Effect of Silica Fillers on Mechanical Properties of Epoxy/Kenaf Composites

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Abstract. The mechanical properties of epoxy/kenaf composites at various silica content has been investigated. Epoxy resin was mixed with silica filler ranging from 6.7 to 33.3 %w/w before applied into kenaf fibre mat (11.5 %w/w) to fabricate epoxy/silica/kenaf composites using hand layup method. The composites were precured at 80 °C for 2 hour and post cured at 110 °C for 1 hour. The mechanical properties of the composite were analysed using flexural and impact test, meanwhile the surface fracture of the composite was examined by means of scanning electron microscope (SEM). It is found that epoxy/kenaf composite with 20 %w/w silica content exhibited the best mechanical properties, with 3.1 kJ/m² of impact strength, 44.5 MPa of flexural strength and 2.7 GPa of flexural modulus. Its surface morphology showed the brittle fracture surface with pull out of fibre which indicated poor interaction between epoxy matrix and kenaf fibre.

1. Introduction

Epoxy resin is one of the most important commercial thermosetting polymer in multitude of industrial applications. It exhibits many desirable properties, such as low shrinkage, high thermal stability, high stiffness and excellent chemical resistance. However, epoxy is generally brittle properties because of its high density of cross-linked structure. Therefore, improving the toughness of epoxy resin is an important concerned in epoxy composites for better product performance [1]. Several types of rigid inorganic particulate fillers have been studied to improve the mechanical properties of epoxy composite, such as silica, alumina, nanoclay and others. Among the numerous fillers of epoxy, silica is the most commercially used as a filler in literature due to its significant improvement in flexural modulus, fracture toughness and impact strength in epoxy composite [2]. It has been reported that fracture toughness of epoxy resins can be significantly improved by silica modification [3].

Carbon fibres and glass fibres integrated in thermoset polymer are the traditional reinforced composite material. Natural fibre reinforced composites found to be an alternative solution for greener environment and thus received increasingly attention in recent years. Among the natural fibre, kenaf fibre is more attractive since it is available commercially, comparatively cheap and strong [4]. Therefore, implementation of kenaf as a reinforce materials would be an interesting to study in
improving the mechanical properties of epoxy composites. Several researchers have studied on epoxy modification using different type of fillers to improve the mechanical properties of epoxy composites [1-3]. Marjetka (2013) has investigated the addition of silica improved the impact strength of epoxy from 6.4 kJ/m² to 8.9 kJ/m² [2]. Fiore et al. (2014) has proved that the used of multi-axial kenaf fibre mat increased the flexural modulus of epoxy from 3 to 7.82 GPa [5].

In this present work, polymer composites have been developed using kenaf fibre reinforced epoxy composites filled with different percentage of silica particles. The mechanical properties such as flexural strength, flexural modulus and impact strength have been investigated.

2. Experimental study

2.1 Materials

The epoxy resin used in this study was bisphenol A from Dow Chemical Pacific Singapore (DOW) whereas curing agent was cycloaliphatic amine (Jointmine 905-3S) from Epochemie. Nonwoven kenaf fibre mat was purchased from Kenaf Agro Vet Sdn Bhd, Malaysia. Silica powder with particle size 10 micron was bought from Maju Saintifik, Malaysia.

2.2 Preparation of epoxy/kenaf composites

Epoxy composites were prepared using hand lay-up method. The percentage weight of epoxy and kenaf mat were 88.5 % w/w and 11.5 % w/w respectively. The carbon steel mould (170 mm x 170 mm) was coated with mould release agent before placed with kenaf fibre mat. Epoxy resin was thoroughly mixed with curing agent before pouring into the mould to wet the kenaf fibre mat. The mould was then covered with top section and pressed with 1 kg load and heated at 80 °C for 2 hours. The curing was continued for another 1 hour at 110 °C for post curing process. The epoxy/kenaf composite sheet label as composite A was removed from the mould after cooled at room temperature and cut into specimen for mechanical testing.

2.3 Preparation of silica modified epoxy/kenaf composite

Epoxy resin was modified with silica filler by mechanical mixing process with silica content from 10, 15, 20, 25 and 30 %w/w for composites B, C, D, E and F respectively. The percentage weights of modified resin epoxy and kenaf mat were fixed at 88.5 % w/w and 11.5 % w/w respectively. The carbon steel mould (170 mm x 170 mm) was coated with mould release agent before placed with kenaf fibre mat. Similar to the above procedure, modified epoxy resin was thoroughly mixed with curing agent before pouring into the mould to wet the kenaf fibre mat. The mould was then covered with top section and pressed with 1 kg load and heated at 80 °C for 2 hours. The curing was continued for another 1 hour at 110 °C. The epoxy/kenaf composite sheet was removed from the mould after cooling at room temperature and cut into specimen for mechanical testing.

2.4 Mechanical properties

2.4.1 Flexural strength. Five samples from each epoxy composite were prepared for the test according to the ASTM D790 standard. Samples were cut using bandsaw machine and finished using milling machine. The samples were prepared in dimensions of 8 mm x 12.7 mm x 101 mm.

2.4.2 Impact properties. Charpy impact test was conducted according to ASTM D6110 standard. Five samples from each epoxy composites were prepared for the test. The samples were cut using bandsaw machine and finished using milling machine. The dimension of the sample was 5 mm x 12.7 mm x
63.5 mm. A v-notch was cut into the specimens using a sample notcher with an angle of 45°. The notched samples were left for 40 hours in room temperature before proceed for impact test in order to remove the stress concentration created by notching process.

3. Results and discussion

3.1 Flexural strength of epoxy/silica/kenaf composites

Figure 1 show the flexural strength of fully cured epoxy/kenaf composite (A) and epoxy/silica/kenaf composite labelled as B, C, D, E and F for 10, 15, 20, 25 and 30 %w/w silica content, respectively. Epoxy/kenaf composite (A) is able to withstand flexural strength up to 37.3 MPa as compared to 92.6 MPa in neat epoxy. The use of multi-axial kenaf fibre mat in composite was reported to lower flexural strength as compared to neat epoxy due to incompatibility of kenaf fibre and epoxy matrix which lead to poor interfacial bonding. Poor interaction between fibres, void formation and dispersion problem were also reported among the reason for lower flexural strength [5]. However, the present of silica fillers in the matrix composites B to F have improved the flexural strengths as compared to epoxy composite without silica (composite A). Composite C with 13 %w/w silica content show the highest flexural strength. Rigid particulate filler such as silica able to enhance the stiffness of the epoxy matrix [6].

![Figure 1. Flexural strength of epoxy/kenaf composites with different silica composition.](image)

3.2 Flexural Modulus of epoxy/kenaf/silica composites

Figure 2 shows the flexural modulus of epoxy/kenaf composites (A) without silica and epoxy/silica/kenaf composite labelled as B, C, D, E and F. Similar pattern to the result of flexural strength was observed for flexural modulus. Epoxy/kenaf composites exhibits flexural modulus of 1.96 GPa as compared to 3.00 GPa as recorded for neat epoxy. This result is however not in agreement with the finding by Fiore et al. (2014), where epoxy composites with kenaf fibre mat should exhibits
higher flexural modulus than neat epoxy [7]. This may because the fibres applied were not properly compacted and a lot of voids between the resin which may reduce the strength of the composites. Furthermore, the surface of fibre was not treated so it was not properly attached to the epoxy matrix. Therefore, the interfacial adhesion between fibre and epoxy resin is weak [8].

![Fig 2. Flexural moduli of silica modified epoxy composites.](image)

Addition of silica content has found increasing the flexural modulus as shown on composites B to D. The addition of 20 %w/w of silica in composites D denotes the highest flexural strength among the silica modified epoxy composites. However, there is no significant flexural strength improvement in composite E as compared to composite C. The results show that addition of silica above 20% w/w has reduced the flexural modulus. It is probably due to silica content overloaded and caused agglomeration formation in epoxy resin, which subsequently leads to poor stress transfer [9].

### 3.3 Impact strength of epoxy/kenaf/silica composites

Figure 3 shows the impact strength of epoxy/kenaf composite (A) as compared to epoxy/silica/kenaf composites (B to F) using charpy impact test. It is found that the impact strength of epoxy/kenaf composites is about 1.6 kJ/m² which is higher than the impact strength of neat epoxy (1.5 kJ/m²). The impact strength was further increased as the silica content increased in the epoxy matrix of composites. Addition of continuous fibre of kenaf mat gave additional energy absorption during impact [10]. However, increasing silica content up to 15 %w/w gave significant improvement of impact strength by 93 %. The highest impact strength (3.1 kJ/m²) was observed when 20 %w/w silica content was used in composites. The impact strength significantly reduced when the silica filler content above 20 %w/w.
Overall results of mechanical properties proved that the addition of silica content improves the flexural strength, flexural modulus and impact strength of epoxy/kenaf composites. This is because the presence of silica creates plastic void growth around debonded particles, which may increase the difficulty of crack propagation during fracture [3]. In general, a relatively high loading of silica (up to 20 %w/w) is required to achieve reasonable toughening effect in epoxy composite, which would also increase the viscosity of the resin mixtures [11]. Composite D denotes the best impact strength and the best flexural modulus among silica modified epoxy composites. Above 20 %w/w of silica loading reduce the flexural strength, flexural modulus and impact strength of epoxy/kenaf composites. At higher filler loading disturb the matrix continuity leading to stress concentrations, which can act as micro-crack initiator and reduce the adhesion and energy absorption capacity of composites [6].

3.4 Morphology analysis of epoxy/kenaf/silica composites

Figure 4 reveals the morphology of fracture surface of composite D. Based on the SEM image, the cloudy spherical shape in matrix indicating the voids from silica particles were pulled out from the matrix of the composites. Silica particles at 20 %w/w was found well distributed in epoxy matrix, this it was in agreement with the findings of Xu et al. (2015) [12]. The dark circles in matrix indicated that the void have created by fibre pull-out from the matrix. The arrows in Figure 4(a) show the unbroken fibre pulled out from the opposite sample’s surface. The clear pull out of the fibres indicate the poor interfacial adhesion of the kenaf fibres with the epoxy matrix. The existence of dark circles reflects to the failure of composite which is contributed by the poor interfacial adhesion between fibre and matrix, instead of the strength of the fibre itself [13]. Figure 4(b) shows the cross section of kenaf fibre embedded in the epoxy matrix. Small gaps were seen between the fibre and matrix (indicated by arrows), which supported that the interfacial bonding between kenaf fibre and epoxy resin was poor. The kenaf fibres found in the matrix were hollow in shape which hindered the resin to penetrate the bundle of the fibre as a result gave poor mechanical properties of composite [13].

High magnification image of fibre surface in epoxy composites is shown in Figure 4(c). It indicated that the smooth surface of kenaf fibre may limits the fibre and epoxy matrix interaction, thus
decreased the mechanical properties [10]. A higher mechanical property of composites may be obtained if the surface of fibre is treated to become more hydrophobic for better interfacial adhesion with the epoxy matrix.

Figure 4. SEM images of (a) fracture surfaces composite E, (b) cross section of kenaf fibre and (c) fibre surface.

4. Conclusion

Overall results found that mechanical properties of epoxy composites has increased as the silica content increased up to 20% w/w. Addition of 20% w/w silica into epoxy composites have shown the highest impact strength i.e 3.0 kJ/m² and flexural modulus i.e 2.9 GPa whereas the flexural strength is 49 MPa. Beyond the addition of 20% w/w silica content, epoxy composites exhibit reduction in flexural strength, modulus and impact strength. Surface fracture analysis using SEM has revealed that no interfacial interaction between kenaf and epoxy matrix.

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