

Research Article

Thermal Properties of Woven Kenaf/Carbon Fibre-Reinforced Epoxy Hybrid Composite Panels

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The effects of carbon fibre hybridisation on the thermal properties of woven kenaf-reinforced epoxy composites were studied. Woven kenaf hybrid composites of different weave designs of plain and satin and fabric counts of 5×5 and 6×6 were manually prepared by a vacuum infusion technique. A composite made from 100% carbon fibre was served for a comparison purpose. Thermal properties of pure carbon fibre and hybrid composites were determined by using a thermogravimetric analyser (TGA) and differential scanning calorimeter (DSC). It was found that a hybrid composite with higher kenaf fibre content (fabric count 6×6) showed better thermal stability while the highest thermal stability was found in the pure carbon fibre composite. The TG and DTG results showed that the amount of residue decreased in the plain-designed hybrid composite compared to the satin-designed hybrid composite. The DSC data revealed that the presence of woven kenaf increased the decomposition temperature.

1. Introduction

Over the last decade, natural fibre is known as a reinforcement material in a polymer composite. Natural fibre acts as a substitution to synthetic or man-made fibres due to the environmental concerns raised by the latter. A 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], and ramie [8]. These fibres are widely used as reinforcements in the NFRPC due to the fact that they are low in density, good in mechanical properties, and recyclable and have excellent 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 one of the remarkable natural fibres that can be potentially

used in the biobased composite production because of its lower price, good properties, and fast-growing characteristic [11–13]. Kenaf bast has good prospective as a reinforcement agent for the 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, ranging from 2.48 to 3.6 mm, and thinner, which makes it has higher 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]. Apart from that, bast fibre has low lignin content (14.7%) which is favourable to its quality [16]. For the past several years, kenaf fibres have been proven suitable for fibre-reinforced composite applications such as particleboard, medium density fibreboard (MDF), polymer matrix composite (PMC), pultruded products, nonwoven materials, and woven materials.

However, the performance of the NFRPC is still not comparable with the synthetic polymeric composite. Therefore, hybridisation of natural fibres with synthetic fibre is recommended in order to decrease the moisture absorption and increase the mechanical properties of the composites. Hybridisation is able to overcome the disadvantages of NFRRC such as poor fibre-matrix bonding adhesion [19, 20], higher water absorption and hydrophilic nature [21, 22], inferior wettability [23, 24], and low thermal properties [25]. Thus, hybridisation with synthetic fibre such as carbon fibre, glass fibre, aramid fibre, and Kevlar is believed to be able to improve the mechanical and thermal properties of the composites. Various past investigations using hybridisation of natural and synthetic fibres have been carried out such as kenaf/Kevlar [26], flax/glass fibre [27], jute/carbon fibre [28], sisal/carbon fibre [29], ramie/glass fibre [30], and flax/carbon fibre [31] composites.

Several studies have proved that the thermal properties of NFRPC have been improved by hybridisation with synthetic fibre. A comprehensive review conducted by Ibrahim et al. [32], Madhusudhan and Keerthi Swaroop [33], and Jawaidd and Khalil [34] on the hybrid composite has confirmed that thermal properties of natural and synthetic fibre hybrid composites are better than those of the nonhybrid 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 polyurethane had better thermal properties as revealed by thermogravimetric analysis (TGA) and dynamic mechanical analysis (DMA). Thermal properties of glass/sisal fibre-reinforced polypropylene composites showed improvement with addition of glass fibre [36]. Better thermal stability has been observed in the hybrid composite of sisal/glass polypropylene [37]. In addition, Nayak et al. [38] also noticed that better thermal stability was attained with the addition of bamboo and glass fibres into the polymeric composite.

In this study, investigation was carried out to study the thermal properties of woven kenaf and carbon fibre-reinforced epoxy composite. The effect of carbon fibre addition 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

The woven kenaf fabric was prepared manually using a hand loom. Two weaving designs, namely, plain and satin, and two different fabric counts, namely, 5×5 and 6×6 (number of warp yarn \times number of weft yarn), were used in this study. Carbon fibre in a balanced woven fabric form (SPN.B 200.P-3K) was supplied by Spinteks Tekstil Ins. Specific properties of the carbon fibre are tabulated in Table 1. Epoxy EPIKOTE 240 resin (5300 mmol/kg epoxy group content, density of 1.12 g/cm^3) and hardener EPIKURE Curing Agent 309 were purchased from Chemrex Corporation Sdn. Bhd.

TABLE 1: Properties of carbon fibre supplied by Spinteks Tekstil Ins.

Properties	Carbon fibre
Weave pattern	Plain
Warp tow size	3K
Weft tow size	3K
Density	1.78 g/m^3
Thickness	0.20 mm
Tensile strength	3800 MPa
Tensile modulus	240 GPa
Strain	1.6%

TABLE 2: Composition of kenaf woven in the hybrid composite.

Composite code	Laminate sequence	Weave design	Woven kenaf	
			Fabric count	Volume fraction (%)
CP5	CF+PKF+CF	Plain	5×5	17.45
CP6	CF+PKF+CF	Plain	6×6	16.33
CS5	CF+SKF+CF	Satin	5×5	20.14
CS6	CF+SKF+CF	Satin	6×6	18.89

Note: CF: carbon fibre; PKF: Plain kenaf fabric; SKF: Satin kenaf fabric.

The manufacturing of the woven kenaf/carbon fibre hybrid composite was conducted in accordance with the procedures specified in the previous work [39]. The composite samples were prepared in a dimension of $300 \times 300 \text{ mm}$ by a hand lay-up method followed by the vacuum infusion process (VIP). The composite was prepared through VIP to efficiently pull the epoxy resin into the layer of woven kenaf and carbon fibre by removing the air from the system. In the preparation of composites, the layer of woven kenaf and carbon fibres was placed one by one by the hand lay-up method. Each composite consists of a single ply woven kenaf as the reinforcement at the centre and carbon fibres at the upper and lower layers as shown in Table 2. The epoxy resin is applied after placing each layer of fabrics. In this process, a pressure is applied to the laminated plies through a vacuum bagging film. The resin was then allowed to flow for a few minutes to ensure that the resin penetrated all the layers. Finally, the infused fabric composite was left to cure for 24 h at room temperature. In this study, five types of composites, namely, 100% carbon fibre, and woven kenaf/carbon fibre hybrid composite were tested for their thermal properties within the scope of this work. TGA and DSC analyses were carried out to further investigate the thermal properties of hybrid woven composites after the incorporation with carbon fibre.

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 the 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

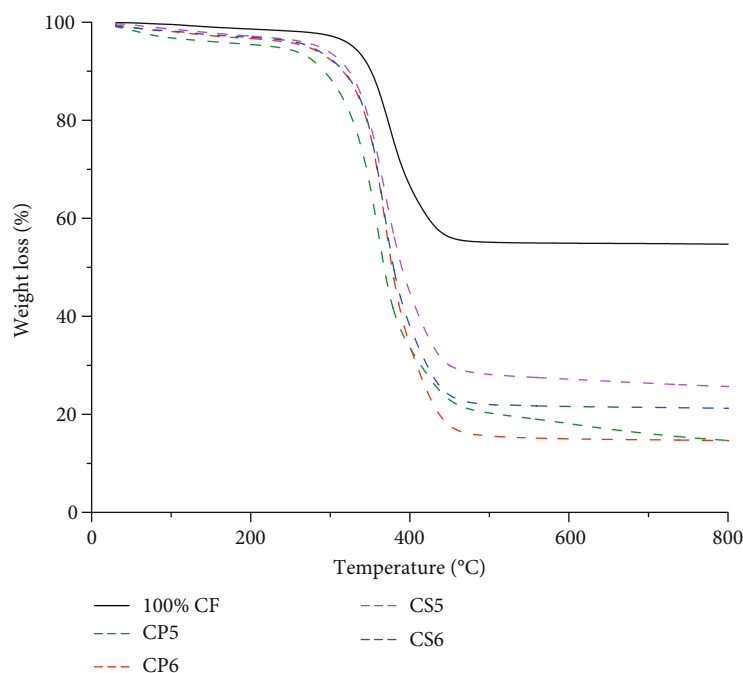


FIGURE 1: TGA curve on the effect of hybridization of woven kenaf in kenaf/carbon fibre hybrid woven composite on thermal properties.

(derivative thermogravimetric, weight loss rate as a function of temperature). DSC analysis was carried out to measure changes in heat flows associated with material transitions of the composites using a DSC Q20 from TA Instruments. A sample weight of 3–4 mg in an aluminium crucible with a pin hole was heated at a rate of 10°C/min from 25–350°C under nitrogen air. Three samples from each composition were analysed. 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.

4. Thermogravimetric Analysis (TGA)

The TG and DTG curves of the composite are presented in Figures 1 and 2, respectively. The T_{on} , T_{max} , weight loss, and residual at 800°C are tabulated in Table 3. The weight loss percentage are in Figure 1; 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 revealed a single decomposition step with the highest T_{on} of 341°C,

compared to the woven kenaf/carbon fibre hybrid composite. From the starting temperature of 25°C to the temperature of 100°C, the composite lost only 0.53% of the initial weight that resembles the evaporation of solvent materials. Mass loss of 37.38% over the temperature range of 250–500°C could be observed in 100% CF composite. The mass loss at this temperature range could be attributed to the decomposition of an organic-based sizing compound on the carbon fibre [40, 41]. 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 as illustrated (Figure 2) was observed corresponding to an apparent step of decomposition. The thermal properties of the carbon fibre composite and their hybrid were studied by Dhakal et al. [42]. The authors found that the carbon fibre composite has higher T_{on} of 365°C compared to those of the composites hybridise with flax fibre.

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

The second step 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]

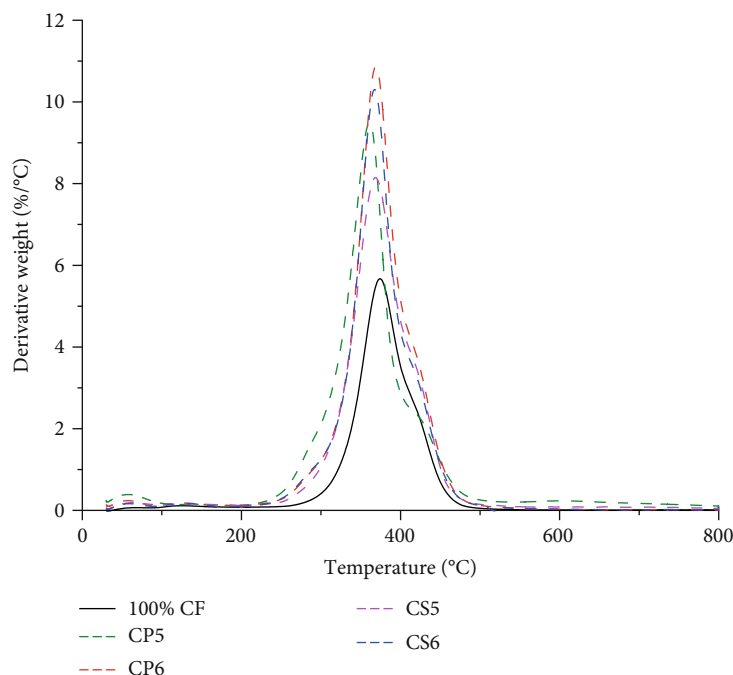


FIGURE 2: DTG curve on the effect of hybridization of woven kenaf in kenaf/carbon fibre hybrid woven composite on thermal properties.

TABLE 3: 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.

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 a random amorphous structure with little strength, thus easily hydrolysed. In contrast, the decomposition of cellulose takes place at a higher temperature (315–390°C) than those of the hemicellulose because of its long polymer of glucose units and high crystalline nature, making cellulose 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].

From Table 3, 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 addition 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, the composite with woven kenaf with plain fabrics including CP5 and CP6 shows a move in the decomposition process towards a higher temperature level at 364–368°C. The substantial increase in thermal stability upon the weave design in the composite structure also can be related to the fibre and fibre interaction. It appears that another factor which contributes to the higher thermal stability of polymeric composites is enhanced interaction between fibre and the matrix, resulting in additional intermolecular bonding between them [48]. This can be supported by the high tensile and impact properties of the plain composite in the previous study [39].

It is clear that, the T_{max} of the hybrid composite increased from 363.58 to 368.91°C and 364.13 to 368.02°C by increasing the fabric density of woven kenaf from 5×5 to 6×6 for both plain and satin fabrics. The increase in T_{max} could be associated with the amount of cellulosic content in kenaf fibre with higher fabric density. 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 fabric density (CP6 and CS6) shifted to higher temperatures. 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 a highly ordered region could reduce the mobility of cellulose chains and eventually strain and weaken the existing hydrogen bonding thus increasing the thermal stability of the composite.

Based on the results shown in Table 3, the highest final residue at 800°C was observed in the 100% carbon fibre

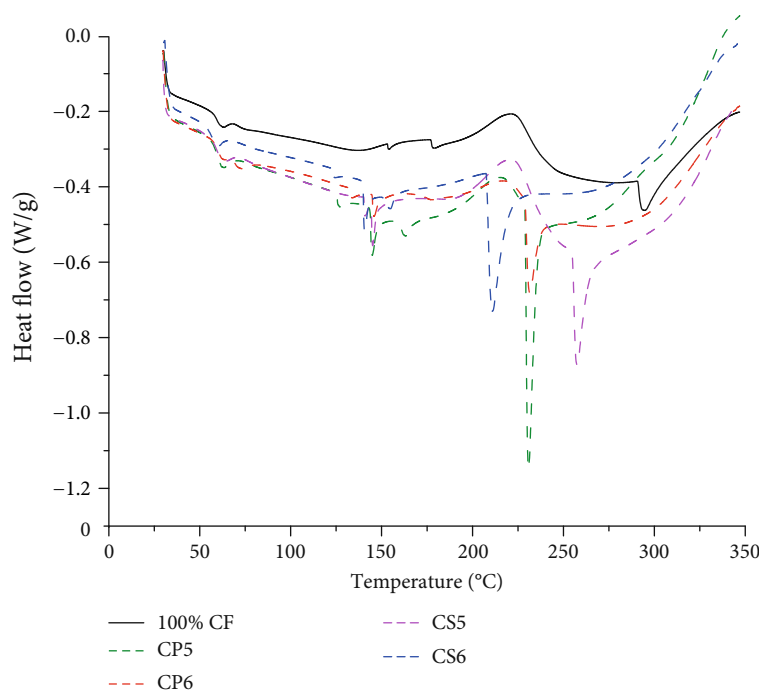


FIGURE 3: Thermal transitions DSC of the woven kenaf/carbon fibre hybrid composite vs. 100% carbon fibre epoxy composite.

composite, which was 54.14%. Meanwhile, the lowest residue was found in woven kenaf with plain fabric (CP5 and CP6). This could be 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%. 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 a higher temperature (300°C and above), epoxy starts to decompose. As the satin fabric contains more epoxy that is 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 composite decomposition and caused higher residual content [50].

5. Differential Scanning Calorimetric (DSC)

The DSC analysis was carried out to further investigate the thermal behaviour of the pure carbon fibre and hybrid composites. Figure 3 shows the DSC curves of hybrid composites 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 for all the composites show an exothermic and endothermic behaviour, indicating the melting and crystallisation of the

TABLE 4: 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

composite samples. The thermal parameters of melting and crystallisation of the samples are summarised in Table 4.

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 the 100% CF composite was higher than that of the hybrid composite. From the curves, the curing temperature (T_c) and melting temperature (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 the pure carbon fibre composite. This could be probably due to the incorporation of woven kenaf that reduces the total energy needed to be absorbed to break up the polymer chains of composites.

The first drop of curvature was at around 58°C (point A), displaying 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 composite samples, 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 was in line with a study conducted by Mofokeng et al. [51], who determined that, as the fibre content of sisal fibre in poly(lactic acid) (PLA) and polypropylene (PP) composites increased, 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 consist of cellulose that represents the crystalline part of the materials, which could also result in increased crystallinity of the composites.

Subsequently, there were very strong endothermic peaks for all the hybrid composite known as T_m . At the temperature range of $211\text{--}261^\circ\text{C}$ (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 start 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 T_m increased in the presence of higher kenaf fibre content (woven fabric of 6×6) probably due to the fact that more heat is required to be absorbed to break up the polymer chains in composites. Additionally, the 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 was higher than the hybrid samples (293°C) indicating that this melting temperature of carbon fibre contributed to the crystalline structure in carbon fibre through evaporation of the solvent during heating [55]. It is notable that the hybrid samples showed a 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 starts to decompose at the 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 becomes 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].

6. Conclusions

The effects of hybridisation of carbon and kenaf fibre on the thermal stability of woven kenaf/carbon fibre-reinforced

epoxy hybrid composite were examined. Although TG and DTG curves revealed that the thermal stability of the pure carbon fibre composite was higher than that of the hybrid composite, the thermal stability of the hybrid composites improved as higher content of kenaf fibre was used, i.e., fabric density of 6×6 . It was found that using the plain weave design of woven kenaf has improved the overall thermal stability of the samples compared to that of the satin design. The addition of carbon fibre in woven kenaf hybrid composites has improved the TGA properties of hybrid composites. The DSC results show 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 might be subjected to elevated temperatures. The DSC results discovered that the hybrid composite is more stable as shown by the high decomposition temperature.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that there is no conflict of interests regarding the publication of this paper.

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