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Physical and flammability properties of kenaf and pineapple leaf fibre hybrid composites

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Abstract. Pineapple leaf fibre (PALF) was hybridized with kenaf fibre in order to achieve superior physical and flammability performance. The mixing effects of kenaf fibre and pineapple leaf fibre in phenolic composites are evaluated at various fibre ratios and investigated various physical properties such as density, void content water absorption, thickness swelling and flammability. Pure kenaf fibre composite showed lowest void content, water absorption and thickness swelling while pineapple leaf fibre revealed the opposite trend. After adding pineapple leaf fibre in kenaf composites, it absorbed more moisture and developed more void contents. Density of PALF composite was lower and kenaf fibre composite was highest but PALF/kenaf hybrid composite increased density with higher PALF. The flammability of hybrid composites were analysed by using UL-94 test method. Vertical and horizontal UL-94 test conducted to analyse fire resistance of composites from different angles. Phenolic resin is itself a fire resistant polymer and the percentage of polymer in all samples are fixed though ratios of both fibres pineapple leaf fibre and kenaf were change to analyse fire resistant of fibres and compatibility with polymers. The 70 % pineapple leaf fibre and 30 % kenaf fibre hybrid composites were very affective to improve the flammability of PALF/KF hybrid composites.

1. Introduction

As far as the environment is the concern, renewable resources of materials are the best alternative material of good economic potential and protection of nature, since, the renewable resources are characterized as biodegradable and recyclable, lignocellulosic fibres belong to this category [1]. Nowadays, natural fibres as reinforced material have been increased considerably due its low price compared to synthetic fibers [2]. Waste management of lignocellulosic and renewable materials and poor mechanical strength and thermal stability of polymers can be improved by developing natural fibres reinforced polymer composites [3-5]. Some plants fibres have potential to be applied in industries as raw materials such as pineapple, kenaf, coir, abaca, sisal, cotton, jute, bamboo, banana, Palmyra, talipot, hemp, and flex [6-7]. Pineapple leaf fibre (PALF) is one of the waste materials in agriculture sector, which is widely grown in Malaysia as well as Asia. After banana and citrus, pineapple (*Ananas comosus*) is one of the most essential tropical fruits in the world [8]. Natural fibres influenced mechanical strength due to chemical composition such as to cellulose, hemicellulose and lignin and its orientation and some other factors such as type of plant, environmental factors during growth as well as extraction methods used [9]. Usage of natural fibre can replace synthetic fibres to minimise health hazards, low density and high flexibility [10].

Recently, polymer hybrid composites have become the attraction into production of mechanical stable and renewable products [11]. Polymer hybrid composites are formed by incorporating two and more reinforcing materials into a polymeric matrix via-versa. The hybridization of two natural fibers or a synthetic fibre with a natural one can improve the physical, mechanical and thermal properties of composites [12]. The reinforcing materials provide a synergetic effect called a “hybrid effect” that determines new or enhanced properties [13]. The hybrid composites based on fiber fillers have become extremely important materials [14]. It is extensively used in aerospace, automotive, and construction industry etc. The main reason of using fibres as reinforced materials for fabricating the hybrid composites are their mechanical strength, lightweight, durability and competitiveness and eco-friendly [15].

Natural fibre reinforced polymer composites are sensitive to water absorption, humidity, dimension stability. Water absorption affects the mechanical strength and compatibility between fibre and matrix and leading towards poor stress transfer efficiencies from matrix to reinforcement [16]. Mechanism of water absorption in fibre composites is based on three ways; first method is diffusion method [17], water molecules flow into micro-cracks of polymer chains. Second method is capillary flow[18]; water flow along with the interface if interfacial bonding of fibre and matrix is weak. Last mechanism is moisture content; storage of water in micro-cracks present in both polymer and natural fibre [19]. Study of water absorption in natural fibre composite is necessary for different applications such as outdoor component, waste water treatment, packaging and building industry. In natural fibre reinforcement, fibre absorb water due to many reason such as temperature, amount of fibre, orientation of reinforcement, fibre characteristics, area of exposed surfaces, diffusivity, and surface protection [20].

Origin of most of the polymers are petroleum-based, due to this they are highly flammable/combustible [21]. Phenolic resins have been deeply studied for commercial purpose and still there is no substitute of phenolic in stability and heat-resistance quality [22]. Phenolic resin is very brittle in nature and due to this mechanical strength is very low and it is not biodegradable. Many natural fibres have been used as reinforced material to enhance the mechanical properties. Pineapple leaf fiber (PALF) was selected as reinforcement materials for phenolic resin due to the following reasons. First, PALF has good tensile strength (400–1600 MPa) and modulus (59 GPa) due to its high cellulose content 70–82% and high degree of crystallinity [8]. Second, since pineapple can be cultivated throughout the South-East Asia, it is possible for significant domestic production of PALF [23]. Third, PALF has already been used as a reinforcement to strengthen LDPE, rubber, thermosetting polyester, polyhydroxylbutyrate (PHB) and polyester amide [24]. Kenaf is comparatively very cheap and commercially available natural fibre to use as reinforcing material. Kenaf fibre (KF) showed very good compatibility with the different type of polymers such as epoxy and phenolic resin [25].

In this research, we have fabricated PALF and KF hybrid composites by using hand layup technique. It is assumed that kenaf will reduce water absorption due to low cellulosic content good compatibility with polymers. Phenolic resin will help to improve fire resistant properties of composites, where, PALF will improve compatibility due to its physical properties.

2. Materials and method

Novolac type Phenolic formaldehyde resin (Grade PH-4055) was supplied by Chemovate Girinagar, Banglore, India. Pineapple leaf fibres (PALF) (*Ananas comosus*) were harvested in Indonesia and kenaf were harvested from Malaysia and retted by immersion into water having 0.5% wt Urea. The chemical compositions of PALF and kenaf were collected from previous study [1] were examined in MARDI, Malaysia. The data of mechanical properties and fibre diameter were collected from the previous studies [23] shown in Table 1.

Table 1. Chemical composition, physical and mechanical properties of PALF and KF [1,23].

Properties	PALF	KF
Tensile strength (MPa)	290.61	282.60

Tensile modulus (GPa)	5.83	7.13
Cellulose content (%)	70.51	66.89
Hemicellulose content (%)	14.21	14.98
Lignin content (%)	2.93	6.85
Fibre diameter (μm)	78.80	83.50

2.1. *Fabrication of composites*
PALF and KF used as filler in the fabrication of composites. PALF and KF were ground into 0.8–1 mm by using grinding machine; fibres maintain 6–8 % moisture content. PALF and KF reinforced Phenolic composites were fabricated by using hand lay-up technique in 15 × 15 × 3 mm stainless metal plate. The 3 mm stainless steel plate was placed into hydraulic pressure hot press at 160 °C temperature. The stainless steel plate was removed from the press after 8 min and kept for cooling to room temperature, and then samples were cut for testing according to ASTM standard. The ratio of fibres and matrix of composites are tabulated in Table 2.

Table 2. Formulation of composites and hybrid composites

Type of composites and hybrid composites	PF (wt%)	PALF(wt%)	KF(wt%)
50% PALF	50	50	0
3P7K	50	15	35
1P1K	50	25	25
7P3K	50	35	15
50% KF	50	0	50

2.2. *Density*
Density was measured by using ASTM D 1895 standard. The density of the samples was calculated by using following equation

$$\text{Density } (g/cm^3) = m/v \quad (1)$$

Where, m represents mass of fibre/matrix and v represents volume of fibre/matrix.

2.3. *Void content*
For determination of voids in PALF and KF composites and its hybrid composites, ASTM-D-2734-70 [26] method was used. The void content was determined from the theoretical and experimental density of the composites through following equation:

$$\text{Void content} = 100/\,(R/D + r/d) \quad (2)$$

Where, R is the weight % of the resin in the composite;
D is the density of the resin matrix;
r is the weight % of the reinforcement in the composite;
d is the density of the reinforcement.

2.4. *Water Absorption Test*
Natural fibre composites sample were immersed into distilled water at room temperature. Water absorption was calculated by the equation 3 [27] for various period of time. All the samples were weighed before and after the soaking samples and recorded carefully. At the three intervals, water absorption was examined such as 3 day, 5 days and 7 days. After taking out samples from water, all the samples were wipe out to remove excess amount of water on the surfaces. The percentage equilibrium

water absorption was calculated as an average value of several measurements. The percentage of water absorption was calculated from Equation (3) using ASTM D570,

$$\text{Water absorption (\%)} = [(W_n\text{-}W_d)/W_d]X100 \quad (3)$$

where, W_n is the weight of composites samples after immersion and
W_d is the weight of the composite samples before immersion

2.5. Thickness Swelling

Ten samples of pure phenolic and each different PALF fibre loadings composites were prepared for the experiment of thickness welling. Thickness swelling was calculated according to ASTM D570 by using Equation (4)[27]. The thickness swelling experiment was conducted by measuring the swelling of the each sample. Measurement of the composite was taken before and after the soaking of composites. Samples were measured three time, first measurement was taken on third day of immersion second was fifth days of immersion and last was seven days of immersion.

$$\text{Thickness Swelling (\%)} = [(T_1\text{- } T_0)/T_0]X100 \quad (4)$$

where, T_1 is the thickness after soaking and
T_0 is the thickness before soaking.

2.6. Flammability test

Flammability of the untreated and treated composites and hybrid composites were studied by a horizontal UL-94 test and a vertical UL-94 test according to ASTM D635 and ASTM D3801, respectively. For the horizontal UL-94 test, the specimen was fixed horizontally and a flame was applied to one end of the specimen. The time for the flame travel from 25 mm (from the end) to 100 mm was recorded. Then, burning rates of the composites were calculated as follows:

$$V = 60L/t \quad (5)$$

Where, V is the burning rates (mm/min),
L is burned length (mm), and
t is the time of burning (sec).

The UL-94 vertical test was carried by using a test specimen bar 125 mm long, 13 mm wide, and 3 mm thick. Specimen bars were suspended vertically over surgical cotton, and ignited with a Bunsen burner. The flammability types were classified from V-0 toV-2. The lower end of the specimen was exposed to the flame for 10 s and then removed. The time was recorded.

The specimen would be then assigned to class UL-94 V-0 if flaming time is less than 10 sec. The lowest flame retardancy rating is V-2 if flaming time is less than 50s and cotton indicator ignited by flaming particles or drops. V-1 is the intermediate class of V-0 and V-2, the flaming time should not exceed 30s and flaming drops could not ignite the surgical cotton below the specimen. The flammable materials which cannot be classified according to this method are marked with code NC (no classification, complete combustion of the specimen).

3. Results and discussion

3.1 Density

The theoretical and experimental densities of PALF and KF composites and their hybrid composites were calculated shown Table 1. The experimental density of PALF composites is lower (1.07 g/cm^3) than theoretical value (1.3 g/cm^3). However, the experimental density of KF composites is lower (1.25 g/cm^3) than theoretical value (1.27 g/cm^3) though this difference is very less in comparison of PALF composites. After addition of PALF at different fibre loading in KF composites, experimental densities reduced with inconsistency and at the same time the theoretical densities increased. Among the all hybrid composites, the 7P3K hybrid composites showed highest theoretical as well as experimental densities. The difference in theoretical and experimental densities are due to air trap in composites during manufacturing process [28].

3.2 Void content

The density of the both, measured and theoretical value of PALF and KF composites and their hybrid composites are given in Table 3. From the table, it is clearly seen that the specific gravity of PALF and KF composites and their hybrid composites are vary in different fibre fractions composites, which indicated the contribution of fibres in density of composites. The void contents is the difference of theoretical and practical density of substance, the most common cause of voids is the incapability of the matrix to displace all the air which is trapped within the woven or chopped fibres as it passes through the matrix impregnation. The void content (%) of PALF and KF composites and their hybrid composite samples are presented in Table 3. The PALF composite showed 12.50 % void content, however, KF composites revealed very low void content (2.36 %). It is also observed that after adding PALF in to KF composites, the percentage of void content increased. PALF was added up to 70 % of total fibre loading and the void content also increased up to 11.11 %. The increment in void content is may be due to incompatibility between the phenolic resin and PALF and high percentage of fibre fraction were inadequate to mix with the polymer properly.

Table 3. Density and void content of PALF and KF-Phenolic composites and their hybrid composites

Weight Fraction of fibres (%)	Theoretical density (g/cm^3)	Measured density (g/cm^3)	Void content (%)
50% PALF	1.4	1.22	12.5
3P7K	1.3	1.13	13.07
1P1K	1.32	1.18	10.22
7P3K	1.35	1.2	11.11
50% KF	1.27	1.24	2.36

According to researchers [29-30], incomplete wetting out of the fibres by the matrix would lead to the formation of voids. During impregnation of fibres into the matrix or during manufacturing of fibre reinforced composites, the trapped air or other volatiles exist in the composites [31]. KF composites showed very less void content percentage among all composites and hybrid composites, it showed KF has very good interfacial bonding with Phenolic matrix and have very less hollow space. The reduction in void content is evidence of improvement in mechanical strength because the presence of void content in the composites significantly reduces the mechanical and physical properties of the composites [32].

3.3 Water absorption

Water absorption (WA) percentage of PALF and KF composites and PALF/KF hybrid composites were shown in Fig.1. Among the all types of composites, KF composite showed lowest WA percentage and the deference in WA percentage was very high. However, WA of rest of the composites and hybrid composites were very close to each other. PALF composites revealed lower WA than all hybrid composites though the difference was very less. Among the all hybrid composites, 3P7K and 1P1K hybrid composites showed the highest WA. However 7P3K hybrid composites revealed its WA in the middle of highest WA and PALF composites shown in fig.1. From the results, KF composites showed lowest WA due to lower content of cellulose and PALF composites should show highest WA due to higher content of cellulose.

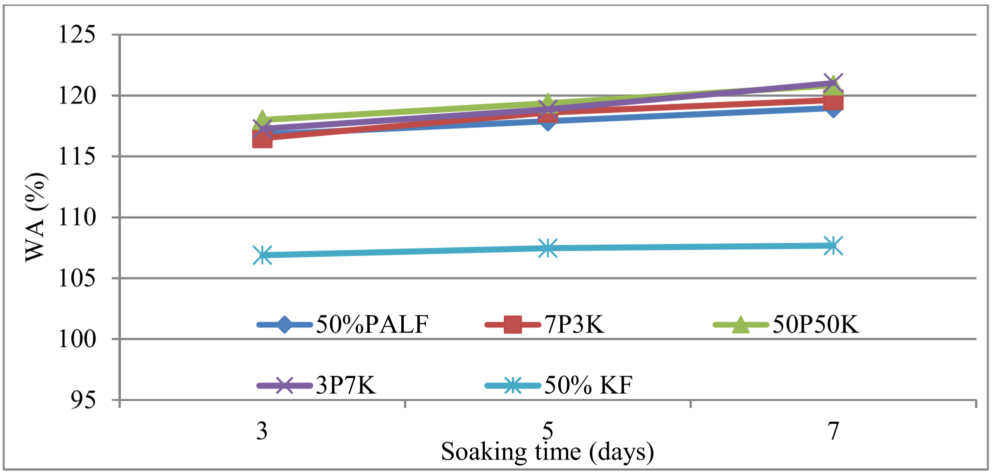

Figure 1. Water absorption of PALF and KF hybrid composites.

The expected results were to reduce the WA by adding KF in PALF composites; in the contrary the results showed that hybrid composites absorb more water content. By this research, it can be assume that in despite of having comparatively less quantity of cellulosic content, WA is increasing due to high void contents. The water molecules easily enter into the composites through such pores and these molecules are attracted by hydroxyl groups of natural fibres causing a rise of water absorption. This hydroxyl groups present in cellulose and hemicelluloses provide easy access to water in to composites. Besides that, there are many other factors such as porosity, void content, lumen size, and fibre-matrix adhesion also effect water absorption behaviour of the composites [33]. The inter-fibrillar spaces of cellulosic structure of natural fibres contain water molecules that crack and micro voids at the surface of composites [34]. The immersed composites into the water for long time, absorb water molecules through capillary action and fills the voids and cracks in the composites [27,35].

3.4 Thickness swelling

The natural fibre composites are highly hydrophilic in nature due to the presence of hydroxyl groups, which invite water molecules through hydrogen bonding [34]. These water molecules are the cause to build the moisture into cell walls and fibre/matrix interface. This is one of the main reason of dimensional changes of natural fibre reinforced polymer composites especially thickness and linear expansion due to reversible and irreversible swelling of the composites[36,37]. The Fig. 2 showed the TS of PALF and KF composites and their hybrid composites. KF composites showed very less impact of moisture on TS, with the immersing period the moisture swelling was increased.

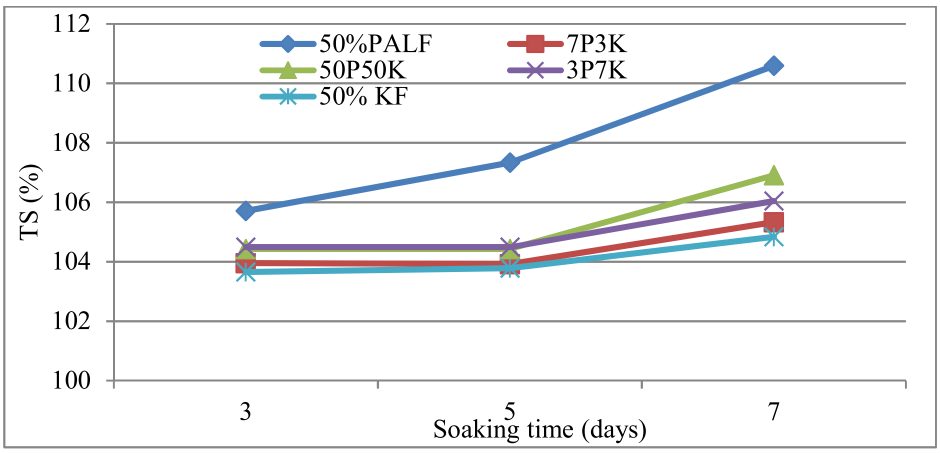

Figure 2. Thickness swelling of PALF and KF hybrid composites.

The hybrid composites 3P7K and 7P3K showed the higher TS values than KF composites but the pattern in dimensional changes are same affected by soaking periods. The hybrid composite 1P1K was showed more affected by last soaking period. The PALF composites showed highly prone to TS over the all soaking period of time. From these results, it is assumed that PALF has higher quantity of cellulosic materials and hydroxyl groups than KF, which absorb moisture content without hindrance. However, KF composite and KF hybrid composites showed less TS may due to have good interfacial bonding, less cracks and voids [28]. As can be seen from Fig. 2, the thickness swelling values of composites slightly increased with the increasing water exposure time. The swelling of the cell wall of fibres develop stress on the surrounding matrix and cause micro cracking, which reduce the mechanical properties of composite. As a consequence, the fiber–matrix adhesion is weak and the dimensional stability of composites particularly for outdoor applications will be greatly affected [34].

3.5 Under writer laboratory (UL-94)

The vertical and horizontal UL-94 tests were conducted to quantify and rank the flame retardancy of the composites. The results of the UL-94 tests for the PALF and KF composites and their hybrid composites are presented in Table 4. In Vertical UL-94, the flame retardancy of KF composite was very good and achieved V-0 grade because their combustion times were less than 10 seconds. After the addition of 30%, 50% and 70% PALF, the fire retardancy was reduced, and were considered in V-1 grade due to their combustion times were less than 30 seconds. On the other hand, PALF composites showed lower fire retardancy than KF composites and the combustion time was less than 13sec, considered in V-1 grade. No dripping was observed for any of the samples. Overall burning rate was low due to Phenolic matrix because of chemical structure of phenol resin is deformed and forms a layer of char on the surface of composites that protect the surface from heat [38].

Table 4. Horizontal and vertical burning rate of PALF and KF composites and PALF/KF hybrid composites

Sample	**Vertical UL-94**			**Horizontal UL-94** (mm/min)
	Flaming drops	**Cotton ignited**	**Classifications**	
50% PALF	No	No	V-1	15.71
7P3K	No	No	V-1	18
1P1K	No	No	V-1	17
3P7K	No	No	V-1	15.56
50% KF	No	No	V-0	13.6

Horizontal UL-94 showed the combustion rate of KF composite lowest among all composites however after addition of PALF into KF composites, the combustion rate was increased shown in Table 4 Burning rate of 7P3K was highest it may be due to incompatibility of both fibres with matrix that exposed the uncovered cellulosic content of fibre to the fire.

4. Conclusion

- PALF composites showed lowest density and higher void content. After adding KF, density of hybrid composite increased and void content decreased.
- Water absorption of PALF composites was higher and after adding KF it did not show significant affect. Alone KF composites showed very low water absorption.
- Thickness swelling of PALF composites was highest and KF composite was lowest. After hybridisation of PALF and KF, thickness swelling was reduced.
- All hybrid composites showed good fire resistant properties. In vertical UL-94, KF composites showed better property. However, in horizontal UL-94, PALF composite showed slower burning rate.

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