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## THERMAL DEGRADATION STUDY OF KENAF FIBRE/EPOXY COMPOSITES USING THERMO GRAVIMETRIC ANALYSIS

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### ABSTRACT

Kenaf fibres are receiving much attention in the natural fibre composite industry due to its potential as polymer reinforcements. However, like all natural fibres, kenaf fibres have lower thermal resistance as compared to synthetic fibres. In this current work, the characteristics of kenaf fibre/epoxy composites, both treated and untreated using alkalization process, exposed to high temperature were studied. Thermo gravimetric analysis (TGA) was used to study the thermal decomposition behaviour of treated and untreated kenaf fibre/epoxy composites, glass fibre/epoxy composite as well as neat epoxy from room temperature up to 600 °C. Surface morphology of both kenaf fibre/epoxy composites after exposure at 100 °C was observed using scanning electron microscopy (SEM). The results from the TGA showed that the addition of kenaf fibres into the epoxy slightly improves both the charring and thermal stability of the samples. However, it was observed that alkalization of fibres causes reduction in these behaviours. At 100 °C, the SEMs show more voids in the untreated composites than the treated ones, suggesting higher moisture content within the voids which influences the higher weight loss of untreated composite at this temperature.

**Keywords:** natural fibres composites, kenaf fibre, thermal degradation, Thermo Gravimetric Analysis (TGA).

### INTRODUCTION

Fibre reinforced polymer composite or more easily referred to as FRP is a relatively new material in the construction industry as compared to steel and concrete (Awad et al., 2012). The commonly used synthetic fibres are glass, aramid and carbon (Hollaway, 2010). However, these fibres face some competition from natural fibres due to the emerging concept of green-building. The advantages of natural fibres over its synthetic counterparts include low weight, low cost, low density, biodegradable, availability from renewable resources, and good thermal and acoustic insulation properties (Dittenber and GangaRao, 2012, Liu et al., 2012, Araújo et al., 2008). Also, they are non-abrasive on processing equipments and provide safer and healthier working environment (John and Thomas, 2008). What makes natural fibre appealing as a composite reinforcement material is its biodegradability which can contribute to the reduction of construction waste (Dittenber and GangaRao, 2012) thus reduces landfill dependency. However, this very attraction also imposes a great drawback on its durability. The disadvantages of natural fibre includes the incompatibility between fibre and polymer, low moisture resistance, inferior fire resistance, limited processing temperature, lower durability, and variation in quality and price (Dittenber and GangaRao, 2012, Araújo et al., 2008). Presence of hydroxyl groups in natural fibres makes them hydrophilic in nature and this

causes incompatibility with the hydrophobic polymer matrix (Araújo et al., 2008, Dittenber and GangaRao, 2012, Shih, 2007, Chen et al., 2009). The hydrophilicity of natural fibres leads to low interfacial strength and generates high moisture absorption that causes composites to fail in wet condition through fibre swelling and delamination (Joseph et al., 2002, Assarar et al., 2011). In terms of exposure to high temperature, majority of natural fibres have low degradation temperatures which are inadequate for processing with thermoplastics with processing temperatures higher than 200°C (Araújo et al., 2008).

The major issue that hinders the widespread use of FRP in structural engineering application is the degree of fire resistance of the material and the limited amount of information regarding FRP behaviour in fire. At lower temperature of 100-200 °C, FRP soften, creep and distort causing buckling for load bearing structures while at 300-500 °C, polymer matrix decomposes, releasing heat and toxic volatiles (Hollaway, 2010). Decomposition of burning polymers includes the production of combustible gases, non-combustible gases, liquids, solids (usually char), and entrained solid particles (smoke). These outcomes may produce hazards such as the evolution of toxic gases (in the case of PVC), loss of physical integrity, and melting and dripping thereby providing other ignition sources (in the case of polyolefins and polyethylene) (García et al., 2009, Stark et al., 2010).

For natural fibres, flammability is in part due to differences in chemical composition. Higher cellulose content results in higher flammability while higher lignin content results in greater char formation with lower degradation temperature (Dittenber and GangaRao, 2012, Manfredi et al., 2006, Suardana et al., 2011). Char formation is important during a fire as it protects core of material and structural integrity. Other attributes that provide better fire performance includes the present of silica or ash and higher crystallinity and lower polymerization of fibre microstructure (Dittenber and GangaRao, 2012). Among the natural fibres studied, flax fibres are considered the best for thermal resistance due to their low lignin content (Manfredi et al., 2006).

Temperature plays an influential role in the thermal stability of natural fibre composite where it causes direct thermal expansion or contraction and affects rate and volume of moisture absorption that leads to fibre swelling (Wang et al., 2005). The degradation process of natural fibres includes dehydration combined with emission of volatile components initiating at a temperature of about 260 °C, and rapid weight loss due to oxidative decomposition corresponding to the formation of char as the temperature increased (Beg and Pickering, 2008). Thermo gravimetric analysis is one of the methods used to study the thermal degradation behaviour of natural fibre/polymer composites and its constituents. Approximately 60% of the thermal decomposition of most natural fibres occurred within a temperature range between 215 and 310 °C with an apparent activation energy of 160-170 kJ/mol (Yao et al., 2008). For example, fibres from water hyacinth, reed, sisal and roselle decompose at 290-490 °C (Methacanon et al., 2010) while bamboo fibres degrade at 250-420 °C (Lee and Wang, 2006). In this current work, investigation was carried out to study the degradability behaviour of kenaf/epoxy composite exposed to high temperature in comparison with neat epoxy. The effect of alkalization treatment on the thermal degradation of kenaf fibre/epoxy composite was also observed. It is important to analyse the behaviour of natural fibre/polymer composite subjected to thermal exposure in comparison to its synthetic counterpart in order to draw a conclusion to whether natural fibres are technically capable to replace synthetic fibres.

## EXPERIMENTAL DETAILS

### Materials Selection

Raw kenaf fibres were supplied by Malaysian Agricultural Research and Development Institute (MARDI). The fibres were soaked in warm water for 3 hours and then rinsed with tap water to remove any dirt or debris. The colour of the rinsed water was monitored to ensure that the fibres were cleaned thoroughly. The fibres were combed and then dried for 24 hours in an oven at a temperature of 40 °C. In the fibre treatment process, the cleaned kenaf fibres were cut into an average length of 100 mm. Sodium hydroxide (NaOH) solution was prepared with a 6wt% concentration. The selected fibres were immersed in NaOH aqueous solution for 24 hours at room temperature. After treatment, the fibres were thoroughly washed with tap water until all traces of NaOH were removed from the fibres and then dried for 24 hours in an oven at a temperature of 40 °C. The resin used in the current work is liquid epoxy (DER 331) which is a liquid reaction product of epichlorohydrin and dispanol A. It is suitable for applications such as casting and tooling, composites, and automotive parts. The curing agent used for the selected epoxy is JOINTMINE 905-3S, which is a low viscosity aliphatic amine for room temperature curing. It has good wetting properties and impact resistance.

### Epoxy Composites Preparation

In the fabrication process, the epoxy resin and hardener, with a ratio of 2:1, was uniformly mixed using an electric mixer and poured into the desired mould. The mould was placed in a vacuum chamber (MCP 004PLC) with a 0.5 bar pressure to get rid of any air bubbles which may have been trapped in the mould in between the fibres. The vacuumed block was kept for curing at room temperature for 24 hours. The volume fraction of the fibre in the matrix was controlled to be about 48% Vol.

### Thermal Gravimetric Analysis

Thermal gravimetric analysis was conducted using TGA Q500 machine. Samples weighing approximately 10mg were subjected to pyrolysis in nitrogen environment to a maximum temperature of 600 °C at a heating rate of 10 °C/min. The weight loss was recorded in response to increasing temperature, with final residue yield on set of degradation temperature and number of degradation steps reported. Additionally, glass fibre/epoxy composite sample was also tested for comparison.

### Scanning Electron Microscopy

Rectangular cubes of treated kenaf fibre/epoxy composites and untreated kenaf fibre/epoxy composites with the dimensions of 10x10x15 mm were prepared. Samples were placed into a laboratory muffle furnace and were heated to maximum temperatures of 100°C at a heating rate of 5 °C/min for 1 hour. Scanning electron microscopy was performed using JEOL JCM-6000 with a balance scale range of ± 0.1µg. The samples were coated with gold prior to conduction and the final morphologies of the samples were examined.

## RESULTS AND DISCUSSION

### Thermal Decomposition through Thermo Gravimetric Analysis

Figures 1 and 2 shows the TGA and DTA curves obtained from the runs. The data from both curves were extracted and presented in Table 1.

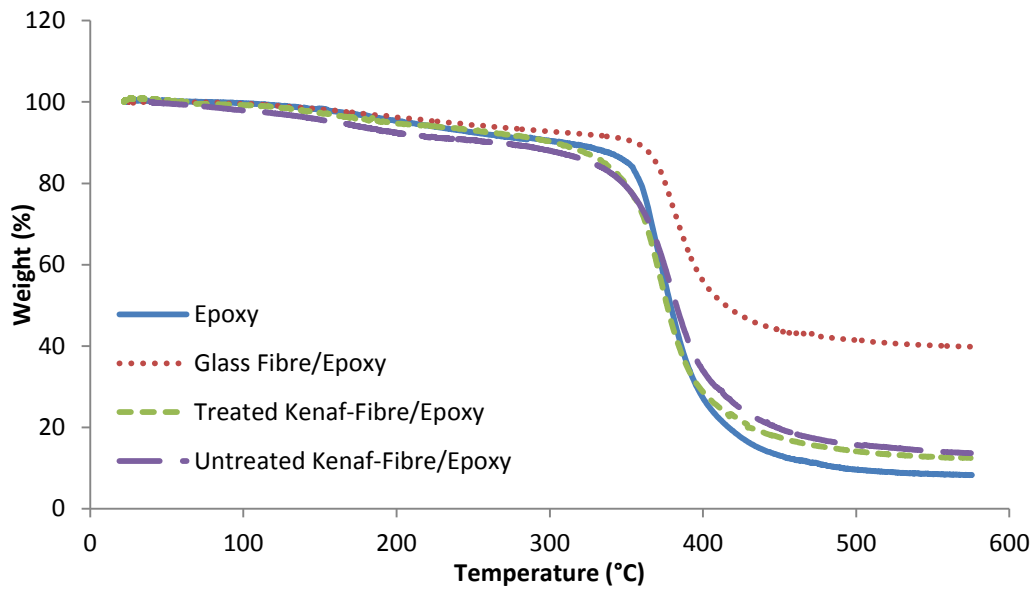


Figure 1. Thermo Gravimetric Analysis (TGA) curves

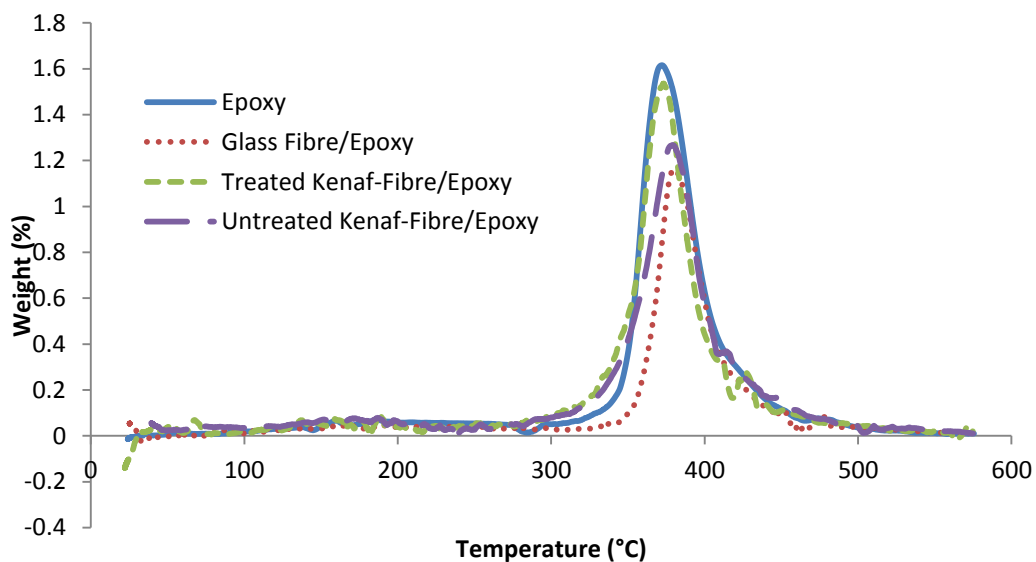


Figure 2. Differential Thermal Analysis (DTA) curves

Table 1. Decomposition temperature and charring of samples for epoxy and its composites.

Alkali Treatment (wt.%)	Epoxy	Glass Fibre/Epoxy	Treated Kenaf-Fibre/Epoxy	Untreated Kenaf-Fibre/Epoxy
Decomposition Temperature (°C)	371.99	380.12	373.47	378.64
Final Weight after Decomposition (%)	8.29	39.83	12.46	13.67
Increment in Thermal Stability (%)	-	2.19	0.40	1.79
Increment in Char Production (%)	-	380.46	50.30	64.90

From Figure 1, it can be seen that the untreated kenaf/epoxy composite starts to lose weight earlier than the other samples. This is attributed to the higher moisture content of untreated fibres whereby, the presence of hemicelluloses has caused higher moisture absorption of the composite (Methacanon et al., 2010). Moisture evaporates from the fibres starting at 80 °C. The percentage of weight reduction at 500°C reflects the amount of residues left after the composites were degraded. Epoxy has the lowest residue due to the absence of char. Treated kenaf/epoxy composite had lower residue than untreated kenaf/epoxy composite due to the removal of lignin through alkalization. Lignin in kenaf is responsible for charring thus untreated kenaf/epoxy composite will have more char (Beg and Pickering, 2008). The peaks of the DTA curves correspond to the decomposition temperature of each constituent of the composites. However, from Figure 2, only one peak is obvious for all curves due to the overlapping peaks of the fibres and epoxy (Azwa et al., 2013). It seems that neat epoxy has the lowest decomposition temperature at 371.99 °C while the addition of fibres had shifted the curves to higher temperatures. Some researchers have found that the addition of natural fibres causes reduction in the thermal stability of the composite due to the influence of the less stable fibres (Lee and Wang, 2006). It seems that in this study, the kenaf fibre plays a synergistic role in improving the thermal resistance of the composite. The charring provided by the degradation of the kenaf fibre may have contributed to this. Further investigation is needed to explain this behaviour.

Treated kenaf-fibre/epoxy is less stable than the untreated ones. This contradicts some studies that states that with the reduction of hemicelluloses and lignin from fibre treatment, thermal stability is increased (Methacanon et al., 2010, Beg and Pickering, 2008). However, through alkalization, cell walls of fibres are removed which may reduce the thermal protection of the treated fibres. Also, the exposure of cellulose to direct heat without any layering from the cell walls and hemicelluloses may contribute to this lack in thermal stability. This hypothesis is based on reports suggesting that flammability of natural fibres is contributed by its cellulose content (Suardana et al., 2011). As expected, glass fibre improved the thermal resistance of the composite the most and gives a charring structure at the end of the test.

### Morphology of Composites through Scanning Electron Microscopy

To study the reason on why untreated kenaf fibre/epoxy composite lost its weight at a lower temperature than the other samples, its morphology was observed using scanning electron microscopy (SEM) and compared to the treated sample. The SEM observations

reflect the results of the weight loss of the samples as discussed earlier. Figures 3 and 4 represent the micrographs of treated and untreated composites at 100 °C, respectively.

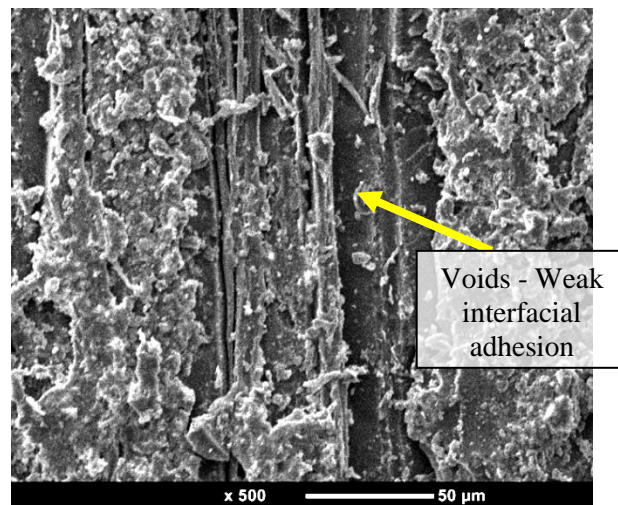


Figure 3. Micrograph of untreated kenaf/epoxy composite at 100 °C.

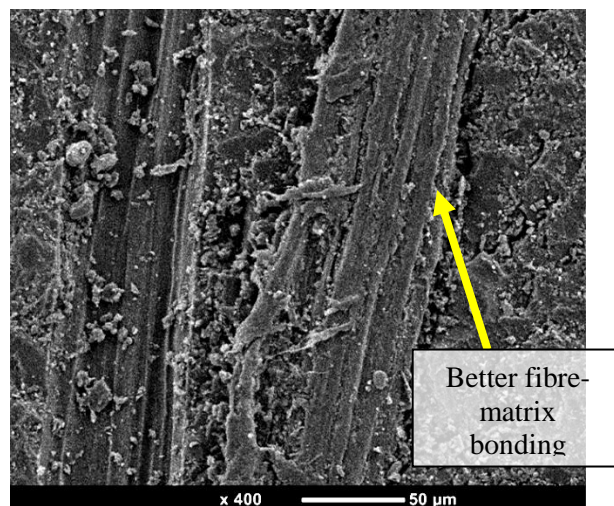


Figure 4. Micrograph of treated kenaf/epoxy composite at 100 °C.

For the untreated kenaf-fibre/epoxy composites, it is obvious that more voids are present along the fibre-matrix interface, as shown in Figure 3, suggesting weak interfacial adhesion. These areas provide spaces for moisture to occupy thus making untreated kenaf composites more vulnerable to moisture attack as compared to the treated ones (Azwa et al., 2013, Dittenber and GangaRao, 2012, Chen et al., 2009), which explains its higher initial weight loss. At lower temperature, treated kenaf-fibre/epoxy composite has better fibre/matrix bonding due to the removal of hydrophobic components of the fibre, allowing better compatibility between the kenaf fibre and the epoxy. This compatibility provides better interfacial adhesion and mechanical interlocking between the fibres and the matrix as observed in Figure 4, whereby the fibre surface is seen to be filled up by the epoxy. This improves the resistance of the composite to moisture thus, the lack of moisture leads to comparable percentage of weight loss to epoxy at temperature below 300 °C.

Observations on bamboo fibres at various degrees of alkalization was conducted by Wong et al. (2010) through SEM and it was discussed that the present of voids suggest that impurities and soluble substances were gradually removed as the concentration of alkali is increased. This increased in surface area provides more interface for fibre/matrix adhesion and with the absence of the hydrophilic component of the fibres, the resin has a better chance to seep through and bound with the fibres. Such improvement was also observed for treated kenaf fibres alone whereby the morphological changes provided better tensile strength and modulus of the fibres (Yousif et al., 2012). SEM studies were performed on tensile-tested coir/polyester composites which highlighted that the treated coir fibres showed a reduction in fibre pull-out with better dispersion of matrix in the fibres as compared to the untreated coir fibres (Rout et al., 2001). This is in agreement with Figure 4.

## CONCLUSION

Treated and untreated kenaf fibre/epoxy composites, glass fibre/epoxy composite as well as neat epoxy were subjected to thermal degradation by means of Thermo Gravimetric Analysis (TGA). From this study, it can be concluded that the addition of fibres into the epoxy improves the thermal stability of the samples as well as its charring capability, with glass fibres giving the best results. However, alkalization reduces the decomposition temperature of the kenaf fibre/epoxy composite and produces lesser char than untreated composite caused by the removal of lignin. Through SEM, it was confirmed that weak fibre-matrix adhesion of the kenaf fibre with the epoxy causes high moisture content of the composite which was removed at a temperature of 80 °C.

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