

Flexural, Compression, Chemical Resistance, and Morphology Studies on Granite Powder-Filled Epoxy and Acrylonitrile Butadiene Styrene-Toughened Epoxy Matrices

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ABSTRACT: Granite powder is an inexpensive material that can reduce the overall cost of a composite if used as a filler in epoxy and acrylonitrile butadiene styrene (ABS)-toughened epoxy matrices. Epoxy and ABS-toughened epoxy resins filled with granite powder were cast into sheets. To enhance the properties of these composites, granite powder was treated with triethoxymethyl silane coupling agent. Flexural properties, compression properties, chemical resistance, and morphology of these composites were studied. The filler used varied from 0 to 60 wt %. Composites consisting of ABS-toughened epoxy with treated granite powder were found to be superior in mechanical properties to composites with treated and untreated granite

powder. Composites with 50 wt % of granite powder was found to have maximum mechanical properties in all cases. All the three composites, i.e., untreated, treated and ABS toughened composites showed good resistance toward acids, alkalis, and solvents. Treating granite powder with silane coupling agent enhances its mechanical properties and improves the interfacial bond between granite powder and the matrix. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 104: 171–177, 2007

Key words: epoxy resin; granite powder; filler; flexural strength; compression strength; chemical resistance; SEM

INTRODUCTION

Epoxy resins are considered to be the most important class of thermosetting polymers. They have found remarkable applications as bonding agents and adhesives, protective coatings, electrical laminates, apparel finishes, fiber-reinforced plastics, flooring and paving, and composites pipes. Once cured, epoxy resins are characterized by high resistance to chemicals and corrosion. However, in all these applications, epoxy resins have one major disadvantage: they are very brittle, with poor resistance to crack propagation and low impact strength.¹ Our aim is to toughen these brittle polymers without significantly decreasing other important properties they possess. Many studies have been carried out with the addition of suitable rubber such as liquid amine-terminated,² carboxyl-terminated,³ and hydroxyl-terminated⁴ copolymers of butadiene, and acrylonitrile. In general, toughness can be achieved when elastomer particles are dispersed on a micro level. However, toughness improvements in most elastomer-modified epoxy systems usually result in a significant decrease in the modulus and glass transition temperature (T_g) of the cured resin.

Recently, many attempts have been made to modify epoxy resins with engineering thermoplastic polymers such as polysulfone,⁵ polyetherimide,⁶ and polyimide.⁷ Significant improvement in toughness is obtained only at higher thermoplastic content, where the thermoplastic polymer forms a continuous phase with the epoxy spherical domain, or the thermoplastic polymer and epoxy form a cocontinuous phase. However, epoxy resins modified with higher thermoplastic content show a steep increase in viscosity which causes difficulty in handling the resin. In addition, the T_g of the resins modified with the thermoplastic polymer is equal to or higher than that of the unmodified resin.

Although composite materials are usually more expensive than conventional building material on a weight to weight basis, they are more economical in the long run, considering their properties, high strength to weight ratio, modulus to weight ratio, and good impact and chemical resistance. Fillers are added to the epoxy matrix system to improve handling, molding characteristics, cured properties, and to reduce the overall cost of the system. Almost any powdered material can be used as a filler. Most commonly used fillers are various grades of CaCO_3 , silica flour, talc, various kinds of clay,⁸ and, recently, flyash.⁹

Granite powder is a byproduct of granite cutting industries. Granite powder is obtained during sizing of granite slabs. Since granite powder is an environ-

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mental hazard, we need to find appropriate methods for its use and disposal. Further, granite powder-filled thermosetting composites are more economical to produce than the original thermosets. However, not much work has been reported to date on the utilization of granite powder in thermosets as filler. So, the authors thought that if granite powder can be used as filler, it would be beneficial in reducing the overall cost of composites, and it would be useful for proper disposal of granite powder, which is otherwise a hazard to the surrounding environment.

Many studies have been published concerning the mechanical properties of polymer composites with a variety of fillers. Raj et al.¹⁰ studied the effect of cellulose-based filler in polymeric materials. Kaolin has been used as filler, and also as toughening agent for unsaturated polyester and epoxy resin, and the properties of these composites have been studied.^{11,12}

Mechanical properties, water absorption, and chemical resistance of the composites are important, since these materials are continuously exposed to different chemicals during their end use. So, the authors thought that it would be useful if studies were carried out in this direction.

EXPERIMENTAL

Materials and Methods

For the present study, a commercially available epoxy resin DGEBA (LY556) was procured from M/s Vantico Ltd. (Mumbai), and the Hardener HY 951 (triethylene tetramine) was used as the polymer matrix. Triethoxy methyl silane used as the coupling agent for modifying granite powder was procured from E. Merck (India). Acrylonitrile butadiene styrene (ABS) was obtained from Absolac (India) as unfilled grade. Granite powder was obtained from locally available granite industries. The particle size of the granite powder ranges from 1 to 100 μm . The density of the granite powder was found to be 2.196. The chemical composition of the granite powder is given in Table I. The powder was washed thoroughly with water and dried in an oven at 100°C for 2 h to remove any moisture content before using it as a filler for epoxy resin. For blending ABS and epoxy, ABS pellets were dis-

solved in dichloromethane. The solution of dichloromethane containing ABS was poured into a beaker containing the required amount of epoxy resin. The amount of ABS in epoxy resin was varied from 2 to 10% by weight. Above this content, the epoxy resin becomes highly viscous and is very difficult to process. The resin to hardener mixture was maintained to 100:10 parts by weight. The amount of ABS in epoxy resin was varied from 2 to 10% by weight. Then the dichloromethane was evaporated by heating at a temperature of 80°C. After complete evaporation of dichloromethane, the mixture of ABS and epoxy resin were stirred at 120°C continuously for 1 h, and then cooled to room temperature. The degassed mixture of ABS and epoxy was mixed with a calculated amount of hardener by continuous stirring, and then poured into molds. The epoxy and hardener system was cured at room temperature for 24 h, and then post-cured at 100°C for 2 h to ensure complete curing.

Fabrication

Teflon sheet of 10 mm thickness with dimensions 26 \times 13 \times 0.3 cm^3 , grooved using CNC milling machine, was employed to cast flexural specimens. Teflon sheet was employed as a mold as it is nonsticky in nature. Epoxy resin with required amount of hardener was poured into the mold. The mold was allowed to stand for 24 h at room temperature for complete curing. A sample sheet was taken out and cut to the required specimen size according to ASTM standards. All the samples were postcured at 100°C in an oven for 2 h to ensure complete curing before being subjected to mechanical tests. In a similar way, blends of ABS/epoxy were made by varying ABS content in the epoxy. The composites of untreated, coupling agent-treated, and 4% ABS-toughened epoxy/coupling agent-treated granite powder-filled composites were prepared by varying the granite powder content by weight percentage. These were cast into sheets, and specimens were cut out. For making samples for impact test, a steel mold with dimensions of 100 \times 65 \times 10 mm^3 was employed. The inner cavity of the steel mold was coated with wax. In each case, ten specimens of each type were made, and the average value was reported.

Flexural strength measurement

Flexural strength was determined using Lloyd's LR 100 K type universal testing machine as per ASTM D 790-procedure. Test speed was maintained between 1.3 and 1.5 mm/min.

Compression strength measurement

Compression strength measurement is carried out on a Lloyds LR 100 K type computer-controlled universal

TABLE I
Chemical Composition of Granite Powder

Compound	wt %
Silica	70–77
Alumina	11–13
Potassium oxide	3–5
Soda	3–5
Lime	1
Iron	2–3
Magnesia and Titania	< 1

TABLE II
Flexural and Compression Properties of ABS/Epoxy Blends

% of ABS content in the matrix	Flexural properties		Compression properties	
	Flexural strength (MPa)	Flexural modulus (MPa)	Compression strength (MPa)	Compression modulus (MPa)
0	45.86	640	48.36	2931
2	49.70	603	83.35	2120
4	52.70	816	96.09	2762
6	47.40	989	91.45	3141
8	44.04	1014	87.13	3216
10	37.63	1123	80.09	3914

testing machine. The compression test was performed on ten sets of specimens of differing granite powder content as per ASTM D-695 standard. The machine cross head speed was maintained at 5 mm/min. From the load stroke history, the compression modulus and strength were determined.

Chemical resistance test

Chemical resistance of the composites was studied as per ASTM D 543 method. For this purpose three strong acids—conc. HCl, conc. HNO₃, and glacial acetic acid; two bases viz., aqueous solutions of 40% NaOH, and 20% Na₂CO₃; three organic solvents benzene, carbon tetrachloride, and toluene were selected. Preweighed samples were dipped in chemicals for 24 h, removed, washed thoroughly in distilled water, and dried immediately by pressing them on both sides by filter paper. From the final weight of the samples, percentage weight loss/gain was determined. The chemical test was repeated for ten samples in each case, and the average value was reported.

Scanning electron microscopy

Impact-fractured samples were examined in a JEOL JSM 840A type scanning electron microscopy. The samples were wrapped in a silver foil and the fracture

surface was coated with gold under vacuum in an ion-sputtering unit to make them conducting.

RESULTS AND DISCUSSION

The flexural properties of ABS/epoxy blend as a function of ABS content in the epoxy are presented in Table II. From this table, it is concluded that the flexural strength of ABS/epoxy blend increases up to 4 wt % of ABS content in the epoxy and then found to decrease on further addition of ABS plastic. Flexural strength was increased by 12.9% over the pure epoxy matrix when epoxy contains 4 wt % of ABS. Further, the flexural strength was found to decrease as the ABS content increased. This decrease is mainly due to the dispersed phase of ABS, which hinders the crosslinking of epoxy with the amine hardener. The decrease in strength and modulus may be explained by the plasticizing influence of the dispersed phase of ABS particles within the epoxy network. This creates weak sites, reducing the effective cross section that bears the load, causing a reduction in strength.

The flexural properties of untreated, coupling agent-treated, and 4% ABS-toughened epoxy/coupling agent-treated granite powder composites are given in Table III. From this data, it is confirmed that the flexural strength of untreated, coupling agent-treated, and 4% ABS-toughened epoxy/coupling agent-treated

TABLE III
Flexural Strength, Modulus of Untreated, Coupling Agent-Treated, and ABS-Toughened Epoxy/Coupling Agent-Treated Granite Powder Composites

% of filler content in the matrix	Epoxy/untreated granite powder composites		Epoxy/coupling agent-treated granite powder composites		ABS-toughened epoxy/coupling agent-treated granite powder composites	
	Flexural strength (MPa)	Flexural modulus (MPa)	Flexural strength (MPa)	Flexural modulus (MPa)	Flexural strength (MPa)	Flexural modulus (MPa)
0	45.86	640	45.86	640	52.08	816
30	47.05	1505	52.80	1711	57.66	1565
40	54.33	2222	66.76	4062	68.67	1833
50	68.95	3174	78.92	5126	88.52	2656
60	62.53	3483	71.44	6617	58.46	2660

granite powder composites increase linearly up to 50 wt % of granite powder content, and then decrease on further addition of granite powder. This decrease in flexural strength may be due to the non-wetting of the granite powder particles with the matrix, and the formation of agglomeration of the granite powder particles. The initial increase in flexural strength from 30 to 50% of granite powder content was mainly due to the good filler matrix interaction, which was largely due to the platy nature of the fillers. Platy fillers have high aspect ratios, and this increases the wettability of the fillers by the matrix, thus creating fewer microvoids between the fillers and the matrix.¹³ The decrease in flexural strength on addition of granite powder from 50 to 60% is caused due to the smaller particle size of granite powder. The granite powder, which has a higher surface area due to its small particle size, tends to agglomerate, and this results in the reduction of surface interaction between the epoxy and the granite powder. This explains the reduction in the flexural strength of granite powder-filled epoxy composites at higher filler loadings. The percentage increment of flexural strength of untreated granite powder composites over pure epoxy matrix was found to be 33.48% for 50% filler loading, which is an optimum filler content in the matrix. The increment in flexural strength of coupling agent-treated granite powder composites was about 41.89% over pure epoxy matrix. Both coupling agent-treated granite powder composite and untreated granite powder composites contain 50 wt % of filler loading. The flexural strength of the former was found to be 12.63% higher than that of the latter. This indicates that good interfacial bond exists between the treated granite powder and epoxy resin. In the case of 4% ABS-toughened epoxy/coupling agent-treated granite powder composites, flexural strength increases linearly up to 50 wt %, and then decrease as in the case of other two types of composites. The flexural strength of granite powder-filled 4% ABS/epoxy blend matrix composites increased upto 50 wt % of filler content, showing 48.19% increment over pure epoxy matrix and

41.16% over pure ABS/epoxy blend. The increment of flexural strength of 4% ABS-toughened epoxy/coupling agent-treated granite powder composites over untreated and coupling agent-treated granite powder composites was about 22.10 and 10.84% respectively, corresponding to 50 wt % of granite powder loading.

The flexural modulus data are presented in Table III. This data indicates that the flexural modulus of all the types of composites are found to increase linearly on increasing the granite powder content. Coupling agent-treated granite powder-filled composites show higher modulus indicating that these composites are stiff in nature. It is a well-known fact that incorporation of rigid particulate fillers improves the stiffness of plastic matrices.¹⁴ 4% ABS-toughened epoxy/coupling agent-treated granite powder composites show decreased modulus when compared to those of untreated and coupling agent-treated granite powder-filled composites. This reduction in modulus is due to the dispersed phase of ABS. As reported earlier, elastomer-modified epoxy systems usually result in a significant decrease in the modulus and the glass transition temperature of the cured epoxy resin.

The compression strength of ABS/epoxy blend is given in Table II. It was found that the compression strength increased up to 4 wt % of ABS content, and decreased linearly on increasing the ABS content in the epoxy. The 4 wt % of ABS in epoxy blend shows 49.67% increment in compression strength over pure epoxy matrix. The decrease in compression strength on increasing the ABS content is again due to the plasticizing effect caused by the dispersed phase of ABS in epoxy-rich matrix. The compression modulus of these ABS/epoxy blends increased linearly on increasing the ABS content. The compression properties of all the types of composites studied are given in Table IV. From these data, the compression strength of all the composites is found to increase gradually on increasing the granite powder content. The compression strength of untreated granite powder composites corresponding to 50 wt % filler loading shows 43.36% increment over pure epoxy matrix. The percentage incre-

TABLE IV
Compression Strength, Modulus of Untreated, Coupling Agent-Treated, and ABS-toughened Epoxy/Coupling Agent-Treated Granite Powder Composites

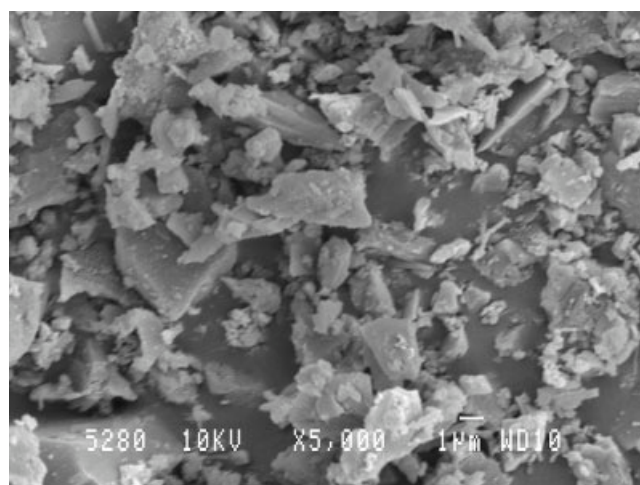
% of filler content in the matrix	Epoxy/Untreated granite powder composites		Epoxy/coupling agent-treated granite powder composites		ABS-toughened epoxy/coupling agent-treated granite powder composites	
	Compression strength (MPa)	Compression modulus (MPa)	Compression strength (MPa)	Compression modulus (MPa)	Compression strength (MPa)	Compression modulus (MPa)
0	48.36	2931	48.36	2931	96.09	2762
30	66.22	3542	77.88	4172	88.57	5111
40	73.43	3978	96.77	4395	109.49	6063
50	80.93	4085	106.68	5054	115.25	7868
60	85.39	4192	114.65	5369	123.1	9156

TABLE V
Chemical Resistance Properties of Untreated and Coupling Agent-Treated Granite Powder/Epoxy Composites

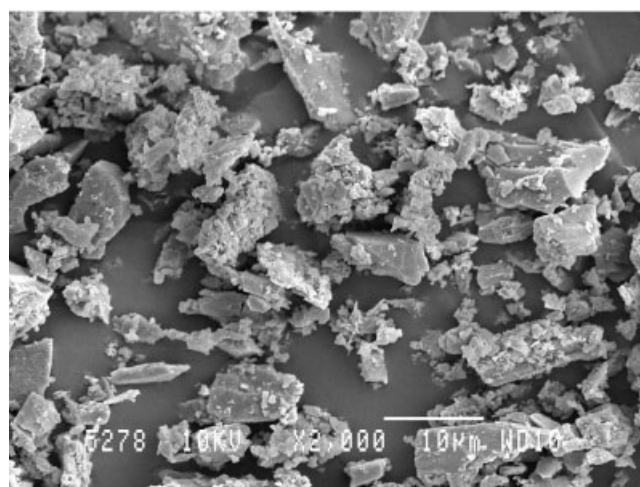
Chemicals	Percentage of filler content in matrix by weight							
	30		40		50		60	
	Untreated	Treated	Untreated	Treated	Untreated	Treated	Untreated	Treated
Acetic acid	0.6029	0.5994	0.5496	0.5301	0.5215	0.5121	0.5111	0.4981
Hydrochloric acid	1.2936	1.1136	1.1894	1.1711	1.1798	1.1634	1.1674	1.1598
Nitric acid	Dissolved	Dissolved	Dissolved	Dissolved	Dissolved	Dissolved	Dissolved	Dissolved
Sodium hydroxide	0.1761	0.1608	0.1222	0.1121	0.1022	0.0911	0.0934	0.0892
Sodium carbonate	0.3941	0.3021	0.3143	0.3001	0.2154	0.1914	0.1923	0.1811
Benzene	0.0753	0.0701	0.0643	0.0612	0.0101	0.0010	0.0091	0.0082
Carbon tetrachloride	No change	No change	No change	No change	No change	No change	No change	No change
Toluene	0.0816	0.0700	0.0698	0.0601	0.0530	0.0492	0.0521	0.0501
Lactic acid	0.3996	0.3111	0.2346	0.2009	0.1163	0.9636	0.1041	0.0943

ment of these composites increased from 26.97 to 43.36% on increasing the granite powder content in the matrix. The compression strength of coupling agent-treated granite powder composites increases on increasing the granite powder content. The percentage increment of compression strength corresponding to 60 wt % filler content over pure epoxy matrix was found to be 57.81%. The percentage increment of coupling agent-treated granite powder composites increased from 37.90 to 57.81% over pure epoxy matrix, indicating good dispersion of the granite powder particles due to the coupling agent treatment. The silane-treated granite powder composites show increased compression strength over untreated granite powder, indicating a good interfacial bond between the granite powder and epoxy. The increment of strength was about 14.97 to 25.52% linearly over those of untreated granite powder composites. In the case of 4% ABS-toughened epoxy/coupling agent-treated granite powder composites, the strength increased linearly on increasing the granite powder content. The increment in compression strength of the composite of maximum filler fraction (60 wt %) was about 30.63 and 6.86% over untreated and coupling agent-treated granite powder composites, respectively. Overall, 4 wt % ABS-tough-

ened epoxy/coupling agent-treated granite powder composites show higher compression strength over other two types of composites.



(a)

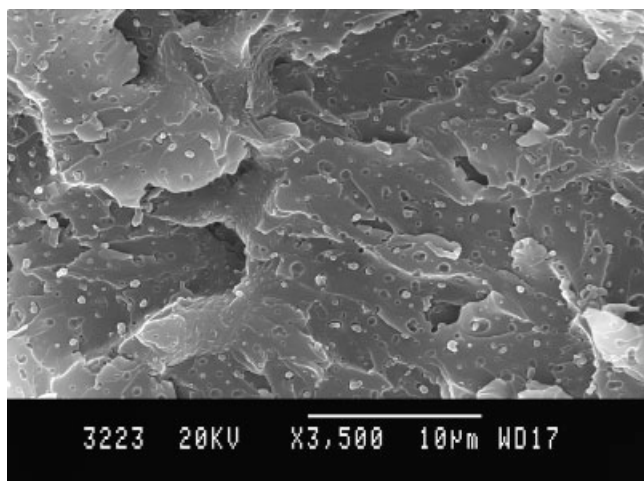


(b)

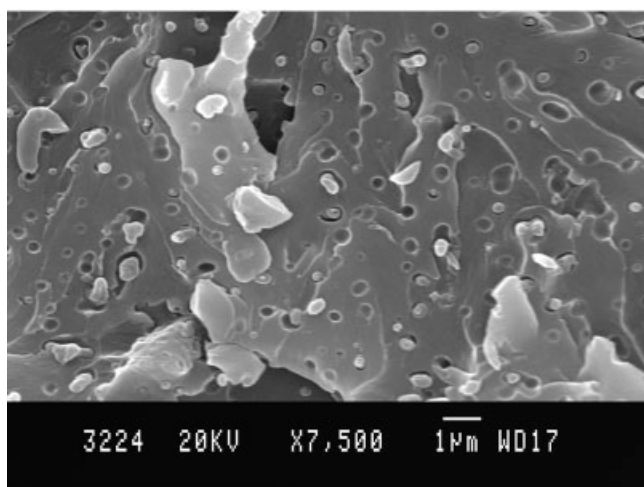
TABLE VI
Chemical Resistance of Coupling Agent-Treated Granite Powder-Filled 4% ABS-Toughened Epoxy Blend Matrix Composites

Chemicals	Percentage of filler content in the matrix by weight			
	30	40	50	60
Acetic acid	0.3798	0.9394	0.7848	0.4827
Hydrochloric acid	0.5120	0.5703	0.5392	0.8256
Nitric acid	35.57	38.493	23.1367	17.38
Sodium hydroxide	-0.0127	-0.0112	0.0130	0.0192
Sodium carbonate	0.1263	0.1347	0.0668	0.0709
Benzene	0.0838	0.0616	0.0318	-0.0319
Carbon tetrachloride	0.0622	0.0714	0.0104	0.0179
Toluene	0.0769	0.1009	0.0001	-0.0008
Lactic acid	0.2224	0.3618	0.1708	0.1113

Figure 1 Scanning electron micrographs of granite powder particles at different magnifications: (a) $\times 2000$, (b) $\times 5000$.



(a)



(b)

Figure 2 Scanning electron micrographs of impact-fractured samples of 4% ABS-toughened epoxy blend matrix at different magnifications: (a) $\times 3500$, (b) $\times 7500$.

The compression modulus data of the composites are presented in Table IV. From these data, it is concluded that the compression modulus of all the composites increased as granite powder content increases. Addition of rigid mineral filler increases the modulus of the composites. Svehlova and Poloucek¹⁵ have shown that a better filler dispersion leads to a greater modulus. This development was explained by the percolation theory described by He and Ziang.¹⁶ According to this theory, there is a matrix zone around each particle affected by a stress concentration. If the distance between these particles is small enough, these zones join together and form a percolation network, which increases the modulus. For constant filler loading, if the particles are fine and well dispersed, the total volume affected is bigger and the distance between the particles is shorter. The percolation network therefore develops more easily, and the modulus increases.

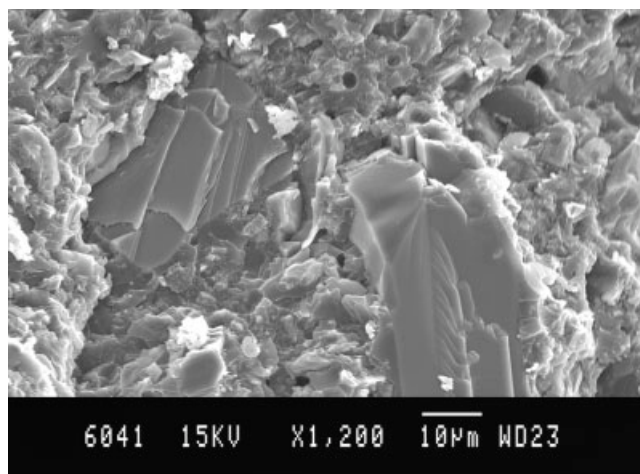


Figure 3 Scanning electron micrograph of impact-fractured sample of untreated granite powder-filled epoxy composite corresponding to 50% filler loading.

In the case of 4% ABS-toughened epoxy/coupling agent-treated granite powder composites, modulus increases linearly on increasing the filler content, showing higher modulus compared to other two types of composites.

The percentage weight gain (+) or weight loss (–) data, when the composites of untreated, coupling agent-treated, and 4 wt % ABS-toughened coupling agent-treated granite powder composites immersed in different chemicals are presented in Tables V and VI. From Table V, it is clearly evident that weight gain is observed in both the cases of untreated and coupling agent-treated granite powder-filled composites. In the case of coupling agent-treated granite powder-filled composites, less weight gain is observed when compared to untreated granite powder-filled composites. This result indicates that the coupling agent added

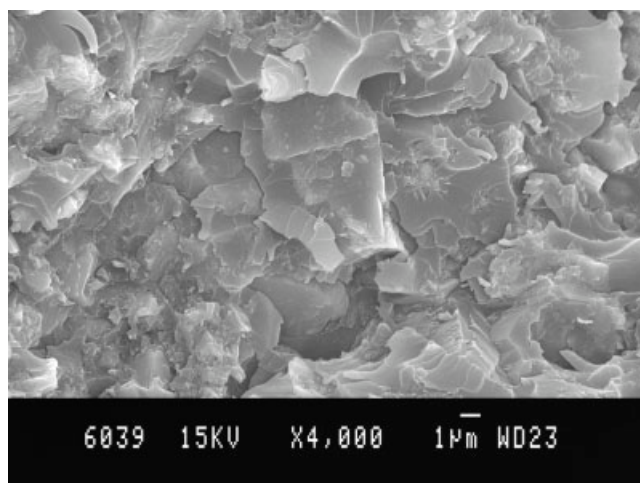


Figure 4 Scanning electron micrograph of impact-fractured sample of coupling agent-treated granite powder-filled epoxy composite corresponding to 50% filler loading.

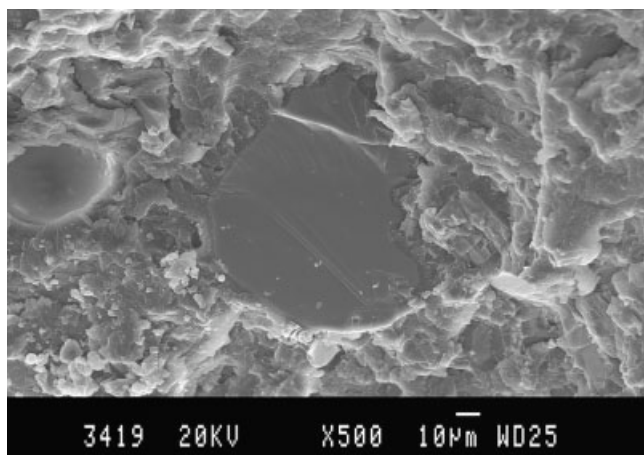


Figure 5 Scanning electron micrograph of impact-fractured sample of 4% ABS-toughened epoxy/coupling agent-treated granite powder composite corresponding to 50% filler loading.

brings the components closer, as a result a close packing is ensured in the composites. Generally, close packing enhances the chemical resistance of the materials. From Table VI, it is clearly evident that, when 4 wt % ABS-toughened epoxy/granite powder composite material is immersed in some strong acids, alkalis, and solvents, weight gain is observed. The weight gain observed in the entire composites is understandable, as crosslinked systems form three-dimensional networks that are chemically more stable.

The SEM micrographs of granite powder particles are shown in Figure 1(a,b). The particle size varies from 1 to 100 μm . SEM micrographs of granite powder revealed that granite powder exists as aggregates/agglomerates. The particles are angular in nature. Figure 2(a,b) illustrates the fractured surface of the 4% ABS/epoxy blend. The ABS particles are dispersed uniformly in the epoxy-rich matrix. The dispersed ABS particles in epoxy matrix result in two-phase morphology. One of the most striking features of this morphology is the presence of small pits, indicating the slip out of the particle during the fracture. Plastic deformation is also observed from the micrographs of ABS/epoxy blend.

Figure 3 shows the SEM micrographs of fractured surface of untreated granite powder epoxy with 50% filler content. It was observed that the fractured surface has a homogeneous structure. The presence of dark voids indicates the filler pull out in the matrix. SEM micrograph corresponding to 50% filler content of coupling agent-treated granite powder-filled epoxy composite is shown in Figure 4. These micrographs revealed the denser structure of the composite with

the presence of granite powder firmly attached to the matrix. This is an indication of good interfacial bonding established between the granite powder and epoxy. The SEM micrographs of 4 wt % ABS-toughened epoxy with coupling agent-treated granite powder-filled composite corresponding to 50 wt % filler content is given in Figure 5. It is observed that a good interfacial bonding between the matrix and the granite powder is established with a homogeneous and denser structure. The granite powder is distributed evenly in the toughened matrix system.

CONCLUSIONS

The flexural strength, compression strength, and compression modulus of the 4% ABS-toughened epoxy/coupling agent-treated granite powder composites are found to be superior over untreated and coupling agent-treated granite powder-filled composites, but the flexural modulus of 4% ABS-toughened epoxy/coupling agent-treated granite powder-filled composites was found to be lower. All the composites were found to have good chemical resistance except in the case of nitric acid. All the types of composites studied show poor resistance to nitric acid. Treatment of granite powder with coupling agent enhances the interfacial bonding leading to improved mechanical properties of the composites.

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