C}$)	T_g ($^\circ\text{C}$)
Neat	0	167.2	172.8
UT	10	165.9	170.8
	20	163.0	167.4
	30	163.4	168.7
	60	155.9	160.7
LZ	60	156.2	161.0
AS	60	159.7	163.1
GS	60	156.9	161.9

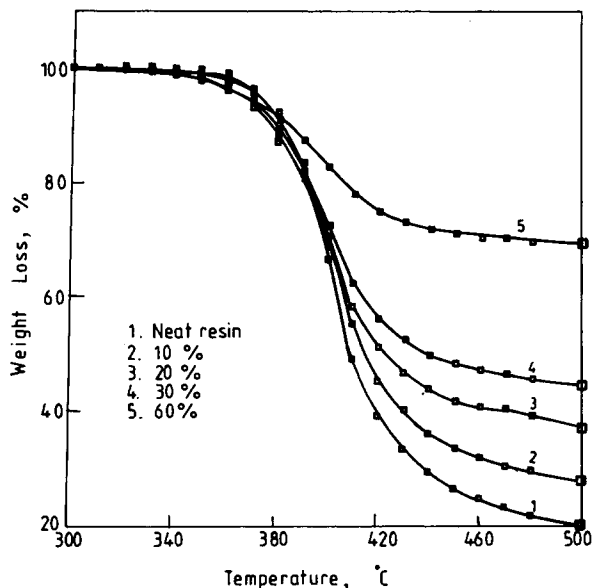


Figure 5 Plots of percentage weight loss vs. temperature for neat and untreated mica/epoxy samples.

temperatures and percentage residue for all the samples are given in Table III. Addition of 60% mica has lowered the initial and peak decomposition temperatures by about 12°C compared to the neat resin. However, the final decomposition temperature did not show any change for neat resin and 60% mica-loaded sample. As seen in Figure 6, mica accelerates the decomposition process at the initial stages but the rate at the maximum decomposition was significantly lower than that of the neat resin. The residue obtained (Table III) from these samples was directly proportional to the resin fraction, which implies that mica, being an inert material at the temperature studied, did not participate in the reaction.

The rate of decomposition at T_p showed a decreasing trend as the mica content increased. The rate of decomposition at T_p further dropped in treated mica epoxy samples.

The following Arrhenius-type equation was used to obtain the kinetic parameters of the decomposition process:

$$dW/dt = A \exp(E_a/RT) W^n \quad (5)$$

where dW/dt is the rate of weight loss; A , the pre-exponential factor; E_a , the activation energy of thermal decomposition; n , the order of the reaction; and W , the weight at time "t."

Both the activation energy and the order of reaction showed a decreasing trend with increasing mica percentage (Table IV). It was found that mica

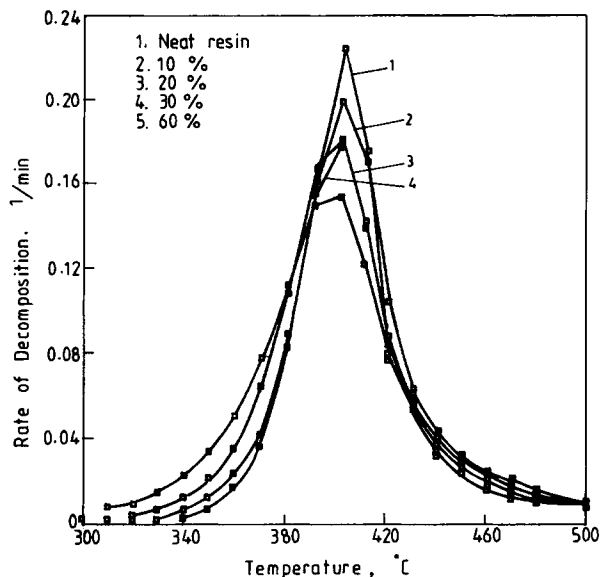


Figure 6 Plots of rate of decomposition vs. temperature for neat and untreated mica/epoxy samples.

accelerated the epoxy resin cure² and decreased the T_g of the cured resin. From these observations, it can be concluded that mica affects the structural properties of epoxy composites, such as crosslink density. The structural change occurring during the network formation could also be a factor which affects the thermal decomposition behavior of mica/epoxy composites.

The plots of percentage weight loss and rate of decomposition vs. temperature for the coupling

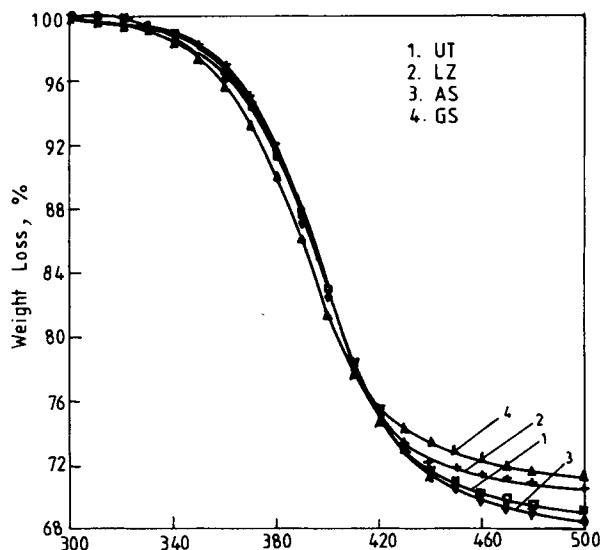


Figure 7 Plots of percentage weight loss vs. temperature for 60% untreated and surface-treated mica/epoxy samples.

Table III Thermogravimetric Studies of Untreated, and Coupling Agent-Treated Mica/Epoxy Samples

Sample	Mica (Wt %)	Decomposition Temp			Residue (%)	Residue of Resin Component (%)
		T_i	T_p	T_f		
Neat resin	0	381.2	400.9	427.8	16.78	16.78
UT	10	379.1	402.5	431.1	24.73	14.73
	20	373.1	399.0	429.9	33.58	13.58
	30	373.4	398.3	428.8	40.71	10.71
	60	368.9	387.2	427.7	66.81	6.81
	LZ	60	368.4	395.6	422.6	68.75
AS	60	365.8	397.5	428.9	66.74	6.74
GS	60	367.6	391.4	422.6	67.08	7.08

agent-treated mica (60%)-filled epoxy samples are shown in Figures 6 and 8. The data obtained and the kinetic parameters calculated are listed in Tables III and IV. All the coupling agent-treated mica (60%)/epoxy samples had higher peak temperatures as compared to the untreated 60%/epoxy sample. The highest T_p was shown by the aminosilane-treated mica/epoxy sample (397.5°C) which is almost close to that of the neat resin. However, the percentage residue and T_p were higher in the case of the zirconate-treated mica/epoxy sample. The activation energy and the order of reaction values were higher for the glycidylxysilane-treated mica/epoxy sample compared to the other samples. The differences in the decomposition behavior of all these samples might have arisen from the structural changes imparted by mica with different surface treatments during the curing process.

Electrical Properties

Dielectric Measurements

The dielectric constant (ϵ') and dissipation factor ($\tan \delta$) values obtained for the neat resin, untreated,

Table IV Kinetic Parameters Obtained from Thermogravimetric Analysis

Sample	Mica (Wt %)	E_a (kJ/mol)	Order n	Rate at T_p (min^{-1})
Neat resin	0	113.2	4.57	0.226
UT	10	108.5	4.41	0.199
	20	105.8	4.44	0.178
	30	95.2	4.29	0.182
	60	57.6	2.74	0.158
	LZ	60	56.6	2.35
AS	60	56.7	2.73	0.155
GS	60	61.2	3.22	0.147

and coupling agent-treated mica/epoxy samples at different frequencies are given in Table V. The dielectric constant and dissipation factor values show an increasing trend as the mica content increases. This is expected, since muscovite mica has a higher dielectric constant value (6–7.5) than that of the epoxy resin. With increasing frequency, the dielectric constant decreased but the dissipation factor increased at 100 kHz. However, it decreased when the frequency was further increased to 10 MHz in mica/epoxy samples. The coupling agent treatment of mica did not seem to have a noticeable effect on either the dielectric constant or the dissipation factor.

Table V shows the ϵ' and $\tan \delta$ values measured for these samples after boiling in water for 2 h. Of

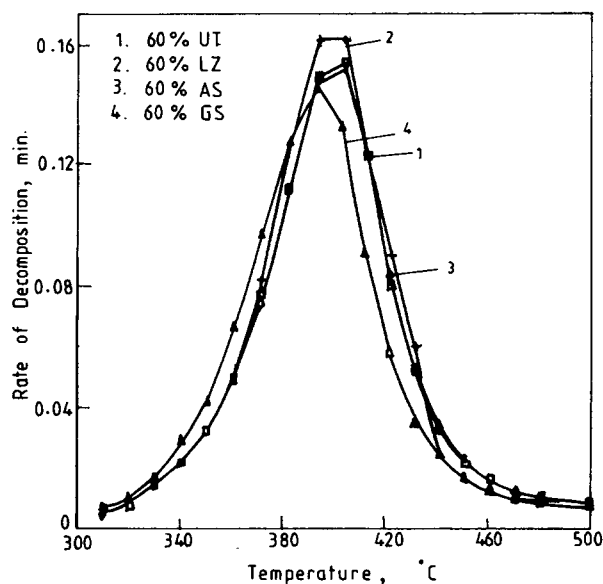
**Figure 8** Plots of rate of decomposition vs. temperature for 60% untreated and surface-treated mica/epoxy samples.

Table V Dielectric Constant and Dissipation Factor Mica/Epoxy Samples

Sample	Dielectric Constant, ϵ'				tan δ		
	Mica	1 kHz	100 kHz	10 mHz	1 kHz	100 kHz	10 mHz
Neat		4.92	4.71	4.47	0.006	0.023	0.024
UT	10	4.48	4.58	4.34	0.019	0.039	0.022
	20	5.19	4.66	4.39	0.028	0.042	0.049
	30	6.13	5.24	4.81	0.047	0.055	0.029
LZ	30	6.21	5.30	4.70	0.051	0.056	0.031
AS	30	5.99	5.27	4.71	0.049	0.056	0.031
GS	30	6.01	5.32	4.99	0.05	0.054	0.029
After boiling in water for 2 h							
Neat		5.1	5.01	4.78	0.010	0.035	0.0270
UT	30	10.36	9.57	9.2	0.186	0.143	0.2130
LZ	30	7.17	7.02	6.7	0.076	0.079	0.1020
AS	30	6.98	6.17	6.01	0.073	0.076	0.0990
GS	30	6.79	6.35	6.2	0.081	0.088	0.0970

all these samples, the neat resin sample suffered minimum damage. Both untreated and coupling agent-treated mica (30%)/epoxy samples were affected by the exposure to boiling water. However, it is worth noting that the coupling agent-treated mica/epoxy samples were affected to a lesser extent as compared to the untreated mica/epoxy sample. Similar observations have been made by Marsden and Zieminsky³² for silane-treated wollastonite/polyester composites. The improvement in retention of these properties might be due to the combined effect of hydrophobicity imparted by the coupling agent and also improved interfacial bonding, thus making the resin-mica interface less moisture-sensitive.

Dielectric Strength and Arc Resistance

The dielectric strength and arc resistance values measured for neat resin, untreated, and coupling agent-treated mica/epoxy samples are given in Table VI. It is seen that as the mica content increases both dielectric strength and arc resistance increase considerably. The untreated 30% mica/epoxy sample had the highest dielectric strength value (26 kV/mm), while the highest arc resistance was shown by zirconate-treated 30% mica/epoxy sample.

CONCLUSIONS

- The thermal expansion was significantly reduced by the addition of mica; 30% mica/epoxy

samples showed a twofold reduction in thermal expansion.

- The glass transition temperature is reduced in the case of mica-filled samples to a considerable extent.
- The thermal stability observed in terms of weight loss as a function of temperature was also affected by the addition of mica.
- Addition of mica increased both the dielectric constant and the dissipation factor at all frequencies. The effect of coupling agents was realized when the samples were exposed to boiling water treatment for 2 h. The retention of properties was better in the case of the coupling agent-treated mica/epoxy samples.
- Both dielectric strength (breakdown voltage) and arc resistance were improved significantly

Table VI Dielectric Strength and Arc Resistance Values for Mica/Epoxy Compositions

	Mica (Wt %)	Dielectric Strength (kV/mm)	Arc Resistance (s)
Neat		17	24
UT	10	21	36
	20	22	72
	30	26	75
LZ	30	24	76
AS	30	23	73
GS	30	22	72

with the addition of mica. However, coupling agents did not influence these properties.

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