

Thermal and Mechanical Properties of Epoxy Composites Reinforced by a Natural Hydrophobic Sand

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ABSTRACT: If a low weight percentage of crude fine fillers can improve properties of polymer materials directly without complicated chemical treatment process involved, it will be significant for many industrial applications. Our previous study indicated that a kind of Cancun natural sand could be an effective filler material for polymer composites. In this current work, the epoxy composites reinforced by this kind of natural sand particles were prepared and thermal and mechanical properties of the composites containing up to 5 wt % of the sand particles were characterized. Results showed that the highest flexural strength appears in the epoxy composite containing 1 wt % sand particles. A damage model was used to interpret the flexural properties, which showed an acceptable agreement with the experimental results. The glass transition temper-

ature, high temperature storage modulus, and dimensional stability of the sand/epoxy composites monotonically increased with the addition of the sand particles. The sand particle/epoxy composites also displayed a noticeable enhancement in thermal conductivity. Theoretical analysis showed that in addition to conduction, other heat transport mechanisms played roles in the improved heat transmission through the composites. As a natural porous micron-scale material, Cancun sand has the potential for applications in cost-effective composites with enhanced mechanical and thermal properties. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 109: 247–255, 2008

Key words: sand particle; epoxy resin; composites; thermal property; flexural property

INTRODUCTION

Polymer materials modified with inorganic fillers have been researched and applied extensively. Many kinds of inorganic particles, including compounds and elementary substances, have been shown to improve the performance of polymers remarkably.^{1–4} However, the size and dispersal characteristics of inorganic particles have a great effect on the properties of polymer composites. Well-dispersed inorganic fillers in a polymer matrix and compatibility between inorganic and organic phases are important to achieve an overall effective performance enhancement.^{5–9} The syntheses of various nanoparticles have made the resulting nanocomposites applicable to many engineering applications, but the full realization of this potential will require great efforts to improve surface properties of nanofillers in order to

achieve eminent interfacial interactions and dispersive stability in the polymer matrix. Many long term studies are still in an embryonic stage.

Crude, fine inorganic filler materials have received increasing attention owing to their low cost, abundant availability, environmental benefits, and the potential for modification of functional characteristics.^{10–12} But most crude fillers are chemically inert to the polymer matrix, and cause a loss in mechanical properties in the majority of cases. In some instances, the required loading of natural filler is very high and limits the role in property enhancement for polymer materials.^{13–15} Some crude filler materials need severe and sophisticated chemical treatments, such as wood fiber and clay.^{16–19} Filler materials that can improve the properties of matrix materials directly without complicated sophisticated treatment processes are highly desired for composite material fabrication.

Our previous study indicated that a kind of beach sand from the area of Cancun, Mexico, whose main chemical composition was calcium carbonate, is a micron porous material with hydrophobic surface property.²⁰ A polymer composite made from mixing Cancun sand particles with poly (methyl methacrylate) showed a good interfacial adhesion as revealed

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from SEM studies, owing to the hydrophobic nature of the sand particle surface and the porous morphology.²⁰ These results prompted us to make further efforts and explore the application value of this porous natural sand as a particulate filler for polymer composite materials. In this present study, epoxy composites containing up to 5 wt % of sand particles were prepared. Characterization on the mechanical properties, thermal analyses, microstructure, and thermal conductivity of the sand composites were carried out. These studies revealed that natural Cancun sand may be a potentially eminent filler for polymer composite materials.

EXPERIMENTAL

Materials

The diglycidyl ether of bisphenol A (Shell EPON[®] 828) and the EPI-CURE Curing Agent W (Miller-Stephenson Chemical Company, US) were used to prepare the neat epoxy resin and the sand/epoxy composites. Cancun sand sample was collected from a beach area in Cancun, Mexico. The average diameter of the collected natural sand particles is about 5.5 μm , as revealed from SEM study. The sand sample was heated at 100°C under vacuum for 10 h to eliminate residual water. Then the sizes of the sand particles were reduced effectively using a ball milling method. Dry ball milling was undertaken in a purified argon protect atmosphere on a planetary ball mill. Steel balls with diameters of 4 and 6 mm were used as grinding medium. A ball to powder weight ratio of 10:1 was selected to ensure a high efficiency. Ball milling was conducted with the disk revolution speed of 180 rpm. To minimize the oxidation during milling, we took 1 h interval after each 2 h milling. After milling, samples were stored in argon and taken out after 24 h. As shown in our previous work, the average size of the sand particles had reached to about 1 μm after 24 h of milling.²¹ From the SEM images of the sand particles milled for 24 h [Fig. 1(a)], it can be seen that there is a wide size distribution of sand particles. In the following magnified image [Fig. 1(b)], it is found that the large particles are actually porous and brittle accumulations of many smaller nanoparticles. The milled sand was used without further treatment to prepare the sand/epoxy composites.

Preparation of the sand/epoxy composites

The neat epoxy resin was prepared by mixing the epoxy resin with the curing agent at room temperature for 2 h, degassing the mixture under vacuum conditions, and curing via a thermal schedule (120°C for 1 h and 160°C for an additional 4 h). For prepa-

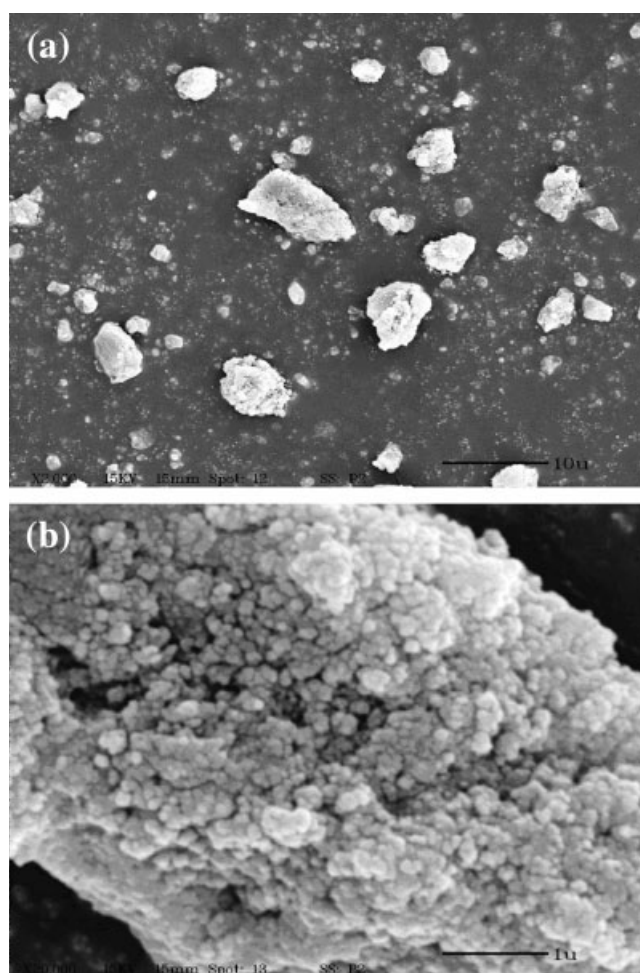


Figure 1 SEM images of the milled sands (a) low resolution; (b) high resolution.

ration of the sand/epoxy composites, sand particles of different loading after 24 h of milling were added into the epoxy resin and the mixture was stirred at 60°C for 3 h. Then the curing agent was added and further mixed with sand/epoxy at room temperature for 2 h. Subsequently, the sand/epoxy/curing agent mixture was degassed under vacuum condition, followed by the same thermal curing schedule as that of neat epoxy resins.

Characterizations of the materials

The size distribution of sand particles was measured by a PSS Nicomp Particle Sizing Systems 380. Differential scanning calorimetry (DSC) analysis was performed using a DSC Q1000 (TA Instruments) with a nitrogen atmosphere. The samples were heated from room temperature to 250°C at a rate of 10°C/min. The dynamic mechanical analysis (DMA) of the neat epoxy and the composites was carried out with a Rheometrics Scientific DMTA 3E Dynamic Mechanical Analyzer in the three-point bending mode at a

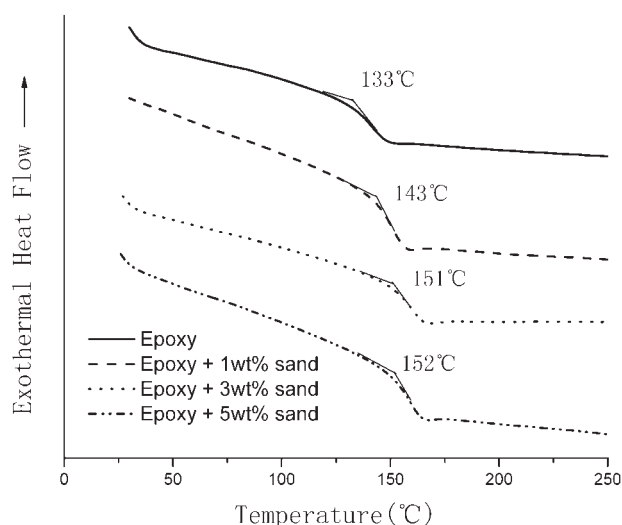


Figure 2 DSC curves of the neat epoxy resins and sand/epoxy composites.

frequency of 1 Hz. The specimens were heated from room temperature to 250°C at a heating rate of 10°C/min. The coefficients of thermal expansion (CTE) of neat epoxy and composites were obtained with TA Instruments 2940 Thermomechanical Analyzer (TMA) at a heating rate of 10°C/min under nitrogen purge. The three point bending tests of neat epoxy resin and sand/epoxy composites were performed using a Q-test machine (MTS) according to the ASTM-D790 standards. The specimens were from both pristine epoxy and epoxy/sand composites and their sizes of the specimens were 2.95 ± 0.03 mm (thickness) \times 12.52 ± 0.20 mm (width) \times 63.83 ± 0.05 mm (length). The speed of the cross-head was 1 mm/min and the results of the bending properties for five samples were averaged. The flexural stress, flexural modulus, and ultimate strains of the samples were calculated according to the equations in ASTM D790. The morphology of the sand particles and fracture interface of neat epoxy and composite samples was examined by a JEOL JSM-6300 model SEM. The size distribution of the sand particles was determined using a Santa Barbara Particle Sizing System at room temperature. Thermal conductivity performances of samples were tested using a Mathis TC-30 Thermal Conductivity Instrument under 25°C.

RESULTS AND DISCUSSION

DSC analyses

To evaluate the effect of sand additives on the material character of epoxy resin, the neat epoxy resin and the sand/epoxy composites were subjected to DSC analyses. The DSC curves are presented in

Figure 2. From the figure we can see that about 10–20°C temperature increases occurred in the glass transition temperature (T_g) of the epoxy matrix after 1–5 wt % of sand was added into the epoxy matrix. This suggested that the sand particles exerted a great influence on the mobility of epoxy molecules, and the T_g of the polymer composites varied accordingly. The effect of reinforcing fillers on T_g of epoxy resin has been investigated by many researchers.^{22–25} Some of them reported similar increases while others reported a decrease. However, increasing the sand particle content between 3 and 5 wt % did not increase the T_g significantly, the reason for which is likely the result of the increasing nonuniform dispersion of the sand particles when such amounts of particles were involved. Increased agglomeration of the sand particles could result in lower enhancement in rigidity of the epoxy crosslinking structure. The dispersion of the sand particles in the composite samples observed from the SEM images are shown later.

DMA tests

If a sinusoidal load is applied to a polymeric sample, it will deform sinusoidally, and the sinusoidal strain depends on elastic and viscous behavior of the sample. Depending on phase angle and amplitude, the elastic modulus (E') and the loss factor ($\tan \delta$) of the epoxy and the sand composites were characterized by DMA as functions of temperature and sand loading. In this work, we examined the effects of sand particles on the dynamic mechanical properties of epoxy resin.

Figure 3(a) shows increases in storage modulus of the epoxy matrix both below and above T_g due to the addition of the sand particles, the effect being more obvious above the T_g of the epoxy matrix. The increase in modulus of the sand composites can be attributed to the high stiffness of the sand particles. In particular, above T_g the epoxy is in a rubbery state, and thus, the high stiffness of the sand relative to the rubbery resin matrix provides larger enhancement in the storage modulus due to the greater restricted movement of epoxy segments.^{22,26,27} It is reasonable to think that the larger modulus ratio of the sand to epoxy resin at higher temperatures is the cause of this phenomenon.²⁷ Therefore, in the rubbery plateau region, the improvement in storage modulus is more significant compared to that in glassy region. However, for the epoxy sample containing 5 wt % sand particles, the storage modulus under room temperature is just slightly higher than that of the neat epoxy resin, and lower than that of the other composite samples such as those containing 1 and 3 wt % sand particles. The prime reason for this result is the increasingly nonuniform dispersion of the sand in the epoxy matrix at the higher

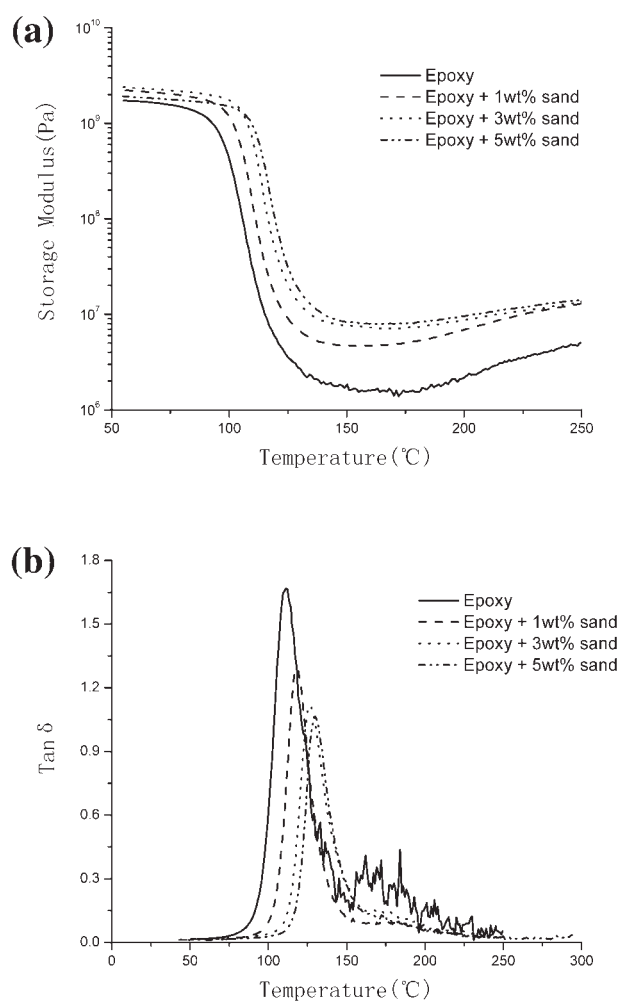


Figure 3 DMA of the epoxy and composites as a function of temperature (a) storage modulus and (b) loss factor.

loadings, which made the reinforcement function less effective than in the sand composites with lower loading of particles. Another reason may be the negative effect of such high loading particles on the crosslinking structure of the epoxy.

Figure 3(b) shows the temperature dependence of $\tan \delta$ of the neat epoxy and sand/epoxy composites. The position of $\tan \delta$ peak in $\tan \delta$ -temperature curve was used to identify the T_g of the material. It can be seen that the peaks of $\tan \delta$ of the composites shift to a higher temperature compared to that of the neat epoxy resin with increasing levels of sand content and correlate very well to DSC experimental results. On the molecular scale, the T_g is indicative of mobility on a macromolecular scale. High T_g means less mobilization of chain segments.²⁸ This reduction in mobility of the segments in the epoxy resin is caused by the addition of the sand particles. In the composites, within the chain segments there are inhibiting sand particles, and to activate the chain segments, both chain segments and internal

sand particles need to be mobilized, which in turn requires more energy: higher temperature corresponding to higher T_g providing more energy for chain segment mobilization. As a result the sand composites show enhanced dynamic mechanical properties, which means the sand particles provide more stable structures to the epoxy composites. Similar results were also observed in other composites.^{29,30} Chio et al.³¹ observed that unconstrained segments of polymer molecules retain T_g whereas segments close to filler particles are less mobile and cause higher T_g .

The height of $\tan \delta$ peak gradually diminishes with the increasing loading of the sand particles [Fig. 3(b)]. $\tan \delta$, which is the ratio of loss modulus and storage modulus, is also referred to as the damping coefficient. Loss modulus corresponds to dissipation of energy in the form of heat, etc. Therefore, the results indicated that the addition of sand particles reduces the damping factor of the epoxy composites. According to the damping theory of polymer materials, damping mainly is a result of friction among the mobile polymer chains/segments during temperature increases. This is why the damping peak always appears in the glass transition zone, since the friction is occurring among a greater number of polymer segments. For polymer composites, the overall damping will be the result of two factors: (a) the amount of the polymer matrix available, and (b) the interface area between the solid fillers and the polymer matrix.³² It should also be noted that the addition of fillers into a polymer results in a lower relative amount of the polymer. So, if the contribution to the damping from the interface source does not exceed the reduction in damping from the lower available polymer material in the composite, the overall damping will be decreased. In this study, the addition of the sand particles reduced the relative amount of the epoxy matrix available for damping, increasingly so with higher loading of particles. Therefore, it can be deduced from the results that the damping is mainly from the epoxy matrix, and the lowered damping of the sand composites as particle content increases is due to the decreased amount of the epoxy matrix, and that the interface contribution to the damping is a relatively less influential factor. In addition, there may be a reinforcing effect from the fact that the better interaction in the interface between the hydrophobic particles and the epoxy matrix may contribute to the retardation of the damping function.³² Regarding the effects of interface between various reinforcements and polymer matrices on the overall damping property, more research is needed as damping of composites is a function of many factors including crosslinking density, molecular weight, interface between particle and epoxy, as well as free volume density etc.^{33,34}

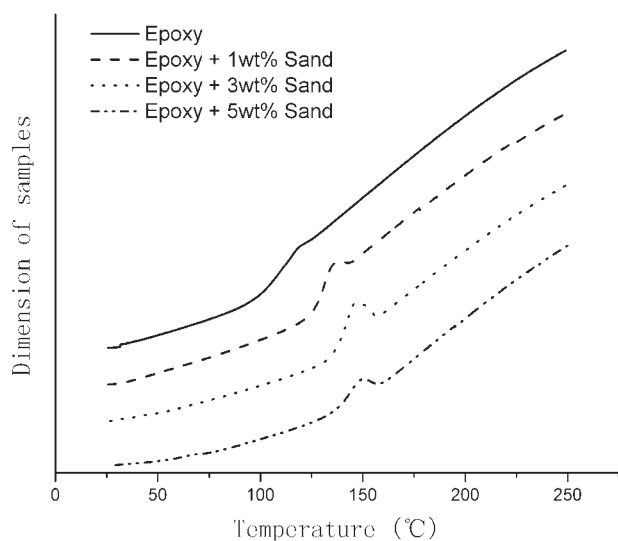


Figure 4 Dimension change versus temperature curves of the epoxy and composites.

TMA tests

TMA is a thermal analysis for samples used to measure changes in dimension taking consideration of adjustable sample strain with temperature and/or time variation. It can be used in determining CTE and T_g . Figure 4 shows the plots of the dimensions versus temperature curves for the neat epoxy and the sand/epoxy composites. It can be seen that with an increase of sand particle content in the epoxy resin, the dimensional stability of the samples were increased and the highest stability was found in the composite sample with 5 wt % sand particles. The change in dimension of the polymer materials is controlled by the macromolecular activity and interaction, and it is phase dependent. Therefore, the CTE values of the epoxy and the sand composites are different below and above the T_g . The CTE values below and above T_g for the neat epoxy and the sand/epoxy composites are listed in Table I. The inorganic sand additives have no obvious deformation and relaxation as observed in the epoxy molecules with the increase of temperature. If the sand particles are dispersed in the epoxy matrix effectively, these rigid additives will retard the thermal expansion of the epoxy molecules when the temperature ascends. Therefore, the addition of the sand particles contributes to the decreasing expansion of the epoxy composite materials, which leads to the thermally dimensional-stable structures. With increased sand loadings, the volume fraction of epoxy resin matrix decreases accordingly, and thus, it is reasonable to observe that the CTE values under both below and above T_g of the sand/epoxy composites decreased slightly with the addition of loading of sand particles.

Flexural properties

The flexural properties of neat epoxy and the sand particle-filled composites were summarized in Table II. The enhancements in flexural strength and modulus for the epoxy resin with 1 wt % sand loading were 7.5 wt % and 8.7%, respectively, compared with that of the neat epoxy. The hydrophobic rigid sand particles play a reinforcing effect on the epoxy matrix. It is important to mention that these enhancements came from natural sand particles without complicated chemical treatment involved. However, with further increases to the sand particle content, flexural strength and modulus of epoxy resin declined. This phenomenon could be explained by the effect of increased particle amount resulting in less effective dispersion of the sand particles with the increasing filler contents. Nielsen and Landel³⁵ reported that increases of particle size (up to micron level), might increase the stiffness while it has a reverse effect on strain to failure. Nicolais et al. also reported that the flexural strength decreases as particle amount increases.³⁶ Stress transfer and elastic deformation from matrix to filler depend on static adhesion strength and interfacial stiffness.³⁷ The overall interaction between the filler particles and the matrix also rely on the surface area of the filler particles. With the increase of particle size, relative surface area decreases; consequently the interaction and thus the adhesion property decreases. Agglomeration of filler particles would result in larger size of the fillers. In the same way this corresponds to the lower strength for the sand composites with higher filler contents such as 3 and 5 wt % because the agglomeration of the sand particles becomes more influential.

A composite is made of filler phase and matrix phase. Because of the rigid nature of inorganic fillers, deformation of composite specimens is a consequence of the polymer behavior. The actual deformation of the composite specimen is very small compared to that of the neat polymer, and it results in a decrease in strain with increase of filler content. In addition, the size of particles affects the strain further; the higher the particle size, the lower the strain.³⁴ As mentioned above, agglomeration of the

TABLE I
Effect of Sand Particles on the CTE of Sand/Epoxy Composite

| | CTE | |
|---------------------|--------------------------------------------------------------|--------------------------------------------------------------|
| | ($\mu\text{m}/\text{m } ^\circ\text{C}$) (Below T_g) | ($\mu\text{m}/\text{m } ^\circ\text{C}$) (Above T_g) |
| Neat epoxy | 73.5 ± 1.5 | 185.2 ± 2.0 |
| Epoxy + 1 wt % sand | 68.5 ± 1.0 | 178.5 ± 1.5 |
| Epoxy + 3 wt % sand | 66.2 ± 0.8 | 175.5 ± 1.5 |
| Epoxy + 5 wt % sand | 64.0 ± 0.8 | 166.3 ± 2.0 |

TABLE II
Flexural Properties of Neat Epoxy and Composites

| | σ_f /MPa (Mean \pm S.D.) | E_f /GPa (Mean \pm S.D.) | ε_f /% (Mean \pm S.D.) | β | α_0 (MPa) |
|---------------------|-----------------------------------|------------------------------|--------------------------------------|---------|------------------|
| Neat epoxy | 149.6 \pm 4.2 | 10.3 \pm 0.2 | 2.5 \pm 0.2 | 82.97 | 150 |
| Epoxy + 1 wt % sand | 160.8 \pm 5.2 | 11.2 \pm 0.3 | 2.3 \pm 0.2 | 18.25 | 165 |
| Epoxy + 3 wt % sand | 114.9 \pm 4.7 | 11.3 \pm 0.3 | 2.0 \pm 0.2 | 19.69 | 117 |
| Epoxy + 5 wt % sand | 106.9 \pm 5.3 | 10.7 \pm 0.3 | 1.7 \pm 0.2 | 12.88 | 109 |

sand particles would result in larger particles. Therefore, the strength and stiffness of the composites obtained from these experiments are reasonable and rational.

The failure modes of the composite specimens resulted from a variety of factors and mechanisms, such as dispersion and size of the particles, stress transfer between the matrix and particles, interface debonding, crack propagation, etc. Use of conventional analytical processes to characterize these modes is difficult to justify. Therefore, we rely on experimental results and the damage constitutive equation (eq. (1)) to assess the specimens in flexural tests.³⁸

$$\sigma = E\varepsilon(1 - \varpi) \quad (1)$$

where σ , ε , π , and E are stress, strain, damage parameter, and elastic modulus, respectively. It is assumed that damage of pure epoxy and sand reinforced epoxy specimens can be characterized by a Weibull strength distribution function (eq. (2)):

$$\varpi = H(E\varepsilon) = 1 - \exp\left[-\left(\frac{E\varepsilon}{\sigma_0}\right)^\beta\right] \quad (2)$$

where H , σ_0 , and β are cumulative probability of failure, Weibull scale parameter and Weibull shape parameter respectively. By taking logarithms on both sides of eqs. (2) and (3) can be obtained:

$$\ln\left[\ln\left(\frac{1}{1-\varpi}\right)\right] = \beta \ln(E\varepsilon) - \beta \ln(\sigma_0) \quad (3)$$

Weibull parameters were evaluated by regression method. In each set of flexural strength results for each type of specimen, the lowest to the highest stress values were assigned to a probability of failure based on its ranking, where ranking starts from 1 to n , and n is the number of specimens in each set. The following equation (eq. (4)) was used to evaluate the probability of failure for p th specimen³⁹:

$$\varpi = \frac{p - 0.5}{n} \quad (4)$$

Eq. (5) represents a linear plot of $\ln\left[\ln\left(\frac{1}{1-\varpi}\right)\right]$ versus $\ln(E\varepsilon)$; β and σ_0 are the slope and intercept of the

plot respectively. In Table II the values of β and σ_0 calculated for different contents are listed. β represents the scattering in strength data of the materials. Higher values of β denote less scattering of composite strength which means more uniform and narrow flaw distribution. From Table II, it can be understood that neat epoxy specimens had narrower flaw distributions and the addition of sand particles increased the distribution width. The standard deviation for each type of specimen listed in Table II demonstrates the same pattern. Stress concentration around the sand particles may contribute to the scattering of strength of the composites. σ_0 measures the nominal strength of materials. With increasing of value of σ_0 , the average strength of composite will increase. The values of σ_0 listed in Table II are as per the flexural strength of specimens.

A decrease in elongation is a typical phenomenon for composites reinforced by inorganic fillers. The hydrophobic characteristics and roughness of the sand particles assist in good adhesion between the epoxy matrix and particles. This improved adhesion and the volumetric restriction by the particles, both resist dilatational and distortional deformation of the polymer matrix and cause a decrease in ductility. However, in this experiment, a drastic decrease of elongation of samples at failure was not observed, and this is analyzed by a morphology study using SEM.

The fracture surfaces of the neat epoxy and the sand/epoxy composites were observed using SEM (see Fig. 5). It could be seen that neat epoxy resin exhibited a brittle fracture surface. This indicated a low fracture toughness of the unfilled epoxy. Compared with the image of neat epoxy, the fracture surfaces of the epoxy composites filled with 1 wt % sand presented different features. Rivers and branches in solid patterns are prominent in the fracture surface. The crack direction was changed, which implied that the path of the crack was distorted due to the presence of filler particles. More external energy could be dissipated during the creation of new surfaces by massive plastic deformation, leading to increased fracture strength of composites. With further loading of the sand particles the epoxy resin composites again presented smooth features on fracture surface with occasional river patterns in some region. Hence, sand particles in the composite

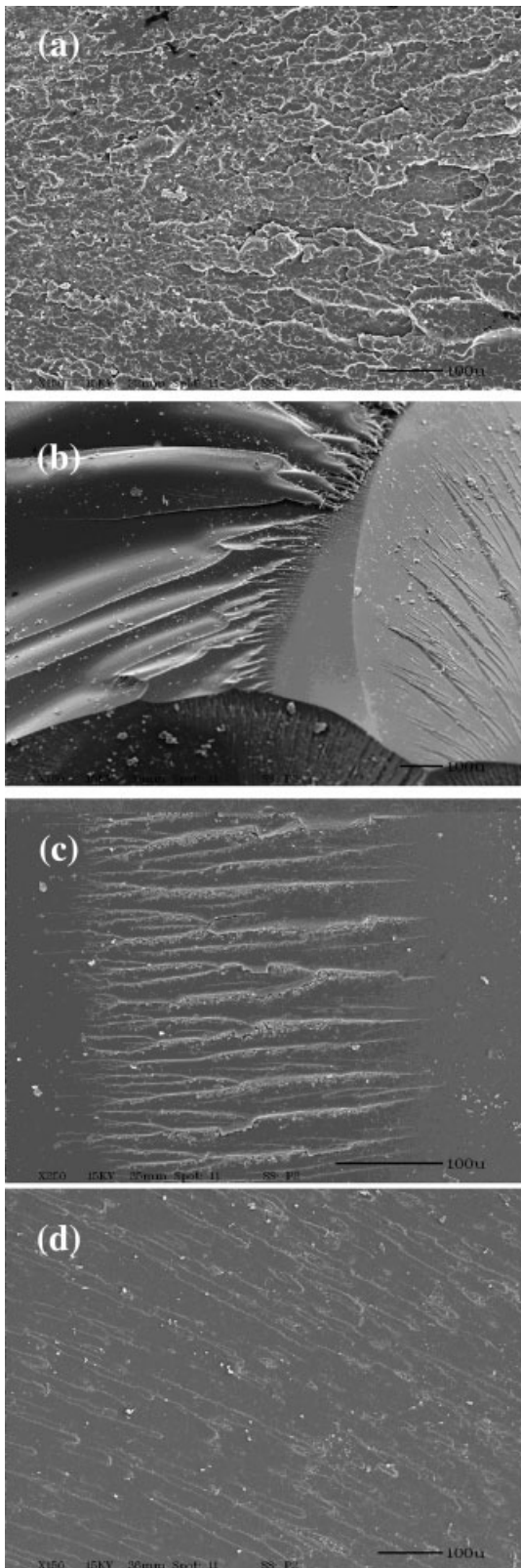


Figure 5 Fracture morphology of the epoxy and composites (a) neat epoxy; (b) epoxy + 1 wt % sand; (c) epoxy + 3 wt % sand; (d) epoxy + 5 wt % sand.

specimens (content >1 wt %) did not play a significant role in dissipating stress and enhancing toughness. This is because of the existence of agglomerations of the sand particles in the epoxy resin matrix, as seen in the SEM image of the composites with 5 wt % sand particles (Fig. 6). These sand particle agglomerates in samples act as structural defects and stress concentration sites, and will result in a drastic drop of mechanical performance of the materials. This would obviously be alleviated by adopting more effective dispersing methods and new technology in the process of materials preparation. Therefore, it is expected that improved processing conditions eliminating agglomeration levels of the sand particles would bring further increase in strength and modulus of the composites at higher filler loading, however as in all such cases there would be a level of filler content at which no further increases would be available as epoxy volume decreased.

Thermal conductivity

There is an extensive interest in the thermal conductivity of composites for their applications in industry. Unlike electrical conductivity, where an obvious percolation threshold exists, thermal conductivity of composites in many cases gradually changes with increasing filler content. Many factors, such as interfaces in polymer composites and additive characteristics impact the thermal conductivity. Inclusion of particles might assist in improving the thermal conductivity of the composite if the thermal conductivity of the particles is higher than that of the matrix and the interface is effective. In this study we used sand particles as inclusions which we assumed to

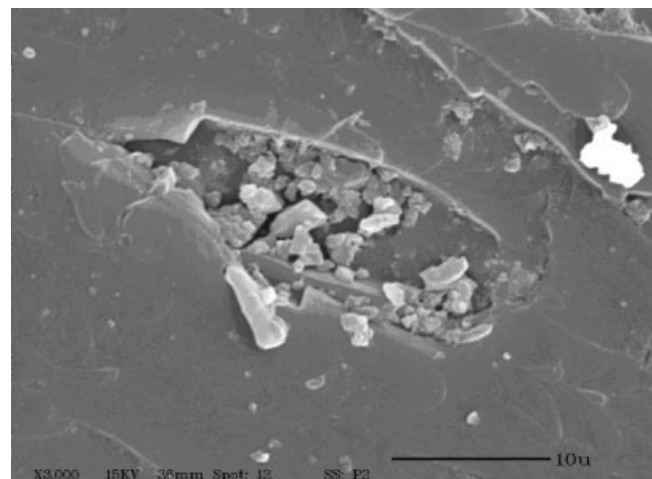


Figure 6 The accumulated sand particles in epoxy resin matrix.

TABLE III
Thermal Conductivity and Thermal Effusivity of Neat Epoxy and Composites

| | Thermal conductivity ($w/m \cdot k$) (experimental) | Thermal conductivity ($w/m \cdot k$) (theoretical) | Thermal effusivity ($w\sqrt{s}/m^2k$) |
|---------------------|----------------------------------------------------------|---------------------------------------------------------|--------------------------------------------|
| Neat Epoxy | 0.229 ± 0.010 | | 551.7 ± 16.3 |
| Epoxy + 1 wt % sand | 0.221 ± 0.012 | 0.231 | 528.9 ± 21.6 |
| Epoxy + 3 wt % sand | 0.241 ± 0.008 | 0.235 | 583.2 ± 18.2 |
| Epoxy + 5 wt % sand | 0.267 ± 0.005 | 0.240 | 622.7 ± 16.1 |

be composed of calcium carbonate²⁰ (some other ingredients were present but were ignored), with a thermal conductivity of $3.89 w/m \cdot k$.⁴⁰ The conductivity of epoxy is $0.229 w/m \cdot k$ (experimental value in Table III). Therefore it is expected that addition of sand particles would improve the conductivity of the composites. Table III shows the experimental thermal conductivity results of the epoxy and epoxy/sand composites, revealing that inclusion of sand in the epoxy did improve the conductivity of the epoxy composite material. Although the thermal conductivity of composites with 1 wt % sand is slightly lower than that of neat epoxy sample, the difference of the thermal conductivity between them is small and within the deviation of data. Thermal conductivity of composites with 5 wt % sand was highest among all composites and the increment was 17% compared to neat epoxy. A recent research work reported that 1 wt % loading of carbon nanotubes in epoxy resin enhanced thermal conductivity of an epoxy matrix from 0.22 to $0.24 w/m \cdot k$.⁴¹ Compared to this reported work, the enhancement of thermal conductivity properties in our experiment is more significant.

Heat transfer resistance can be denoted as the reciprocal of thermal conductivity. The overall thermal resistance can be calculated from the following equation (eq. (5))⁴²:

$$\frac{1}{k_{\text{overall}}} = \sum_{i=1}^N \frac{w_i}{k_i} = \frac{w_1}{k_1} + \frac{w_2}{k_2} + \frac{w_3}{k_3} + \dots + \frac{w_N}{k_N} \quad (5)$$

where w_i and k_i are the weight fraction and thermal conductivity of i th constituent phase respectively. Using eq. (5), the thermal conductivity of composites specimens with different sand loadings was calculated and they are listed in Table III. However, experimental results show greater improvement in thermal conductivity compared to theoretical ones except in composite containing 1 wt % sand. Other factors may play a role in the experimental results which are not accounted for in eq. (5) such as aspect ratio, packing density, volume fractions, tendency of the fillers to form chains, filler contiguity, etc. More investigation and work is planned for this aspect of the sand composites.

Thermal effusivity is a heat transfer property which determines the interfacial temperature when two objects at different temperature come in contact with each other. It is square root of the product of thermal conductivity and heat capacity. Therefore with the increase of thermal conductivity, effusivity increases. Thermal effusivity of the epoxy resin with varying contents of the sand particles are shown in Table III. At the 5 wt % content of the sand particles, the thermal effusivity of the sand filled epoxy resin increases 13 wt % over that of the neat epoxy resin and in other concentrated specimens, conductivity and effusivity tallied to each other values.

CONCLUSIONS

Epoxy composites were prepared and reinforced with a form of natural Cancun sand without complicated chemical treatment involved. Characterization of the mechanical and thermal properties of the epoxy composites containing up to 5 wt % of the sand particles revealed that this porous natural sand may be an effective (with respect to cost and performance) filler material for polymer composite materials with comprehensively enhanced properties. The sand composite with addition of 1 wt % of sand particles showed the highest flexural properties. The damage model used to interpret the flexural properties agrees with experimental results. DMA results showed the storage modulus of the composites at normal temperature exhibits a similar tendency to improved flexural properties. The high temperature storage modulus, glass transition temperature, and dimensional stability of the sand particles/epoxy composites monotonically increase with the addition of sand particles. The enhancement of thermally conductive properties in sand particles/epoxy resin composites is also apparent. Thermal modeling of these kinds of composites is expected to be complex due to natural variations in form and content, however, an initial comparison was conducted and a detailed work is planned for a future article. It is also very likely that improved processing conditions could eliminate or reduce the level of the sand particle aggregates, and thus, a further increase in thermal and

mechanical properties of the epoxy resin composites can be anticipated.

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