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ORIGINAL PAPER

The effect of hydrophobic coating applied to mica reinforced epoxy matrix composites on the water absorption properties of the composite

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Abstract

In this study, epoxy matrix composites containing mica with different particle sizes (40 and 80 μm) and quantities (0, 10, 20, 40 wt.%) were produced by the casting method. Composites were coated with a commercial agent imparting hydrophobicity in order to decrease water absorption. The bulk density and % open porosity values of the finished composites were determined by Archimedes' principle, and detailed microstructural analysis was carried out with scanning electron microscopy (SEM). The surfaces were characterized by the contact angle and roughness measurements. The durability and protective performance of the coating were quantified by the water absorption tests for various immersion times and temperatures (ASTM D570-98) and salt water resistance tests. Microstructure analysis showed that for all filler ratios and particle sizes, mica particles were homogeneously dispersed in the epoxy matrix and the mica–polymer interface was well bonded. It has been observed that the % theoretical density values of the composite products vary between 91.1 and 98.6%, and the total porosity varies between 1.4 and 8.87%. As the amount of mica increased, the total porosity also increased, regardless of the size of the mica particles. The neat epoxy matrix absorbed a higher quantity of water than composites in almost all cases, and the water absorption of the composites decreased with the increasing mica quantities and the decreasing particle size. The hydrophobic coating definitely caused the water absorption rates of the composite samples to decrease and the salt water resistance to increase, rendering them a convenient moisture- and corrosion-resistant composite material.

Keywords Mica · Epoxy matrix composites · Hydrophobic coating · Water absorption · Salt water resistance test

Extended author information available on the last page of the article

Introduction

Mica is a cheap, layered silicate material found in nature and easily obtained. Because of its unique electrical insulation, chemical resistance, thermal stability, and non-toxic properties, it is of great interest as a reinforcing element in polymer matrix composites [1–5]. The addition of mica to the polymer matrix is known to improve chemical resistance, fracture resistance and dimensional stability, as well as reduce gas emissions and provide sound insulation for the composites [6–8]. Because mica is a dielectric, it has very low electrical conductivity. Mica sheets can thus be utilized as electrical insulators in electronic equipment [9, 10]. Therefore, a specific use of mica-reinforced epoxy matrix composites is electrical insulation in electric generators. Mica-reinforced epoxy matrix composites *inherently possess some degree of water absorption* that must be minimized since such applications necessitate a low water absorption of the insulator. The dielectric properties of the composite are adversely affected by both the epoxy matrix and the mica filler's water absorption characteristics [11]. Some methods have been reported in the literature to prevent or diminish water absorption, such as the silanization of the reinforcing phase, gelcoat application, and coating the composite surface with a hydrophobic layer [12–16].

It has been reported in the literature that a protective hydrophobic coating is effective in preventing moisture ingress into carbon fiber-reinforced polymer matrix composites [12]. In this study, the effects of hydrophobic coating on epoxy matrix mica-reinforced composites against deterioration caused by weather conditions for long periods of time were determined and the coating's performance was evaluated.

For this purpose, epoxy matrix composites reinforced with different amounts and sizes of mica particles were produced, and the composites were coated with a commercial hydrophobic solution *to reduce the amount of moisture absorption.* The influence of the hydrophobic coating on the properties and durability of against water and NaCl solutions was investigated. The change in the physical properties with the mica reinforcement amount and particle sizes and its influence on the moisture absorption amount were determined.

Materials and method

Materials

Epoxy resin and hardener were obtained from Smooth-On, Canada. The epoxy resin and hardener were mixed in 73 wt.% and 27 wt.%, respectively. A naturally occurring mica (Işık Madencilik, Eskişehir, Türkiye) in two different particle sizes (40 and 80 μm) was used as a filler.

Characterization of mica

The size and morphology of the mica particles were determined with the FEI Quanta 650 Field Emission model scanning electron microscope (FEG-SEM). Phase

analysis of mica particles was carried out using a Rigaku Smartlab model X-ray diffractometer (XRD) device with a scan speed 2°/min between 5 and 55 degrees. The actual densities of the mica powders were measured with the help of the liquid pycnometer.

Production and characterization of composites

Composites with four different epoxy/mica powder ratios and two different filler particle sizes (40 and 80 μm) were produced by the casting method. A monolithic epoxy casting was used for comparison. The neat epoxy sample production was carried out as follows: The epoxy and hardener were mixed for 5 min using a mechanical mixer at 500 rpm. For composite production, the filling materials mica and epoxy resin were mixed in the specified proportions (Table 1). After mixing, the composite mixture was stirred in three stages at 500, 1000, and 1500 rpm for 5 min each. The mixture was vacuumed for 10 min to remove any remaining air bubbles. The mixture was poured into a mold for shaping. The coding of composite samples was done according to particle size and the amount of mica used. For example, the sample coded E8M20 contains mica particles of 80 μm in size and contains 20 wt.% mica reinforcement phase.

Coating of composite products with hydrophobic solution

The surfaces of the composite products were coated in a water-based hydrophobic solution with the dip coating method. The samples were prepared in 30*30 mm size. After the water-based polymer solution was applied to the composite material surface, the composite samples were heat-treated in an oven at 120 °C for 30 min.

Table 1 Materials and quantities used in the study

Sample Code	Epoxy% wt	Hardener% wt	Mica (40 μm)% wt	Mica (80 μm)% wt
Neat Epoxy	73	27	0	0
E4M10	73	27	10	0
E4M20	73	27	20	0
E4M40	73	27	40	0
E8M10	73	27	0	10
E8M20	73	27	0	20
E8M40	73	27	0	40

Tests and analysis

Microstructural analysis with scanning electron microscopy

Microstructural analysis was performed with the FEI Quanta 650 Field Emission model scanning electron microscope (FEG-SEM) on the sputter Au-coated samples to determine if the added mica reinforcement phase was homogeneously dispersed in the epoxy matrix, to observe the porosity, and to investigate the matrix–reinforcement interface properties.

Density and porosity determination

The bulk density of the composite samples was determined according to Archimedes' principle of water displacement. Bulk density and % open porosity calculations are given by Eq. 2.1 and Eq. 2.2, respectively; where W1 is the dry weight, W2 is the suspended weight, and W3 is the displaced liquid weight.

$$\text{Bulk Density} = \frac{W_1}{W_3 - W_2} x\rho_{su} \tag{2.1}$$

$$\%\ \text{Open Porosity} = \frac{W_3 - W_1}{W_3 - W_2} \times 100 \tag{2.2}$$

The theoretical density (Eq. 2.3) of the composites was determined as the linear combination of the epoxy and the filler phases [17]. The non-porous bulk densities of the mica powders used were measured with a liquid pycnometer. The density of the epoxy was obtained from the specifications of the commercial product. % Theoretical density (Eq. 2.4) and % total porosity amount (Eq. 2.5) were calculated using bulk density and theoretical density values.

$$\text{Theoretical density} = (V_m x \rho_m) + (V_t x \rho_t) \tag{2.3}$$

$$\%\ \text{Theoretical density} = (\text{Bulk density} / \text{Theoretical density}) \times 100 \tag{2.4}$$

$$\%\ \text{Total porosity amount} = (1 - \%\ \text{Theoretical density}) \times 100 \tag{2.5}$$

Surface roughness measurements

The arithmetic mean surface roughness (Ra) was determined with the Mitutoyo Surftest SJ-410 model surface profilometer. The arithmetic mean was taken as the average of five measurements, and the standard deviation was calculated.

Characterization of coated surfaces

The hydrophobicity of the surfaces of the coated and uncoated composite samples was characterized by water wettability (contact angle measurement). The contact angles of the obtained surfaces were measured by Kruss DSA-25 model contact angle measuring device. The surface energy was determined using deionized water and diiodomethane as test liquids. The Owens, Wendt, Rabel and Kaelble method (OWRK) was used to obtain the surface energies [18, 19].

Evaluation of coating performance

Determination of water absorption

The water absorption of coated and uncoated composite samples was determined according to the ASTM D570-98 standard. A Radwag analytical balance with 0.0001 g sensitivity was used for the test. Eight samples of each composition were used for testing. Before the test, the samples were kept at 105 °C for 24 h and cooled down in a desiccator. Water absorption tests were performed according to four different procedures (ASTM D570-98 7.1, 7.2, 7.4, 7.5), namely 2 h, 24 h, and one-week water immersion (at room temperature, ~ 23 °C) tests, and the boiling water test [20]. The amount of water absorbed was calculated using Eq. 2.6 according to ASTM D570-98.

$$\%\ \text{Water absorption amount} = (\text{Wet weight} - \text{Initial weight})/\text{Initial weight} \times 100 \tag{2.6}$$

Salt water resistance test

For the durability tests of the coating, the coated products were kept in 5% NaCl solution for 120 h. The weight increase of the samples was determined, and the contact angle measurements were carried out.

Results and discussion

Characterization of mica particles

In general, mica is classified as a layered silicate mineral with a flake geometry. The sheet/plate morphology and the average particle sizes of 40 µm and 80 µm of the mica powder samples were confirmed with the SEM-SE images, in conformity with the manufacturer specifications (Fig. 1).

The actual density of mica powders as measured by a liquid pycnometer as 2.77 g/cm^3. The XRD patterns of both mica samples showed a main phase of muscovite together with small amounts of quartz and calcite minerals (Fig. 2).

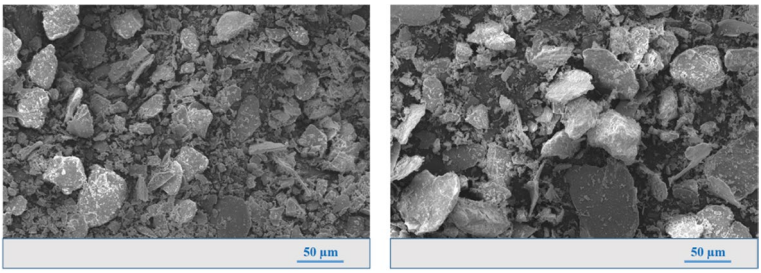


Fig. 1 Representative SEM-SE images of mica particles with different particle sizes **a** 40 µm, **b** 80 µm (1000x)

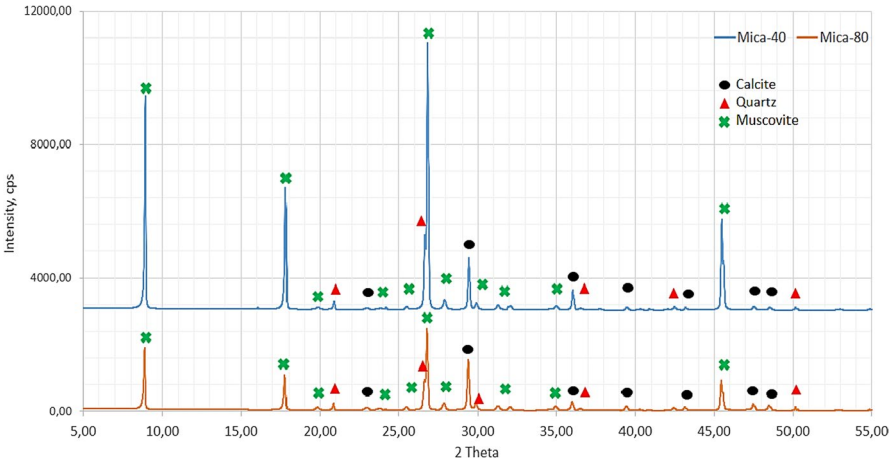


Fig. 2 XRD spectra of mica powders 40 µm and 80 µm

Density and porosity of composite products

Bulk density, % open porosity, theoretical density, % theoretical density, and % total porosity of the neat epoxy and composites with different amounts of mica particles are given in Table 2. The bulk density of neat epoxy was measured to be 1.1241 g/cm^3. An increase in the bulk density was observed with the increasing mica content of the composite samples. While the theoretical density value of the pure epoxy sample was 98.61%, slight decreases were observed in the theoretical density values of the composites. The theoretical density values of the composites were between 91.13 and 98.60%. While the amount of open pores was higher in pure epoxy samples, a decrease was observed with the addition of mica. The reactions that take place during the curing of the epoxy cause outgassing and the formation of open pores. Since the increase in the amount of mica will cause a decrease in the amount

Table 2 Bulk density, % open porosity, theoretical density, % theoretical density and % total porosity of the neat epoxy and composites with different amounts of mica particles

Sample	Bulk density (g/cm^3)	% Open porosity	Theoretical density(g/cm^3)	% Theoretical density	% Total porosity
Neat Epoxy	1.1241 ± 0.071	1.3779 ± 0.194	1.1400	98.61	1.39
E4M10	1.1938 ± 0.001	1.2010 ± 0.138	1.2108	98.60	1.40
E4M20	1.2526 ± 0.003	1.1963 ± 0.056	1.2916	96.98	3.02
E4M40	1.3584 ± 0.003	1.1761 ± 0.094	1.4905	91.13	8.87
E8M10	1.1882 ± 0.005	1.4721 ± 0.040	1.2108	98.13	1.87
E8M20	1.2531 ± 0.006	1.4077 ± 0.267	1.2916	97.02	2.98
E8M40	1.3592 ± 0.010	1.2225 ± 0.069	1.4905	91.19	8.81

of epoxy and less gas output, the amount of open pores decreased with the addition of mica and the increase in the amount of mica.

The effect of mica particle size on bulk density and other physical properties was negligible. The % open porosity amount of the pure epoxy was measured as 1.3779 g/cm3. However, the open porosity contents of composite samples varied between 1.18 and 1.41%. In our other studies, when the particle size of the reinforcement phase is in the range of 40–90 microns, it has been observed that their processability in the epoxy matrix is similar [21]. Since the mica particle sizes are 40 and 80 microns in this study, such a change in particle size did not affect the composite production process in a positive or negative way, and similar physical properties were obtained at the same filling ratios.

As the amount of mica in the composite increased, an increase in the amount of total porosity of the composite was observed. The total porosity of the pure epoxy sample was 1.39%. It increased with the increasing quantity of the mica particles, regardless of the mica particle size, and was in the range of 1.39% to 8.87%. Since mica, which is a layered silicate mineral among mineral fillers, has high water absorption, the total pore amount increased as the amount of mica increased. In other words, the amount of closed pores increased with the increase in mica quantity. On the other hand, in the literature, there was no study on the bulk density and porosity change of epoxy matrix mica-reinforced composites for comparison.

Microstructural analysis

In the SEM images, dark gray areas indicate epoxy matrix, and light gray structures show mica particles. It was observed that the mica particles were homogeneously dispersed in the epoxy matrix for all of the filling ratios and particle sizes (Fig. 3). Mica particles in E8-coded samples are coarser in size than those in E4-coded samples. The open porosity content is low in all the composites, and the microstructural images confirm the open porosity measurements based on the Archimedes principle.

When the matrix–reinforcement particle interface properties are evaluated, it can be said that the mica-polymer phase interface is well bonded in general (Fig. 4). The

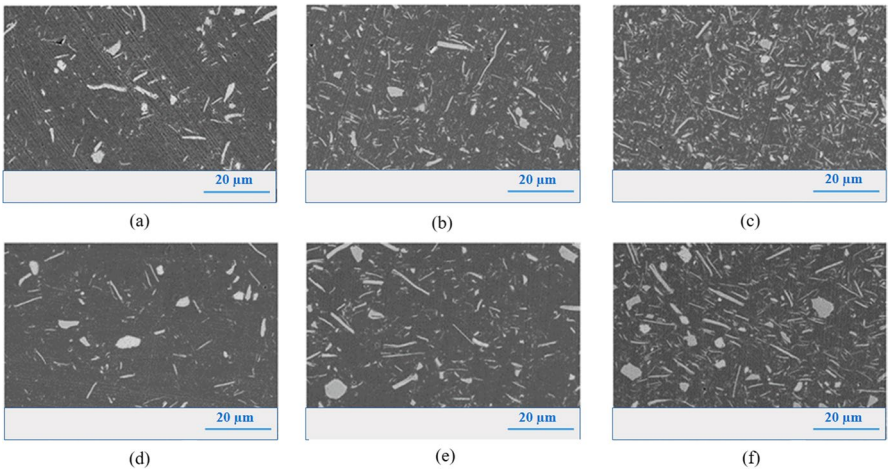


Fig. 3 Representative microstructural images of mica-reinforced epoxy matrix composites **a** E4M10, **b** E4M20, **c** E4M40, **d** E8M10, **e** E8M20, **f** E8M40, (500x)

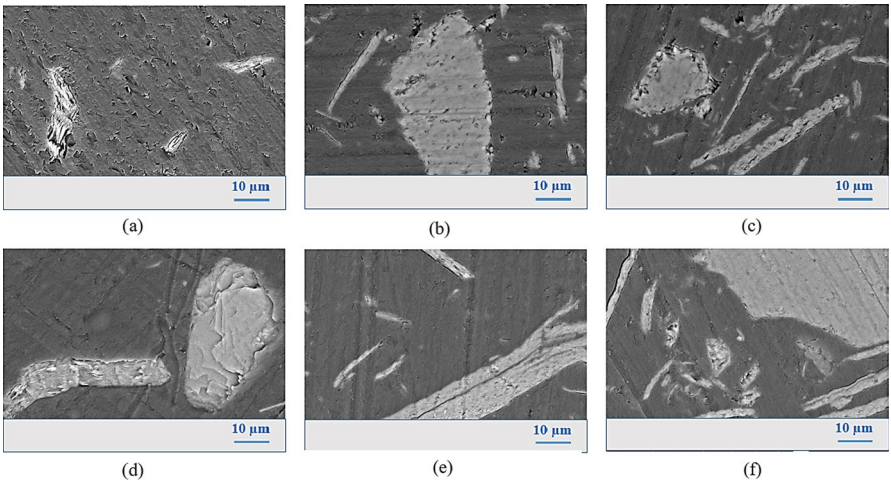


Fig. 4 Representative matrix-interface images of mica-reinforced epoxy matrix composites **a** E4M10, **b** E4M20, **c** E4M40, **d** E8M10, **e** E8M20, **f** E8M40, (5000x)

vacuum process applied during the production of the composites contributed significantly to this end. It has been observed that the matrix–reinforcement interface is more compatible in composites with finer mica particle size (40 µm). Microstructural faults were observed at the particle–matrix interfaces in the composites with 80 µm mica (Fig. 4).

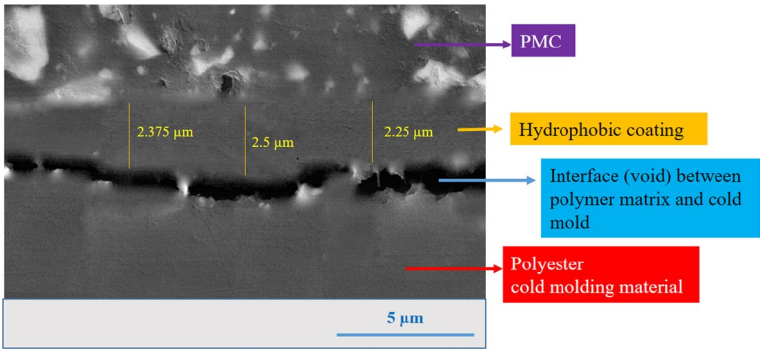


Fig. 5 The cross-sectional SEM-BSE image of E4M40 sample

Characterization of composites after coating

The wettability (contact angle) of the surfaces of the coated and uncoated composite samples was determined. The thickness of the coatings was calculated by the weight difference. The amount of coating material applied to the surface varies between 0.0012 ± 0.0003 g/cm^2 for the single coating and 0.0022 ± 0.0005 g/cm^2 for the double coating. The thicknesses of the coatings were calculated to be 1.3 ± 0.3 µm for the single coatings and 2.5 ± 0.6 µm for the double coatings since the dried polymer coating density is 0.99 g/cm^3 [19]. In addition, a SEM-BSE image was taken from the cross-sectional area of the composite to determine the coating thickness (Fig. 5). Hydrophobic coating thickness was measured at about 2.5 µm for the double coatings. Coating thickness calculations made by weight differences and SEM images support each other. As shown in Fig. 5, the formation of the gap between the hydrophobic coating and the hydrophilic polyester substrate is a generally observed phenomenon due to their inherent repulsion.

Contact angle

Changes in contact angle values before and after hydrophobic coating of mica-reinforced epoxy matrix composites containing different particle sizes and different quantities of mica are given in Table 3. The contact angle of the neat epoxy sample before coating was measured at ~ 59.47°. The contact angle increased with mica reinforcement, and it was observed that it ranged from 64.65° to 74.60°. Mica reinforcement led to an increase in surface roughness as well as an increase in the contact angle (Fig. 6).

The quantity of mica particles used for reinforcement had almost no effect on the contact angle for both 40 µm and 80 µm sizes. The composites reinforced by 80 µm mica particles yielded ~ 5° higher contact angle in comparison with the 40 µm case. The contact angle values of mica-reinforced epoxy matrix composites containing different particle sizes and different mica amounts after double coating were close to each other and varied between 101.65° and 106.40°.

Table 3 Change in contact angle values before and after coating of mica-reinforced epoxy matrix composites containing different particle sizes and different amounts of mica

Sample	Before coating	Coated surface	Sample	Before coating	Coated surface
Neat Epoxy	59.47 ±2.67	121.70 ± 0.14			
E4M10	65.50 ± 2.33	101.65 ± 2.85	E8M10	69.75± 3.89	104.60 ± 1.91
E4M20	64.65± 1.91	104.90 ± 2.89	E8M20	74.60± 2.69	103.45 ± 1.17
E4M40	64.85± 7.85	106.45 ± 3.34	E8M40	67.40± 6.79	106.40± 3.48

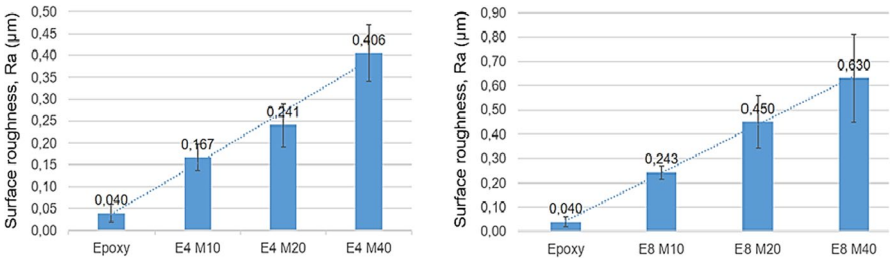


Fig. 6 Surface roughness values of neat epoxy and mica-reinforced epoxy matrix composites with varying quantities and particle sizes of mica

Effect of coating on bulk density and porosity

Bulk density and % open porosity of mica-reinforced epoxy matrix composites containing different particle sizes and different mica amounts were investigated before and after coating (Table 4). As expected for the latter, the bulk density values of all the samples increased, while % open porosity decreased.

Water immersion of composites

The water absorptions of coated and uncoated composite samples were determined according to the ASTM D570-98 standard (Table 5). Water absorption tests were performed according to five different test procedures: at room temperature for 2 h, at boiling water for 2 h, at room temperature for 24 h, at room temperature for 1 week and at room temperature for 2 weeks. In general, the following factors

Table 4 Bulk density and % open pore amounts of mica-reinforced epoxy matrix composites containing different

Sample	Before coating			After coating		
	Bulk density(g/cm^3)	% Open porosity	% Total porosity	Bulk Density (g/cm^3)	% Open porosity	% Total porosity
Neat Epoxy	1.1241 ± 0.071	1.3779 ± 0.194	1.39	1.1244 ± 0.001	1.3389 ± 0.176	1.37
E4M10	1.1938 ± 0.001	1.2610 ± 0.138	1.40	1.1944 ± 0.002	1.1997 ± 0.241	1.35
E4M20	1.2526 ± 0.003	1.1963 ± 0.056	3.02	1.2538 ± 0.003	1.1830 ± 0.105	2.93
E4M40	1.3584 ± 0.003	1.1761 ± 0.094	8.87	1.3586 ± 0.014	1.1685 ± 0.099	8.85
E8M10	1.1882 ± 0.005	1.4721 ± 0.040	1.87	1.1921 ± 0.002	1.4387 ± 0.152	1.54
E8M20	1.2531 ± 0.006	1.4077 ± 0.267	2.98	1.2594 ± 0.004	1.3814 ± 0.136	2.49
E8M40	1.3592 ± 0.010	1.2225 ± 0.069	8.81	1.3596 ± 0.004	1.1317 ± 0.302	8.78

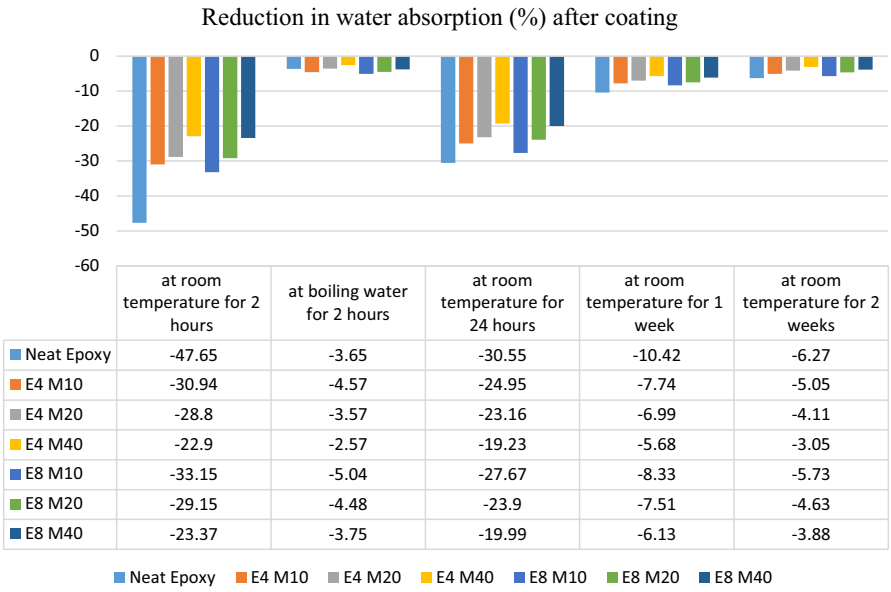
Table 5 Water absorption amounts of mica-reinforced epoxy matrix composites containing different particle sizes and different mica amounts before and after coating

Test conditions	% Weight Gain (Water absorption amounts)				
Sample	At room temperature for 2 h	At boiling water for 2 h	At room temperature for 24 h	At room temperature for 1 week	At room temperature for 2 weeks
Neat Epoxy	0.56 ± 0.14	1.37 ± 0.15	0.60 ± 0.12	0.97 ± 0.07	1.37 ± 0.09
Coated neat epoxy	0.38 ± 0.22	1.32 ± 0.19	0.46 ± 0.09	0.92 ± 0.05	1.25 ± 0.08
E4M10	0.40 ± 0.12	1.25 ± 0.11	0.45 ± 0.16	0.92 ± 0.09	1.25 ± 0.11
Coated-E4M10	0.31 ± 0.12	1.20 ± 0.18	0.36 ± 0.09	0.86 ± 0.13	1.19 ± 0.23
E4M20	0.38 ± 0.11	1.19 ± 0.09	0.42 ± 0.04	0.88 ± 0.23	1.18 ± 0.05
Coated-E4M20	0.29 ± 0.10	1.15 ± 0.05	0.34 ± 0.04	0.83 ± 0.06	1.13 ± 0.09
E4M40	0.31 ± 0.06	1.17 ± 0.06	0.36 ± 0.07	0.74 ± 0.01	1.14 ± 0.10
Coated-E4M40	0.26 ± 0.07	1.14 ± 0.07	0.30 ± 0.11	0.70 ± 0.06	1.11 ± 0.03
E8M10	0.43 ± 0.07	1.47 ± 0.03	0.48 ± 0.15	0.95 ± 0.04	1.46 ± 0.11
Coated-E8M10	0.33 ± 0.13	1.40 ± 0.12	0.38 ± 0.11	0.88 ± 0.06	1.38 ± 0.13
E8M20	0.39 ± 0.12	1.38 ± 0.21	0.43 ± 0.09	0.88 ± 0.12	1.37 ± 0.06
Coated-E8M20	0.30 ± 0.10	1.32 ± 0.11	0.35 ± 0.07	0.81 ± 0.03	1.31 ± 0.07
E8M40	0.35 ± 0.13	1.17 ± 0.05	0.38 ± 0.14	0.77 ± 0.04	1.16 ± 0.07
Coated-E8M40	0.28 ± 0.08	1.13 ± 0.22	0.32 ± 0.10	0.72 ± 0.01	1.12 ± 0.07

influence water absorption: (i) the hydrophilic nature of the matrix and the mica; (ii) the adhesion between the mica and the epoxy matrix; and (iii) the occurrence of voids in the composites. The maximum water absorption was obtained in the pure epoxy matrix as compared to the mica-added composites; that is, the water absorption values decreased with the addition of mica to the epoxy matrix. Similar results were obtained by Liu et al. [23]. Mica absorbs less water than epoxy resin, which can be attributed to a tortuosity effect due to the lamellar nature of the silicate minerals. These lamellar structures require a curved path for water to enter the composites. As a result, composites have lower maximum water absorption values than neat epoxy matrix. In addition, as the quantity of mica increases, the water absorption decreases further due to the increasing tortuosity effect. The second influencing factor in water uptake is adhesion between the mica and the epoxy matrix. As seen from SEM images, the connectivity between the phases is good enough that this does not adversely affect water absorption. The last factor is voids or porosities in the composites. The data obtained from the Archimedes principle showed that the open porosity in composites is less than that of pure epoxy. Outgassing and hence the creation of open pores occur during the curing process of the epoxy. The increasing quantity of mica in the composites results in less gas production and pore reduced.

After coating, the water absorption value of mica-filled epoxy matrix composite samples decreased in all of the test conditions. As the particle size of mica, which was used as a filler, increased, an increase was observed in the water absorption of the composites. As the amount of mica increased for both particle sizes, a diminution in water absorption was observed. The pure epoxy sample showed the highest water absorption values in almost all test condition. The sample with the closest value of water absorption to the epoxy sample was the sample coded E8M10. The highest water absorption values were observed for the E8M10-coded sample after two weeks at room temperature and two hours in boiling water. A linear correlation was observed between the water absorption and % open porosity over the full testing range, indicating that the atmospheric pressure was sufficient for water to fill the pores of the produced samples totally and the negligibility of the capillary forces inside the porous structure. After coating, the water absorption of pure epoxy decreased under all conditions. The water absorption amount of the uncoated epoxy sample changed from 0.56 to 1.37%, a variation of 2.4 times from room temperature to boiling temperature of water for the same duration of 2 h. The same highest water absorption values were observed after 2 h' immersion in boiling water and 2 weeks of soaking in water at room temperature. The water absorption of the samples increased with soaking time and water temperature.

The reduction in the water absorption values after the hydrophobic coating is given in Fig. 7. The reduction amounts were higher for shorter durations of water immersion, with the boiling water immersion test yielding the lowest reduction values, even lower than the 2 weeks' room temperature immersion. The results indicated that the hydrophobic coating layer provided resistance to the water absorption and decreased the water absorption rate, reducing the long-period water absorption value to about 5% and most probably without changing the equilibrium water absorption. Nevertheless, the reduced rate of water absorption of the composites



	at room temperature for 2 hours	at boiling water for 2 hours	at room temperature for 24 hours	at room temperature for 1 week	at room temperature for 2 weeks
Neat Epoxy	-47.65	-3.65	-30.55	-10.42	-6.27
E4 M10	-30.94	-4.57	-24.95	-7.74	-5.05
E4 M20	-28.8	-3.57	-23.16	-6.99	-4.11
E4 M40	-22.9	-2.57	-19.23	-5.68	-3.05
E8 M10	-33.15	-5.04	-27.67	-8.33	-5.73
E8 M20	-29.15	-4.48	-23.9	-7.51	-4.63
E8 M40	-23.37	-3.75	-19.99	-6.13	-3.88

Fig. 7 Reduction in water absorption after coating of mica-reinforced epoxy matrix composites

with the hydrophobic coating can be expected to provide an overall lower absorbed water content operational condition for the material.

The water absorption amount of the uncoated E4M10 sample varied between 0.40 and 1.25, and the water absorption decreased in all test conditions after hydrophobic coating. While the water absorption was 0.40% after 2 h of immersion at room temperature, it increased 3.125 times and reached 1.25% when kept in boiling water for the same duration (2 h). Similar water absorption rates were obtained after 2 h of immersion in boiling water and 2 weeks of soaking in water at room temperature (1.25%). The water absorption of the E4M10 sample after coating decreased by 31% after 2 hours of soaking at room temperature, 5% after 2 hours of soaking in boiling water, 25% after 24 hours of soaking in water at room temperature, 8% after soaking in water at room temperature for 1 week (Fig. 7).

In the immersion tests at room temperature, the coating performance decreased by ~ 5% with the increase in the immersion duration from 2 to 336 h. The amount of water absorption after coating decreased in all test conditions of the sample coded as E4M20. The water absorption of the uncoated E4M20 sample varied between 0.38% and 1.19%. While the water absorption amount was 0.38% after 2 h at room temperature, it increased 3.13 times and reached 1.19% when kept in boiling water for the same duration (2 h). Similar water absorption values were obtained after soaking for 2 h in boiling water and for 2 weeks in water at room temperature (1.19 and 1.18). While the amount of water absorption of the E4M20 sample after coating decreased by 29% after 2 h of soaking at room temperature, 4% after 2 h of soaking in boiling water, 23% after 24 h of soaking in room temperature, and 7% after 1 week of soaking in water. The coating performance

Table 6 Weight changes of pure epoxy and mica filled epoxy matrix composites after standing in salt water and contact angles of coated and uncoated products with hydrophobic solution after salt water resistance test

Sample	Weight gain (%)	Contact angle before NaCl Test		Contact angle after NaCl Test	
		Uncoated	Coated	Uncoated	Coated
Epoxy	0.74 ± 0.13	59.47 ± 2.67	121.70 ± 0.14	74.75 ± 2.25	83.25 ± 1.42
E4 M10	0.65 ± 0.07	65.50 ± 2.33	101.65 ± 2.85	64.45 ± 1.15	87.50 ± 0.25
E4 M20	0.62 ± 0.07	64.65 ± 1.91	104.90 ± 2.80	63.15 ± 2.50	94.20 ± 0.20
E4 M40	0.56 ± 0.04	64.85 ± 7.85	106.45 ± 3.34	68.65 ± 3.35	90.60 ± 0.20
E8 M10	0.69 ± 0.09	69.75 ± 3.89	104.60 ± 1.91	72.40 ± 3.40	81.50 ± 0.35
E8 M20	0.64 ± 0.05	74.60 ± 2.69	103.45 ± 1.17	73.15 ± 1.10	86.80 ± 0.50
E8 M40	0.58 ± 0.16	67.40 ± 6.79	106.40 ± 3.48	64.50 ± 0.20	80.80 ± 0.20

decreased by 4% with the increase in time from 2 to 336 h in the soaking tests at room temperature (Fig. 7). As in the other samples, the amount of water absorption after coating decreased in all test conditions for the sample coded E4M40. The water absorption amount of the uncoated E4M40 sample varied between 0.31 and 1.17. While the water absorption amount was 0.31 after 2 h of holding at room temperature, it increased 4 times and reached 1.17 when kept in boiling water for the same time (2 h). Similar water absorption rates were obtained after 2 h of waiting in boiling water and 2 weeks of soaking in water at room temperature (1.17 and 1.14). The amount of water absorption of the coated E4M40 sample decreased by 23% after 2 h of soaking at room temperature, 3% after 2 h of soaking in boiling water, 19% after 24 h of soaking in water at room temperature, ~ 6% after 1 week of soaking in water at room temperature. The coating performance decreased by 3% with the increase of the time from 2 to 336 h in the soaking tests at room temperature. When the particle size of mica used as the filler was increased from 40 µm to 80 µm, an increase in the water absorption was observed due to the formation of more voids between the matrix–reinforcement phase interphase.

Salt water resistance test

The weight gain values of the composite products after being kept in a 5% NaCl salt solution for 120 h (5 days) are given in Table 6. The results showed that the resistance of pure epoxy against salt water is weaker than that of mica-filled epoxy matrix composites. The sample with the best salt water resistance was the composite sample coded E4M40, containing 40% mica reinforcement and containing mica particles in 40 µm size. The salt water resistance and the water absorption test results were in compliance.

In order to determine the performance of the coating, the contact angles of the products coated with hydrophobic solution and the uncoated products were measured after the salt water resistance test. Since the uncoated pure epoxy sample was damaged after the NaCl test, the roughness level on its surface increased, and an

Table 7 Surface energies of samples before and after NaCl Test

	Surface energies (mJ/m^2)	
	Before NaCl Test	After NaCl Test
Coated Neat Epoxy	27.69	39.54
Coated E4M40	27.73	36.94
Coated E8M40	28.74	38.84

increase in the contact angle was observed. In all uncoated and mica-reinforced samples, there was almost no change in contact angles after the NaCl test. A decrease in contact angles was observed after the NaCl test in all samples coated with a hydrophobic solution. However, the contact angles of the coated samples after soaking in salt water were higher than the uncoated ones. This indicated the resistance provided by the coating against salt water.

Surface energies of samples before and after NaCl test were calculated in order to evaluate the coating performance (Table 7). The surface energies of all the coated pure epoxy and mica-reinforced composites that were subjected to salt corrosion test increased since the coating lost its protective property.

Conclusions

In the study, mica-reinforced epoxy matrix composites were produced by the casting method, and microstructural analysis showed that the mica filler was homogeneously dispersed in the epoxy matrix for all the filling ratios and particle sizes. The porosity of all the composites was low, and the porosity measurements based on the Archimedes principle were confirmed by the SEM images. The vacuum process applied during the production of the composites contributed significantly to the matrix–reinforcement interphase properties. In composites with finer mica particles (40 μm), the matrix–reinforcement interphase was more compatible, and when 80 μm size mica was used, faults were observed at the interphases.

It was observed that the neat epoxy matrix absorbed a higher quantity of water than the composites in almost all cases. The water absorption of the composites was reduced as the amount of mica increased and the particle size decreased. The water absorption and salt water resistance tests, as well as the contact angle measurements, all indicated that the hydrophobic coating of the composite material, which inherently carries a water absorbent characteristic due to its open porosity, even at low, imparted resistance to water absorption by reducing the time rate of water soaking inside the material. The hydrophobic coating provided, besides the decreased water absorption rate, a reduced long-period water absorption value, although equilibrium water absorption might not have changed. The reduced rate of water absorption of the composites with the hydrophobic coating can be expected to yield an overall lower absorbed water content operational condition for the material. Nevertheless, since absorbed water is expected to adversely affect the service performance and life of the material, the hydrophobic

coating of the composite constitutes a major development, for example, in the electrical insulating materials in electric generators, where low moisture absorption is desirable. The lowered rate of salt water absorption provides the additional advantage of diminishing corrosive exposure.

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Authors contribution Gokhan Acikbas was involved in methodology, validation, interpretation of data, visualization, and resources; Kaan Sezer contributed to methodology and validation; Selçuk Özcan was involved in interpretation of data and revised it critically for important intellectual content, writing—editing; and Nurcan Calis Acikbas contributed to supervision, conceptualization, investigation, methodology, writing—original draft and approved the version to be published.

Declarations

Conflicts of interest There are no conflicts to declare.

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