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Thermal and Dynamic Properties of Woven Kenaf/Carbon Fibre Reinforced Epoxy Hybrid Composite

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Abstract: The effects of carbon fiber hybridisation on the thermal properties of woven kenaf reinforced epoxy composites kenaf fibre were studied. Woven kenaf hybrid composites at the different weave designs of plain and satin, and fabric count of 5×5 and 6×6 were manually prepared by vacuum infusion technique. Thermal properties of pure carbon fibre and hybrid composites were conducted by using thermogravimetric analyser (TGA) and differential scanning calorimeter (DSC). It was found that at high kenaf fibre content showed better thermal stability while the highest thermally stable was found in pure carbon fibre composite. The TG and DTG results showed that the amount of residue decreased in plain-designed hybrid composite compared to satin-designed hybrid composite. The DSC data revealed that the presence of woven kenaf increase decomposition temperature.

Keywords: hybrid composites, thermal analysis, kenaf, carbon fibre, epoxy

1. Introduction

Over the last decade, natural fibre is known as a reinforcement material in polymer composite, due to the environmental concerns by substituting synthetic or man-made fibres. Natural fibre reinforced polymer composite (NFRPC) is a composite material made up of a polymer matrix mixed with natural fibres, such as oil palm [1], jute [2, 3], flax [4], banana fibre [5], kenaf [6, 7] ramie [8]. These fibres are widely used as reinforcements in the NFRPC because they are low in density, good in mechanical properties, recyclable, and well in terms of strength per weight materials [9, 10]. Furthermore, natural fibres are favoured over synthetic fibres because they are abundant, renewable, and biodegradable.

Among these fibres, kenaf (*Hibiscus cannabinus*) is remarkable as one of the potential natural fibres that can be used in the bio-based composite production because of its price, properties, and fast to be grown [11-13]. Kenaf bast has good prospective as a reinforcement agent for natural fibre composite because it has long fibre with good mechanical properties and high strength that can be converted to a high performance composite [14, 15]. Compared to softwood fibres, bast fibre is slightly shorter (2.48 to 3.60 mm) and thinner that can increase the ability of bonding and strength development [16,17]. In addition, the slenderness ratio

(fibre length/fibre diameter) of kenaf bast fibres is comparable to those of softwood fibres [18] and bast fibre have low lignin content (14.7%) which attributed to the quality of bast fibres [16]. For the past several years, kenaf fibres have been shown to be suitable for fibre reinforced composite applications such as particleboard, medium density fibreboard (MDF), polymer matrix composite (PMC), pultruded products, non-woven materials, and woven materials.

However, the performance of the NFRPC is still not comparable with the synthetic polymeric composite. Therefore, hybridisation of synthetic fibre with natural fibres is suggested to produce biodegradable composite. Furthermore, hybridisation can overcome NFRRC disadvantages such as poor in fiber–matrix bonding adhesion [19, 20], higher water absorption and hydrophilic in nature [21, 22], lack in wettability [23,24], and low thermal properties [25]. Thus, hybridisation with synthetic fibre such as carbon fibre, glass fibre, aramic, and Kevlar is believed can improve the mechanical and thermal properties of the composites. Many researchers have been worked out to utilise natural fibre by hybridisation with synthetic fibres, such as kenaf/Kevlar [26], flax/glass fibre [27], jute/carbon fibre [28], sisal/carbon fibre [29], ramie/glass fibre [30], and flax/carbon fibres [31] composites.

Various studies have proved that the thermal properties of NFPC improved by hybridisation with synthetic fibre. A comprehensive review conducted by Ibrahim et al. [32], Madhusudhan and Keerthi [33] and Jawaid and Khalil [34] on hybrid composite has confirming that thermal properties of natural and synthetic fibre hybrid composites are better than those of the non-hybrid composite due to the higher thermal stability of synthetic fibres. Atiqah et al. [35] also found that the addition of glass fibre in sugar palm fibre reinforced with polyurethane showed better thermal properties by improved in thermogravimetric analysis (TGA) and dynamic mechanical analysis (DMA). A thermal properties of glass fibre hybridisation with sisal fibre reinforced polypropylene composites were carried out by Jarukumjorn and Suppakam [36] showed an improvement with addition of glass fibre. Thermal properties of sisal/glass hybrid polypropylene investigated by Nayak and Mohanty [37] observed in higher thermal stability in the hybrid composite. In addition, Nayak et al. [38] noticed that better thermal stability of the matrix polymer was obtained with the addition of bamboo and glass fibres, due to the hybridisation when using differential scanning calorimeter (DSC) and TGA.

In this study, investigation was carried out to study the thermal properties with the incorporation of woven kenaf and carbon fibre reinforced epoxy composite. The effect of carbon fibre on the thermal properties of woven kenaf reinforced epoxy composite was also observed. TGA and DSC were used to evaluate the thermal properties of the hybrid composite. Thermogravimetry was used to observe the thermal stability and degradation, while DSC was used to analyse the transition's temperature.

2. Materials and Methods

2.1. Materials

Epoxy resin was used as the polymer matrix and was supplied by Chemrex Corporation Sdn. Bhd. The woven kenaf was prepared manually using a hand loom. Carbon fibre was supplied by Spinteks Tekstil Ins. The properties of these material and the woven kenaf/carbon fibre hybrid composite fabrication comprehensively covered by Aisyah et al. [39]. In this study all types of composites: 100% carbon fibre and woven kenaf/carbon fibre hybrid composite were tested for their thermal properties. TGA and DSC analysis were carried out to further investigate the thermal properties of hybrid woven composites after the incorporation with carbon fibre.

2.2. Thermogravimetric analysis (TGA)

TGA was measured using a thermogravimetric analyser (TGA Q500 from TA Instruments) to observe changes in temperature and time in the controlled environment. The samples were heated from 25 to 800 °C at a heating rate of 10 °C/min in nitrogen gas flow rate of 50 mL/min. A sample of 8–10 mg of the materials was heated in the sample pan and the recorded data were displayed as TG (weight loss as a function of temperature) and as DTG (derivative thermogravimetric, weight loss rate as a function of temperature).

2.3. Differential Scanning Calorimeter (DSC)

DSC analysis was carried out to identify the melting point and the highest temperature limit of the composites using a DSC Q20 from TA Instruments. The samples were heated at a rate of 10 °C/min from 25–350 °C under nitrogen air. Nitrogen was used for efficient heat transfer and removal of volatiles from the sample. Each fibre sample was analysed separately and overlapped for comparison.

3. Results and Discussion

In order to study the effect of hybridisation of carbon fibre on the thermal stability of woven kenaf composites, TGA was conducted. The TG and DTG curves were used to obtain the onset of decomposition temperature (T_{on}), temperature at the decomposition peak (T_{max}), weight loss, and the fraction of material that is not volatile at 800 °C, denoted as residual. Meanwhile, DSC was used to characterise transitions, for instance, crystallisation and melting, with the function of fabric design and fabric density of woven kenaf in the woven kenaf/carbon fibre hybrid composite.

3.1. Thermogravimetric analysis (TGA) Analysis of Composites

The TG and DTG curves of the composite is represented in Figures 1 and 2, respectively. The T_{on} , T_{max} , weight loss, and residual at 800 °C are tabulated in Table 1. The weight loss percentage (Figure 1) shows there was a reduction in the weight loss as a function of temperature in the hybridisation of carbon fibre with woven kenaf,

The TG result of 100% carbon fibre in Figure 1 showed a single decomposition step with the highest T_{on} of 341 °C, compared to the woven kenaf/carbon fibre hybrid composite. From starting temperature of 25 °C to the temperature of 100 °C, the composite lost only 0.53% of the initial weight that resembles to the evaporation of solvent materials. It can be observed that, at temperature range of 250–500 °C, the recorded weight loss was about 37.38% due to the degradation and evaporation of carbon fibres and matrix. Then, the composite continued with a linear weight loss up to 800 °C, where the final residue was 54.14%, which indicated the total weight loss of only 44.25% from the initial weight. Also, only one main peak on the DTG curve (Figure 2) was observed corresponding to an apparent step of decomposition. The decomposition of carbon fibre epoxy composite around 300–500 °C was associated to the decomposition of an organic-based sizing compound on the carbon fibre [40, 41]. In addition, the thermal properties of carbon fibre composite and their hybrid was studied by Dhakal et al. [42], found that the carbon fibre composite have higher T_{on} compared to those of the hybridisation with flax fibre, i.e., 365 °C.

It was observed that the thermal degradation for all hybrid composites show a three steps of degradation processes, with a small noticeable step observed at the temperature below 100 °C, due to the evaporation of water molecules in the kenaf fibre. From the TG and DTG curves, the small step of weight loss at the temperature range of 30–100 °C only found in the hybrid composites, which represents the release of moisture content, due to the water evaporation as the presence of water in lignocellulosic fibres [42, 43]. On the other hand, the pure carbon fibre composites did not show weight loss at 100 °C temperature, which shows the absence of water molecule.

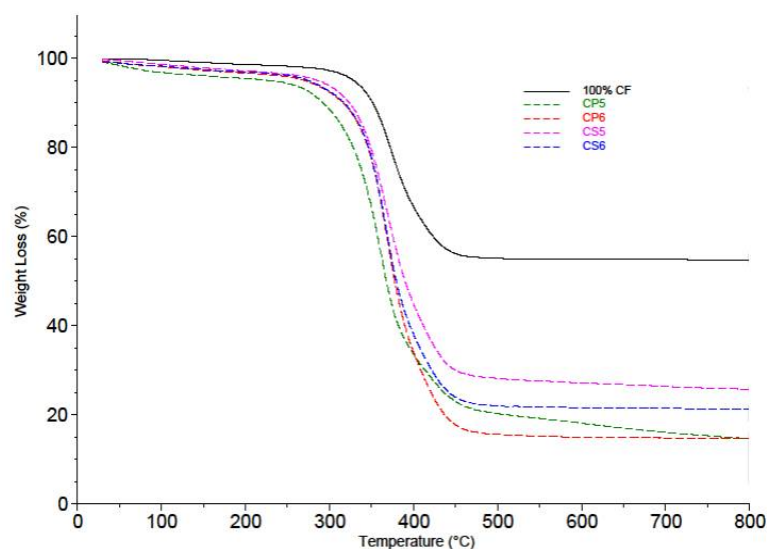


Figure 1. TGA curve on the effect of hybridization of woven kenaf in kenaf/carbon fibre hybrid woven composite on thermal properties.

Second step was corresponded to the weight loss of kenaf fibre, where a major decomposition occurred at a temperature range of 270–380 °C. This was due to the decomposition of hemicellulose, cellulose, and lignin of natural fibres [44] and depolymerisation of the matrix [45]. The decomposition of natural fibres starts with hemicellulose, followed by cellulose, lignin, and ash. The hemicellulose starts to decompose early, normally at temperature of 220 °C due to its chemical structure that consists of random amorphous structure with little strength, thus easily hydrolysed. In contrast, the decomposition of cellulose takes place at higher temperature (315–390 °C) than those of the hemicellulose because of its long polymer of glucose units and high crystalline nature, thus cellulose is relatively thermally stable [46]. The third degradation step is around 370–420 °C, which related to the degradation of carbon fibre chain rupture, where styrene was the primary product [47].

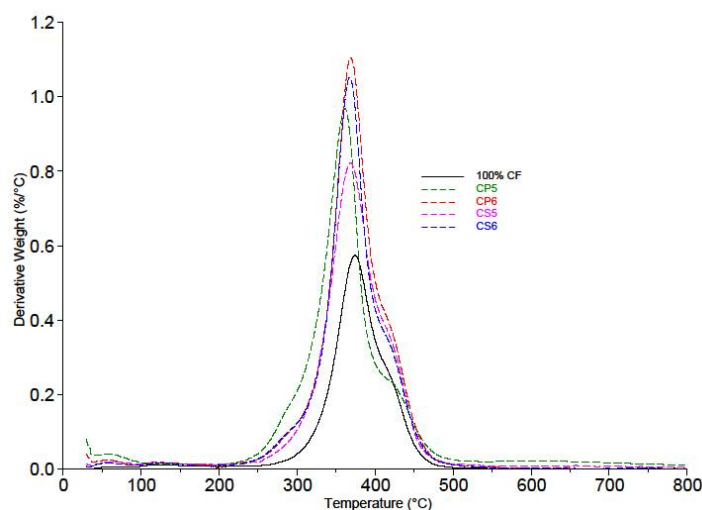


Figure 2. DTG curve on the effect of hybridization of woven kenaf in kenaf/carbon fibre hybrid woven composite on thermal properties.

From Table 1, the T_{on} of 100% carbon fibre was 341.41 °C, and the T_{on} of hybrid composites were around 331.27–336.11 °C. The incorporation of woven kenaf into carbon fibre composites decreased the T_{on} of neat carbon fibre because some portion of the synthetic fibre is replaced with less thermally stable material, i.e., kenaf fibres. Based on the TG curves, composite with woven kenaf with plain fabric (CP5 and CP6) show a move in the decomposition process towards a higher temperature level at 364–368 °C. The remarkable increase in thermal stability upon the weave design in the composite structure also can be related to the fibre and fibre interaction. It can be concluded that another factor that contributes to the higher thermal stability of polymeric composites is the better interaction between fibre and the matrix, which resulted in additional intermolecular bonding between fibre and matrix [48]. This can be support by the high tensile and impact properties of the plain composite in previous study [39].

Table 1 : Characteristic temperature at elevated weight loss

Sample Type	T_{on} (°C)	T_{max} (°C)	Weight Loss (wt.%)	Residue at 800°C (wt.%)
100% CF	341.41	381.34	44.25	54.14
CP5	331.27	363.58	81.90	13.09
CP6	336.11	368.91	82.04	14.44
CS5	331.62	364.13	71.97	20.92
CS6	334.88	368.02	75.89	20.79

Notes : CF = Carbon Fibre Composite, CP5 = Hybrid Plain Kenaf Composite (5 × 5 fabric count) with CF; CP6 = Hybrid Plain Kenaf Composite (6 × 6 fabric count) with CF; CS5 = Hybrid Satin Kenaf Composite (5 × 5 fabric count) with CF; CS6 = Hybrid Satin Kenaf Composite (6 × 6 fabric count) with CF

It is notable that, the T_{max} of hybrid composite increased from 363.58–368.91 °C and 364.13–368.02 °C by increasing the fabric density of woven kenaf from 5 × 5 to 6 × 6 in the plain and satin fabrics, respectively. The increase in T_{max} could be associated with the amount of cellulosic content in kenaf fibre which increased by increasing the fabric density, indicating that the kenaf fibre is able to improve the

thermal stability of woven kenaf/carbon fibre hybrid composite. This is in agreement with a study conducted by Atiqah et al. [35], who found that higher sugar palm fibre loading resulted in more thermally stable sugar palm/glass fibre polyurethane composite. As depicted in Figure 2, the DTG curve for 6×6 of fabric density (CP6 and CS6) was shifted to higher temperatures with the increasing amount of kenaf fibre. This phenomenon might be due to the higher amount of hydrogen bonds between cellulose chains in the 6×6 composites that can lead to more ordered and packed cellulose regions. This may further increase the thermal decomposition temperature of cellulose [49]. In addition, Nair et al. [48] mentioned that more ordered region decreases the mobility of cellulose chains will strain and weaken the existing hydrogen bonding thus increasing the thermal stability of composite.

From the result shown in Table 1, the highest final residue at 800°C was observed in 100% carbon fibre composite (54.14%), while the lowest residue was found in woven kenaf with plain fabric (CP5 and CP6). This is attributed to the resistance of carbon fibre to high temperature and better fibre matrix compatibility. The satin fabric hybrid composites (CS5 and CS6) showed residual of nearly 21%. Conversely, the residual left at 800°C for both plain fabric hybrid composites decreased significantly to approximately 13% and 14%, respectively. This was probably due to the resin-rich area found in the satin composite [39]. At the first stage, the hemicellulose, cellulose, and lignin in kenaf fibres were decomposed and formed charred layers that could prevent further degradation on the polymer matrix. According to Asim et al. [45], at higher temperature (300°C and above), epoxy starts to decompose. As the satin fabric contains more epoxy that are not infused into the inter-yarn due to the fabric structure [39], it resulted in more residual and forms a thicker layer between the heat source and polymeric material. This thicker layer residual results in the higher temperatures required for the composites decomposition and caused higher in residual content [50].

3.2. Differential scanning calorimetric (DSC) Analysis of Composites

The DSC analysis was carried out to further investigate the thermal behavior of hybrid composites after the incorporation with woven kenaf at the core layer of reinforced epoxy laminate composites composite. Figure 3 shows the DSC curves of hybrid composites containing CP5 (composite with plain fabric and 5×5 fabric count), CP6 (composite with plain fabric and 6×6 fabric count), CS5 (composite with satin fabric and 5×5 fabric count), CS6 (composite with satin fabric and 6×6 fabric count) and 100%CF (composite with pure carbon fibre). The curves obtained for the composite are showing an exothermic and endothermic behaviour, indicating the melting and crystallisation of composite samples. The thermal parameters of melting and crystallisation of the samples are summarised in Table 2.

From Figure 3, the addition of woven kenaf into carbon fibre hybrid composites affected the thermal behaviour significantly. It is clearly shown that the heat released from 100% CF composite was higher than that of the hybrid composite. From the curves, the curing temperature (T_c) and melting temperatures (T_m) values of the composites were strongly influenced by the incorporation of woven kenaf in the matrix polymer. The T_m of plain-designed hybrid composites and satin-designed hybrid composites decreased compared to the T_m of pure carbon fibre composite. This is probably due to the incorporation of woven kenaf reduces the total energy needed to be absorbed to break up the polymer chains of composites.

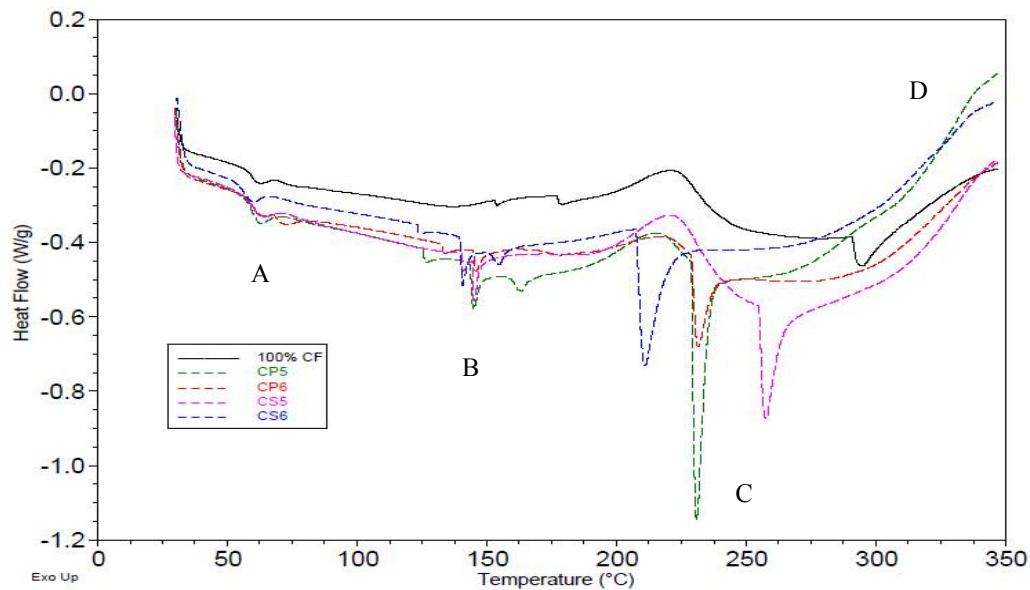


Figure 3. Thermal transitions DSC of the woven kenaf/carbon fibre hybrid composite vs. 100% carbon fibre epoxy composite.

The first drop of curvature was at around 58 °C (point A), displayed the glass transition temperature (T_g), indicated as the starting point for the energy required to change the molecular structure inside the composites from a low energy state i.e., solid or glassy state, to a higher energy state i.e., rubbery state. The thermal decomposition continued until a temperature of 122 °C (point B), and it is observed that there were multiple peaks for hybrid composites sample, but in the carbon fibre composite only two small peaks appeared. At this point, chains in the polymer might start to change the molecular structure to decompose from amorphous solid to crystalline solid by partially arranging their structures. As can be observed, the peak shifts to higher temperatures with the increase in the woven kenaf content (fabric count of 6×6). This finding is in line with a study conducted by Mofokeng et al. [51], who found that, as the fibre content of sisal fibre in poly(lactic acid) (PLA) and polypropylene (PP) composites increase, the crystallisation peak intensity decreased and it shifted to higher temperatures. They concluded that the sisal fibres act as the nucleation sites for the crystallisation of polymer, thus it restricts the mobility of the polymer chains. These sites may be particulates or fibres dispersed in the matrix, thus small crystals were formed around them [52]. In addition, kenaf fibres mainly consists of cellulose that represents the crystalline part of the materials, which could also result in increased crystallinity of the composites.

Table 2 : DSC results of pure carbon fibre and woven kenaf/carbon fibre hybrid composites

Sample Type	T_c (°C)	T_m (°C)
100% CF	178.2	294.4
CP5	145.0	257.4
CP6	140.6	211.4
CS5	145.9	231.3
CS6	144.7	230.7

Notes : CF = Carbon Fibre Composite, CP5 = Hybrid Plain Kenaf Composite (5×5 fabric count) with CF; CP6 = Hybrid Plain Kenaf Composite (6×6 fabric count) with CF; CS5 = Hybrid Satin

Kenaf Composite (5× 5 fabric count) with CF; CS5 = Hybrid Satin Kenaf Composite (6 × 6 fabric count) with CF.

Subsequently, there were very strong endothermic peaks for all hybrid composite known as T_m . At the temperature range of 211–261 °C (point C), there were endothermic peaks for hybrid composite samples, identified as thermal degradation due to the hemicellulose and cellulose degraded in kenaf fibres. It was reported that, chemical constituents in natural fibres starts degrading at a temperature around 200 °C [53, 54]. This peak is attributed to the dehydration, by splitting of hydroxyl groups of the cellulose molecule, resulting in the formation of water molecules and depolymerisation of cellulose, leading to the formation of flammable volatile products. The peak intensity of the T_m increased in the presence of higher kenaf fibre content (woven fabric of 6 × 6) probably due to more heat required to be absorbed to break up the polymer chains in composites. Additionally, hybrid composite with woven fabric of 5 × 5 showed better bonding properties between woven kenaf and carbon fibre with the matrix, thus crystallising at higher temperatures. For 100% carbon fibre, it can also be observed that the heat required to generate these peaks were higher than the hybrid samples (293 °C) indicating that this melting temperature of carbon fibre is contributed to the crystalline structure in carbon fibre through evaporation of the solvent during heating [55]. It is notably that the hybrid samples showed higher exothermic peak (point D) than the pure carbon fibre composite samples, indicating that the degradation of lignin and cellulosic matters from the kenaf fibres as lignin start to decompose at temperature of 340 °C and above [54, 56]. Thus, it can be stated that, the hybridisation of woven kenaf with carbon fibre in the composite structure become more thermally stable. The decomposition in 100% CF is probably due to the decomposition mechanism of epoxy resins through cyclisation of aliphatic chain ends [57].

4. Conclusions

The effects of hybridisation of carbon fibre with woven kenaf composites on thermal stability were examined. Although TG and DTG curves revealed that the thermal stability of carbon fibre composite was higher than that of the hybrid composite, the thermal stability of the hybrid composites improved upon the high content of kenaf fibre by using fabric density of 6 × 6. It was found that using the plain weave design of woven kenaf has improved the thermal stability than the satin design. The addition of carbon fibre in woven kenaf hybrid composites has improved the TGA properties of hybrid composites. The DSC results shows that the plain weave design and fabric count of 5 × 5 owned better interfacial adhesion between fibre and matrix. Thus, the hybrid composites are suitable for various applications that require at elevated temperatures. The DSC results discovered that the hybrid composite is more stable due to high decomposition temperature.

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