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# Mechanical and Thermal Properties of Epoxy/Tamarind Shell Composite

## (Ciri-ciri Mekanikal dan Terma Komposit Epoksi/Kulit Asam Jawa)

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ABSTRACT

*This study explores the potential of tamarind shells as a filler in epoxy composites. The tamarind shells were collected from local supply and processed by washing it repeatedly using distilled water. The tamarind shells were dried and crushed to form fine particles. Epoxy composites were produced by mixing epoxy and hardener with varying (25,40,50,60) wt % of tamarind shells powder to achieve the desired properties. The samples underwent density test, flexural test, hardness test, thermal stability test, and morphology in order to analyse the mechanical and thermal properties of the samples. With the addition of tamarind shell particle, hardness and flexural strength show improvement about ~ 80% and 147%, respectively. However, density and thermal stability show decrement in value.*

*Keywords: Tamarind shell; mechanical; thermal stability; morphology; epoxy composite*

ABSTRAK

*Kertas kajian ini meneroka potensi kulit asam jawa sebagai pengisi dalam komposit epoksi. Kulit asam jawa dikumpul dari pembekal tempatan lalu diproses dengan mencucinya menggunakan air suling. Kulit asam jawa dikeringkan dan dihancurkan sehingga membentuk partikel halus. Komposit epoksi dihasilkan dengan mencampurkan epoksi dan pengeras dengan serbuk kulit asam jawa berlainan berat (25,40,50,60) wt % untuk mencapai ciri-ciri yang dikehendaki. Sampel kemudian menjalani ujian ketumpatan, ujian kelenturan, ujian kekerasan, ujian kestabilan terma, dan morfologi untuk menganalisis sifat sampel. Dengan penambahan partikel kulit asam jawa, kekerasan dan kekuatan lenturan menunjukkan peningkatan sekitar ~ 80% dan 147%. Walau bagaimanapun, ketumpatan dan kestabilan terma menunjukkan penurunan nilai.*

*Kata kunci: Kulit asam jawa; mekanikal; kestabilan terma; morfologi; komposit epoksi*

## INTRODUCTION

In the recent years, natural fiber reinforced polymer composites have found extensive usage in several fields of industries such as automotive industry, deck surfaces, and furniture. Such exposure is due to its appealing mechanical properties such as environmentally friendly, low cost, and non-toxicity compared to synthetic fibers (Zafar & Siddiqui 2017). In this particular study, tamarind fruit fibers were chosen because of the nature of its abundance availability due to the large quantities of this fiber produced in food processing industries as waste products (Maheswari et al. 2008). The vitality of using the wastage as the main component of the composites is mainly to reduce the waste products that are known to seriously contribute in the environmental pollution.

Composites fabricated from epoxy matrix and tamarind shells as a reinforcement have vast potential to be applied in various applications within the composite industry. The possibility to solve the environmental problem due to agricultural waste can be done by recycling the tamarind shells. Other than that, this study provides a possible solution to lessen the cost of productions in composites material due to the ease of producing the tamarind shells. Moreover, the use of tamarind shells as a filler is expected to cater other issues regarding existing glass fibers that can jeopardise the health of its user. Prolonged exposure to fiber glass is known to have harmful effect such as depression of lymphocyte in blood which makes tamarind shells as an excellent possible substitute since up to this date it is not known to have any side effects on the users (Binoj et al. 2017).

Traditionally, tamarind paste has been a substantial ingredient in a wide variety of food products such as traditional food, juices and fast food whose production results in several tons of waste in terms of shell and seeds which contribute to the environmental issues in Malaysia and alike. Agricultural waste is an epitome to the importance of efficient recycling culture since the number of wastage is inclining over the years due to the demand of the products (Goudar & Shetty 2017). The tamarind shells in form of outer crust of tamarind is an inedible product which is normally used and disposed of as a waste in a large amount of quantities. The necessity of venturing into possibilities of reuse and recycle of the tamarind shells waste into environmentally friendly and usable products is expected not only to cater to the environmental issues but also to broaden the versatility of the tamarind itself. Besides, this research can help the manufacturer financially to have add-on product so that they can sell reusable tamarind shell wastes to composite manufacturing industry. By using tamarind shells as a form of filler in composite materials, ways of minimising the cost of manufacturing of composite and reducing the dependence of existing filler such as glass fiber and wood fiber can be established. The existing fiber is mostly expensive and must undergo difficult process to become a filler. Therefore, the use of tamarind shell bio-filler in the polymer matrix such as epoxy resin will help to reduce dependence on the existing filler and minimise the manufacturing cost and time of composite materials.

The main goal of this project is to use tamarind shell research to create the right recycling path. Moreover, the processing of tamarind shells into valuable product is potentially profitable in various manufacturing industries such as food processing industry while also acts as a manufacturing method for the composite industry.

## METHODOLOGY

### MATERIALS AND PREPARATION OF SAMPLE

The tamarind shell wastes that have been collected were cleaned using distilled water repeatedly before being subjected to other processes. After that, the tamarind shells were left at room temperature to dry overnight. The dried tamarind shells were then dried again at constant temperature of 40 ºC in an oven for five hours. The dried tamarind shells were then grinded into smaller particles by using dry grinder until the tamarind shells formed into fine particles. The type of epoxy resin used in this experiment is Epikote 240 and hardener used is Epikure 3090. The epoxy is mixed with tamarind shells with different weight percentage of tamarind shell particle of 25wt%, 40wt%, 50wt%, and 60wt% respectively according to the percentages of weight composition of samples. Around six samples were prepared and the designation of samples are shown in Table 1.

TABLE 1. Designation of sample

| Types of sample | Abbreviation |
|---|---|
| Neat Epoxy | E |
| Epoxy and 25 wt% tamarind shell particle | ET25 |
| Epoxy and 40 wt% tamarind shell particle | ET40 |
| Epoxy and 50 wt% tamarind shell particle | ET50 |
| Epoxy and 60 wt% tamarind shell particle | ET60 |

The mixing process was conducted using mechanical stirrer at a speed of 200 rpm for 15 minutes. Then, the hardener solution was added into the epoxy mixture with ratio of epoxy and hardener 1:0.4. The mixing of the tamarind shell/epoxy composite took about five minutes. Then, the mixture of the epoxy tamarind shells composite was poured into the square mould. The mixture was left in curing stage at room temperature for 24 hours.

## CHARACTERISATION

The actual densities of the epoxy tamarind composites were measured by using the Archimedes principle. According to this principle, when an object is immersed in a liquid, the apparent loss in its weight is equal to the weight of the liquid it displaces. To conduct the test, distilled water was chosen as the medium. All the specimen was weighed using Electronic Weighing FX-300i Precision Balance. This method is covered in ASTM standard D792.

The density of the composite was obtained by using the following Equation (1).

$$\rho_a = \frac{\rho_w W_a}{W_a - W_w} \quad (1)$$

Where

$\rho_a$ is actual density of composite,
$\rho_w$ is the density of distilled water,
$W_a$ is weight of the sample in air and,
$W_w$ is weight of the sample in water.

The hardness of the samples was measured using Durometer Type D. Before undergoing the hardness test process, the samples must be placed on a stable and flat surface. The flexural test of the samples was executed by Universal Testing Machine (UTM), Instron 5569A. The samples were prepared according to ASTM D790 standards with crossheads speed of 2 min/mm. The specimens were placed at the two supports which have a distance of 40 mm and the load was given at the centre which is called as three-point bending test. Thermal stability test was analysed using STA 8000 Perkin Elmer with temperature range of 30–1200 ◦C at a heating rate of 10 ºC/min under a nitrogen flow rate of 20 mL/min. The morphology of composite was studied using a field emission scanning electron microscope (FESEM). The composites were examined with a Gemini 500, Carl Zeiss FESEM, and gold coated before recording the micrograms. The activation voltage varied from 2 to 3 kV with magnification of 1 000x.

## RESULTS AND DISCUSSION

The effect of density tamarind shell particle in the epoxy matrix with composition of ET 0%, ET 25%, ET 40%, ET 50%, and ET 60% are shown in Figure 1. Based on Figure 1, the increasing of tamarind shell particles in epoxy matrix is found to reduce the density of the samples. The sample ET 60% shows the lowest density which is 1.07 g/cm3 while the highest density is neat epoxy (ET 0%) which is 1.487 g/cm3 with 28% percentages of decrement. The decrease in densities of the hybrid composites was due to the presence of low-density tamarind shell particle (Binoj et al. 2018). Naik et al. (2019) reported the mixing of two materials with different densities will result in in-between values of those two material's densities (Naik et al. 2019).

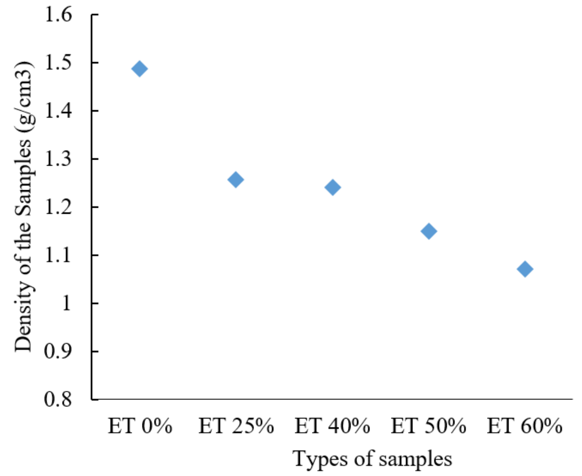


FIGURE 1. Graph of density test of tamarind shells fiber and epoxy composite

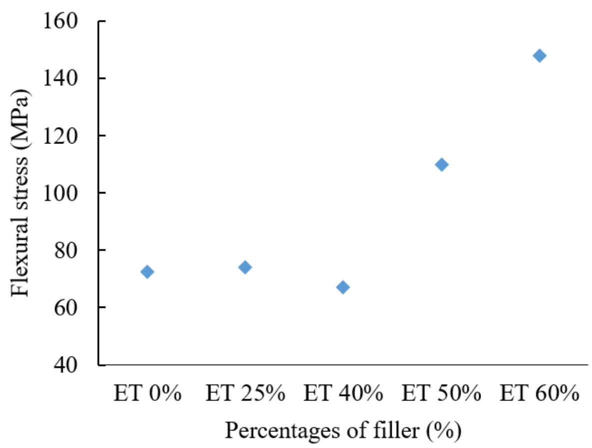


FIGURE 2. Flexural stress curve of tamarind shells fiber and epoxy composite

Based on the data analysis from Figure 2, the maximum flexural stress can be observed in the sample of ET 60% which is 148.00 N/mm2 and the lowest flexural stress can be observed in the sample of ET 40% which is 67 N/mm2. The bonding between the fibers and matrix were evenly distributed at first until the applied load reached the limit which caused the propagation of crack to begin. The propagation of crack was due to poor adhesion bonds between the tamarind shell fibers and epoxy matrix. This statement was supported by the same result of increasing of flexural strength as the filler increases until it reaches the maximum flexural stress (Srinivas et al. 2017). Figure 3 demonstrates the effect of adding tamarind shell particles on the hardness property of epoxy composite. The optimum hardness for epoxy is obtained at 60 wt% tamarind shell particles. It can be seen from the result obtained that hardness value for the composites increases with increasing tamarind shell particle loads. These finding was also supported by (Subramonian et al 2016; Cao et al. 2006; Gomes et al. 2007; Puneeth et al. 2019; Santosha et al. 2020). The hardness increases when the resistance of the materials to the deformation increases. As the percentage of the filler increases, the composite becomes harder and improves the hardness of the samples. The layer of the filler gives better resistance to the plastic deformation.

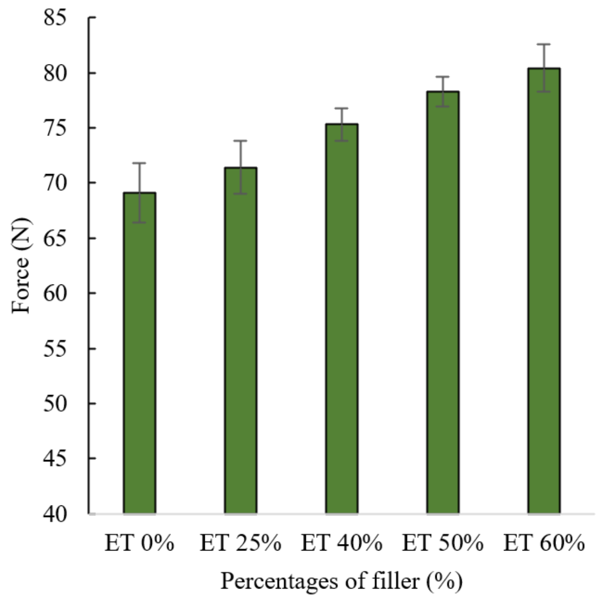


FIGURE 3. Hardness test result

Thermal stability of tamarind shell particle in epoxy matrix was analysed using thermogravimetric analysis. It can be seen in Figure 4 that the TG curves of all samples show similar trends. These trends are apparently exhibiting four sequent stages (or temperature regions) basically associated with: (1) dewatering of the biomass samples, (2) volatilisation of high-reactive chemical components of the shells (cellulose, hemicellulose, and partly lignin) accompanied by volatile oxidation (3) volatilisation of the rest lignin and other high-molecular extractives accompanied by oxidation of volatile matter and char, and (4) low-rate oxidation of chars.

The first stage of mass loss occurs at temperature between 250 °C and 350 °C which causes loss of humidity content of 10 wt%. In the first stage of combustion process, all of the samples show its properties of losing moisture content in the samples (Prabhakar et al. 2015). The highest initial degradation temperature is sample ET 0% which is 326.5 °C and the lowest initial degradation temperature is sample ET 50% which is 261.7 °C. The second stage of decomposition temperature is between 350 °C to 430 °C. The decomposition in this stage is due to the breakdown of the hemicellulose, cellulose, and lignin of the samples. In this stage, the highest degradation temperature is sample ET 0% which is 420.5 °C and the lowest degradation temperature is sample ET 60% which is 366.8 °C.

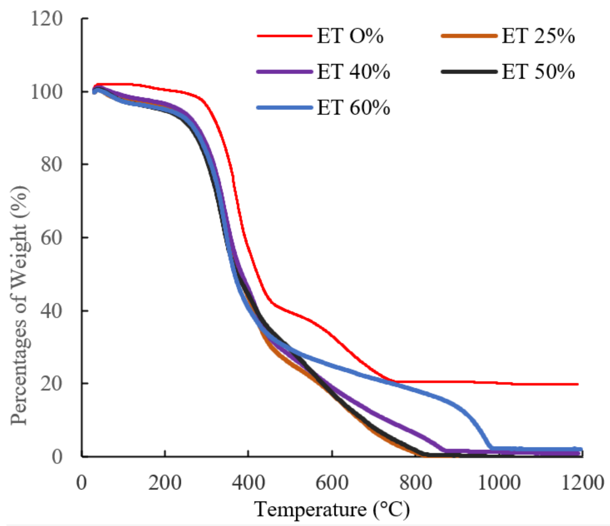


FIGURE 4. Weight loss and degradation temperature curve of different percentages of filler

In the final stage, the highest mass residue is in sample ET 0% raking at 20% whereas the lowest mass residue is in sample ET 25% at 0.05%. This result may be due to poor interfacial characteristics with epoxy matrix compare. Similar result using peanut shell as filler was reported by Prabhakar et.al. (2015). Furthermore, Ujjinappa and Sreepathi (2018) proved the suitability of using tamarind shell as fuel briquette which is due to its good combustion characteristics.

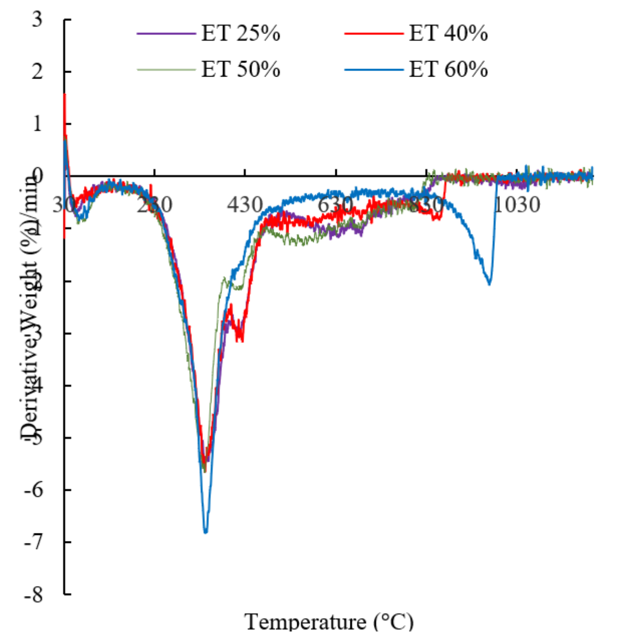


FIGURE 5. Graph of derivative weight of different percentages of tamarind shell particle in epoxy matrix

Figure 5 depicts the DTG curves of the selected composites. The lowest and the highest thermal degradation occurs on sample ET 25% and sample ET 60% at temperature of 55.9 °C and 63.2 °C, respectively. For first decomposition temperature, the presence of tamarind shell fiber is predicted to increase the thermal stability of the epoxy composite.

Next, the second decomposition temperature of epoxy tamarind shell occurs between 350 °C to 430°C. This result was confirmed by using Derivative Thermogravimetric Graph (DTG). In this step, particular data such as corresponding weight loss, onset temperature of thermal degradation, and peak of weight loss temperature of the specimen can be determined using DTG curves. The highest peak of weight loss temperature is in sample ET 40% which is 341.3 °C whereas the lowest peak of weight loss temperature is in sample ET 60% which is 340.4 °C. In this stage, the degradation of some lignin part and hemicellulose part inside the samples occurred (Maheswari et al. 2008). This degradation reaction causes the biggest sample mass losses in this stage and the peak of the degradation temperature occurs between 300 °C and 400 °C.

Graph of DTG for ET 0, ET 40%, and tamarind shell particle are shown in Figure 6. In this analysis, the best samples of epoxy tamarind shell were chosen based on its thermal properties to compare with tamarind shell particle and neat epoxy sample. Based on Figure 6, the neat epoxy shows better thermal behaviour with peak of weight loss temperature at 371 °C. Furthermore, the final mass residue of neat epoxy sample is 20% after thermal degradation process which is higher compared to others.

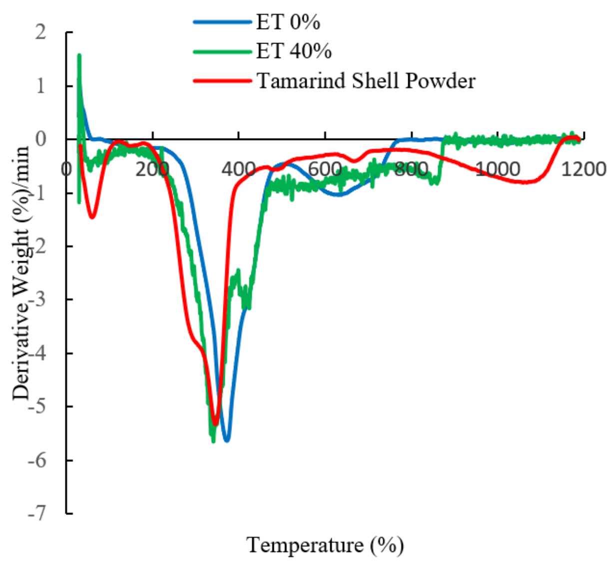


FIGURE 6. Derivative weight curve of neat epoxy (ET 0%), epoxy tamarind shell fiber (ET 40%) and tamarind shell particle

Based on the topographical images done using FESEM analysis, the fractography structure of the composite was observed. Figure 7 shows the surface of the neat epoxy which exhibits brittle properties based on the clean and smooth fracture appearance. The structure of tamarind shell particle is shown in Figure 8. The average size of the particles can be confirmed below than 100 $\mu$m under the microscope observation. The average size of the particles is within the expected range since the acceptable size of particles was fixed in this study to be below 100 $\mu$m. The addition of tamarind shell particle covers the brittle appearance of the neat epoxy and alters the surface appearance of the composites as shown in Figure 9 and Figure 10. The morphological structure of epoxy tamarind shell in Figure 9 with percentage of 50% tamarind shell fiber shows good interfacial bonding of epoxy matrix with tamarind shell fiber. From Figure 9, it can be observed that the tamarind shell fiber was evenly distributed in epoxy matrix compared with epoxy with 25% tamarind shell fiber in Figure 10. This property gives the result of higher mechanical properties in terms of flexural and hardness as the higher force can be exerted in epoxy composite in Figure 9 since the tamarind shell fiber improves the arrangement of particles in epoxy matrix.

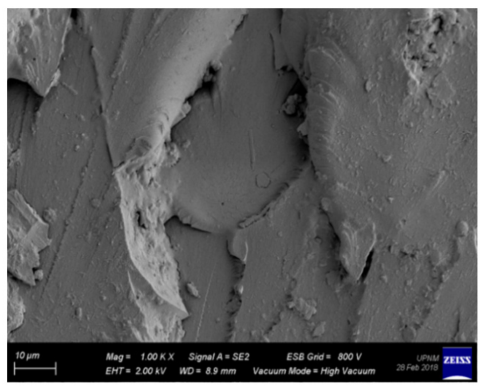

FIGURE 7. The image of neat epoxy samples under 1000x magnification

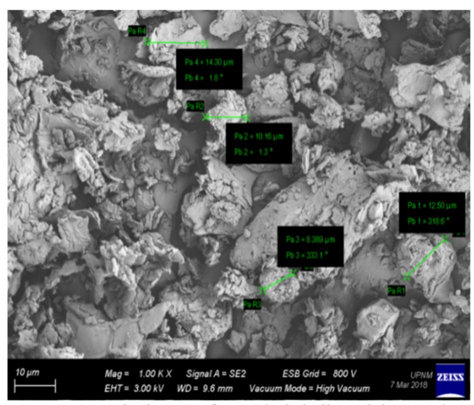

FIGURE 8. The image of tamarind shell particle samples under 1000x magnification

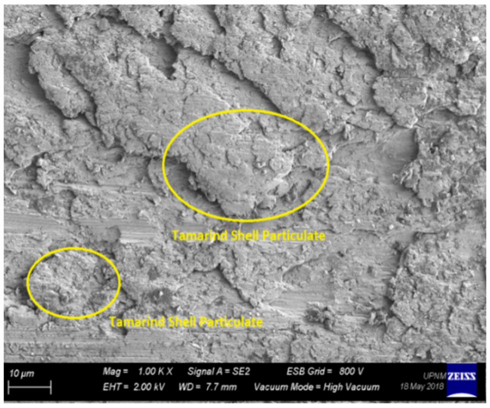


FIGURE 9. The image of 50% tamarind shell particle in epoxy composite samples under 1000x magnification

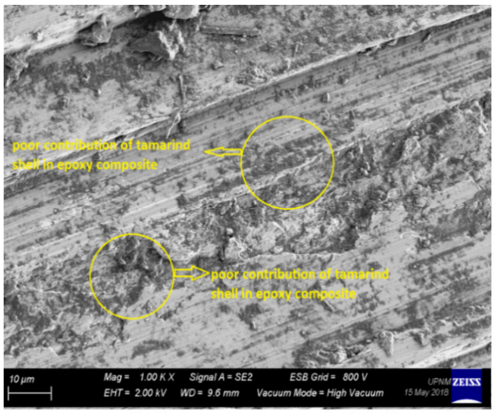


FIGURE 10. The image of 25% tamarind shell particle in epoxy composite samples under 1000x magnification

## CONCLUSION

The density of the composite decreases with the increment of filler percentage. For flexural test, the best sample is sample ET 50% based on its high flexural modulus and stress. The hardness test reveals the result that the higher the percentage of filler, the higher the hardness of the samples. For thermal properties, the result shows that the incorporation of tamarind powder in epoxy matrix gives lower thermal stability.

### DECLARATION OF COMPETING INTEREST

None

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