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Influence of Kenaf Form and Loading on the Properties of Kenaf-Filled Polypropylene/Waste Tire Dust Composites: A Comparison Study

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ABSTRACT: Kenaf (KNF)-filled polypropylene/waste tire dust (PP/WTD) composites containing different KNF loadings (0, 5, 10, 15, and 20 parts per hundred parts of resin (phr)) were prepared using a Thermo Haake Polydrive internal mixer. The influence of the KNF form (KNF short fiber (KNFs) and KNF powder (KNFp)) at different KNF loadings on properties of the composites was studied. Results showed that with increasing KNF loading, the stabilization torque, tensile modulus, water absorption, and thermal properties increased for both KNFp- and KNFs-filled PP/WTD composites. However, the tensile strength and elongation at break decreased by 29.2% and 53.9%, respectively, for KNFp-filled PP/WTD composites, whereas KNFs-filled PP/WTD composites showed a decrement of 24.5% and 63.5%, respectively. The stabilization torque, tensile strength, and tensile modulus increased by 22.4%, 6.7%, and 2.6%, respectively, for KNFs-filled PP/WTD composites at 20 phr KNF loading. The scanning electron microscopy morphological studies on the tensile fractured surfaces revealed poor adhesion between KNFp and PP/WTD matrices as compared to KNFs and PP/WTD matrices. © 2014 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2014**, *131*, 40877.

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INTRODUCTION

Thermoplastic elastomers (TPEs) are unique as they belong to a class of material that combines the properties of both thermoplastics and elastomers. In addition, they can be subjected to injection molding or extrusion processes (just like thermoplastics), and no vulcanization is required.[1] TPEs give better material utilization as the scraps and rejects can be recycled.[1,2] The continuous worldwide accumulation of waste rubber has caused attention over threats to public health and the environment.[3] Recycling is a way of reducing the increasing amount of waste rubber, thus lessening the threat to public health and the environment.[4] Numerous studies on waste rubber have been reported. For instance, Awang and Ismail[3] reported that the addition of dicumyl peroxide and *N*,*N*′-*m*-phenylenebismaleimide imparted noticeable improvement in the overall properties of polypropylene/waste tire dust (PP/WTD) blend. Supri et al.[5] reported that the tensile properties, thermal stability, swelling resistance, and morphology of low-density polyethylene/tyre dust increased with the addition of phthalic anhydride.

The use of various natural fibers in polymer composites has been growing rapidly in recent years, for example, wood, jute, hemp, kenaf (KNF), rattan, etc.[6–10] Natural fibers offer numerous benefits such as low cost, lightweight, abundance, harmless to human health and the environment, less abrasive to machines, and biodegradablity.[6–8] Their nontoxic property allows them to replace glass fibers or other hazardous fibers in developing eco-friendly composites.[9]

KNF is a low cost, renewable, and quick growing annual plant, which is used extensively as a filler in polymer composites.[7,8,11–15] For instance, Ismail et al.[12] used KNF core fiber as a filler in high-density polyethylene/soya powder composites and found that the tensile modulus, flexural strength, and flexural modulus of the composites increased with the addition of KNF core. Recently, the Malaysian government has promoted the development of KNF as a new industrial fiber crop, where incentives have been offered to farmers planting KNF.[16] Consequently, the price of KNF will become less, and the plant will be abundantly available. These are the reasons that why KNF was chosen as the filler for this study, and where the successful utilization

Table I. Compositions of KNF-Filled PP/WTD Composites

Materials	Compositions (phr)				
PP	70	70	70	70	70
WTD	30	30	30	30	30
KNFp or KNFs	0	5	10	15	20

of KNF will ensure inexpensive and more environmentally friendly final composites.

Several researchers have reported on the use of natural fillers in TPE composites.[17,18] However, as far as TPEs are concerned, there are a limited number of studies that have reported the use of KNF fiber as a filler in PP/WTD composites.

The purpose of this study was to study the effect of different KNF forms on the stabilization torque, tensile properties, water uptake, and thermal properties of KNF-filled PP/WTD composites as a function of KNF loading. Morphological studies on the tensile-fractured surfaces of the composites were also carried out.

EXPERIMENTAL

Materials

PP homopolymer was supplied by Titan Pro Polymers (M) Sdn. Bhd., Johor, Malaysia (Grade 6331), with a melt flow index of 14 g/10 min at 230°C and a density of 0.9 g/cm^3. WTD was obtained from Mega Makmur Saintifik Sdn. Bhd., Malaysia, with particle size ranging from 0.2 to 150 μm and specific surface area of 0.089 m^2/g. KNF was obtained from Lembaga Kenaf & Tembakau Negara, Kelantan, Malaysia.

Sample Preparation

Preparation of KNF Powder and KNF Short Fiber. KNF was ground into powder using a mini grinder from Rong Tsong Precision Technology Co. (Product Id: RT-34). Then, KNF was sieved using Endecotts sieve to obtain KNF powder (KNFp) with particle size of ≤75 μm. On the other hand, KNF was manually cut to obtain KNF short fiber (KNFs) with an average length and aspect ratio of 6 mm and 135, respectively.

Preparation of Composites. Compositions of the KNF-filled PP/WTD composites are shown in Table I. The melt-mixing of KNF-filled PP/WTD composites was carried out in a Thermo Haake Polydrive R 600/610 internal mixer at a temperature of 180°C, rotor speed of 50 rpm, and a constant mixing time of 10 min. The compounds were then compression molded using an electrically heated hydraulic press (GoTech Testing Machine Model KT-7014 A) to obtain 1 mm composite sheets. Compression molding was carried out at 180°C with 6 min of preheating, 4 min of compression, and 2 min of cooling. Dumbbell-shaped samples were then punched out using a Wallace die cutter from the 1-mm molded composite sheets.

Characterizations

Tensile test was carried out in accordance to ASTM D412 by using Instron 3366 Universal Testing Machine with crosshead speed of 5 mm/min and a constant gauge length of 50 mm. The tensile strength, tensile modulus, and elongation at break of each composite were measured.

Studies of the tensile fractured surfaces of the composites were carried out using a Leo Supra-35VP field emission scanning electron microscope. The specimens were mounted on the aluminum stubs and sputter coated with a thin layer of gold to avoid electrostatic charging and poor resolution during examination.

The water uptake test was carried out in accordance to ASTM D570. The specimens were dried in oven for 24 h at 50°C until a constant weight was obtained prior to immersion in distilled water. Percentage of water uptake was calculated based on eq. (1).

$$W_t(\%) = \frac{W_2 - W_1}{W_1} \times 100 \quad (1)$$

where W_t is the total water uptake by the specimen, whereas W_1 and W_2 are the weight of specimen before and after immersion in distilled water, respectively.

Thermogravimetric analysis of the composites was carried out using a Perkin Elmer Pyris 6 thermogravimetric analyzer, at the temperature range of 30°C–600°C under nitrogen flow of 50 mL/min and a heating rate of 20°C/min.

RESULTS AND DISCUSSION

Processing Torque

Processing torque can be used as an indicator of the composite viscosity and also to reveal the processability of the composites.[19] Higher processing energy is required with the increase in the viscosity, which indicates that the processing difficulty is increased.[20] Figure 1 shows the stabilization torque at the end of mixing time of 10 min of KNFp-filled and KNFs-filled PP/WTD composites at different KNF loadings. The stabilization torque of both composites increased with increasing KNF loadings. Viscosity of the composites increased with increasing KNF loadings due to the reduced free volume in between the PP/WTD chains. Thus, higher processing energy was required to disperse higher KNF loadings within PP/WTD matrices. Ismail et al.[20] also reported a similar manner where higher processing energy was required due to increase in viscosity of PP/recycled acrylonitrile–butadiene rubber/rice husk powder composite as the content of rice husk powder increased. This result was

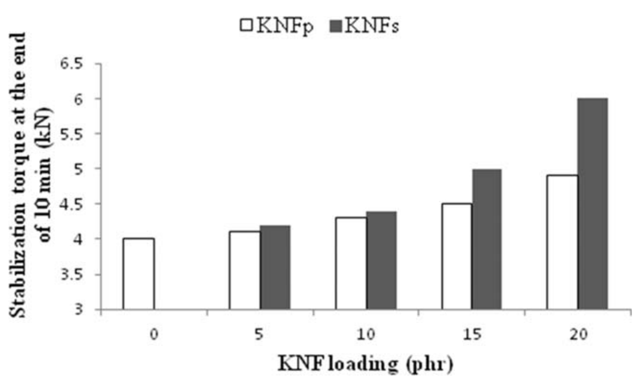

Figure 1. Stabilization torque of KNFp-filled and KNFs-filled PP/WTD composites at various KNF loadings.

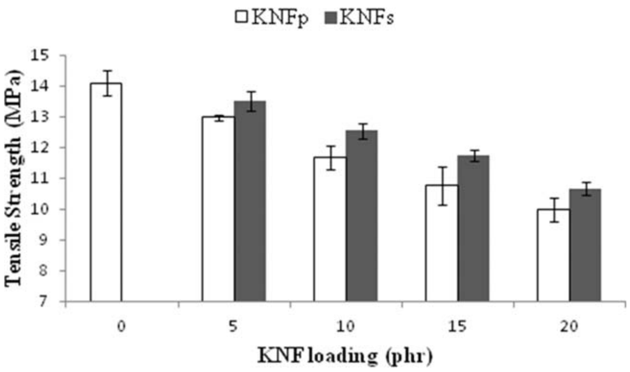

Figure 2. Tensile strength of KNFp-filled and KNFs-filled PP/WTD composites at various KNF loadings.

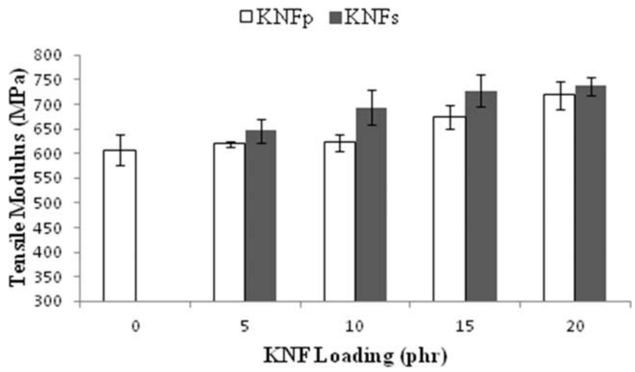

Figure 3. Tensile modulus of KNFp-filled and KNFs-filled PP/WTD composites at various KNF loadings.

expected, as the chain mobility of polymer macromolecules was reduced, due to the increasing filler loading into the polymer matrix that also contributed to a higher resistance of the material to flow.[2]

At similar KNF loadings, KNFs-filled PP/WTD composites exhibited a higher stabilization torque compared with the KNFp-filled PP/WTD composites. KNFp was found easier to mix with PP/WTD matrices compared with KNFs. This was attributed to the larger aspect ratio (L/D) of KNFs as compared with KNFp, which requires higher processing energy for mixing. This result is in well agreement with the previous findings by Migneault et al.,[21] where the mechanical energy and stabilized torque of wood/high-density polyethylene composites significantly increased with increasing wood fiber length.

Tensile Properties

Figure 2 shows the tensile strength of KNFp-filled and KNFs-filled PP/WTD composites with increasing KNF loadings. Tensile strength was found to decrease with increasing KNF loading for both composites. Incompatibility between hydrophilic KNF and hydrophobic PP/WTD matrices may result in poor interfacial adhesion between them. Hence, the efficiency of stress transfer from PP/WTD matrices to KNF, upon stress application, was reduced, and a low resistance for crack propagation resulted in lower tensile strength.[12,20] In addition, the decrement in tensile strength can be explained by the reduction in the amount of effective PP/WTD matrices with increasing KNF loadings. The insufficient wetting of KNF in limited PP/WTD matrices at high KNF loading tends to impart a lower tensile strength to the composites. Demir et al.[19] reported that the low tensile stress of luffa fiber–PP composites with increasing luffa fiber content is due to the reduction in effective matrix cross-section.

At similar KNF loadings, KNFs-filled PP/WTD composites exhibited higher tensile strength compared with KNFp-filled PP/WTD composites. According to Aji et al.,[22] fiber with a larger length-to-diameter ratio (L/D) results in flexible fibers that are good for fiber bonding and entanglement, which imparts greater tear and tensile strengths to natural fiber composites. KNFs-filled PP/WTD composite gives higher tensile strength because of the higher aspect ratio (L/D) of KNFs and because the fiber length is equal to its critical fiber length compared with that of KNFp. The higher aspect ratio of KNFs will increase the stress transfer efficiency from PP/WTD matrices to KNFs along the fiber–matrix interface.[21,23,24] Mathew et al.[25] reported that the enhancement in mechanical properties of wood powder, wood fiber, and microcrystalline cellulose fibers can be achieved if the fiber length is equal to or greater than the critical fiber length. KNFp is in particulate form where the fiber length is less than the critical fiber length (6 mm for KNF); thus, the stress transfer from matrix to fiber is not efficient, which results in fiber debonding and/or fiber pull-out.[26]

Generally, the tensile modulus of the natural fiber composites is greatly affected by fiber modulus, fiber loading, and the fiber aspect ratio.[27] Higher fiber loading may result in high tensile modulus due to the increasing stiffness of the composites.[12] Figure 3 shows the tensile modulus of KNFp-filled and KNFs-filled PP/WTD composites with increasing KNF loadings. The increasing trend of tensile modulus indicates that the incorporation of KNF into PP/WTD matrices improves the stiffness and rigidity to the composites. The addition of KNF reduced the chain mobility of PP/WTD matrices, which is significant at higher KNF loading. Several researchers have reported similar findings.[12,17,20]

At similar KNF loadings, KNFs-filled PP/WTD composites exhibited higher tensile modulus compared with that of KNFp-filled PP/WTD composites. For reasons similar to those of tensile strength, the higher tensile modulus obtained for KNFs-

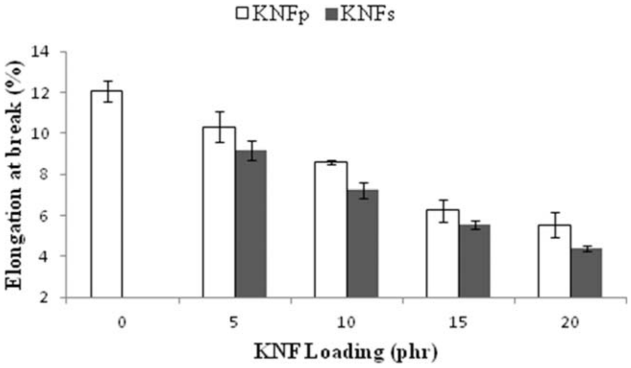

Figure 4. Elongation-at-break of KNFp-filled and KNFs-filled PP/WTD composites at various KNF loadings.

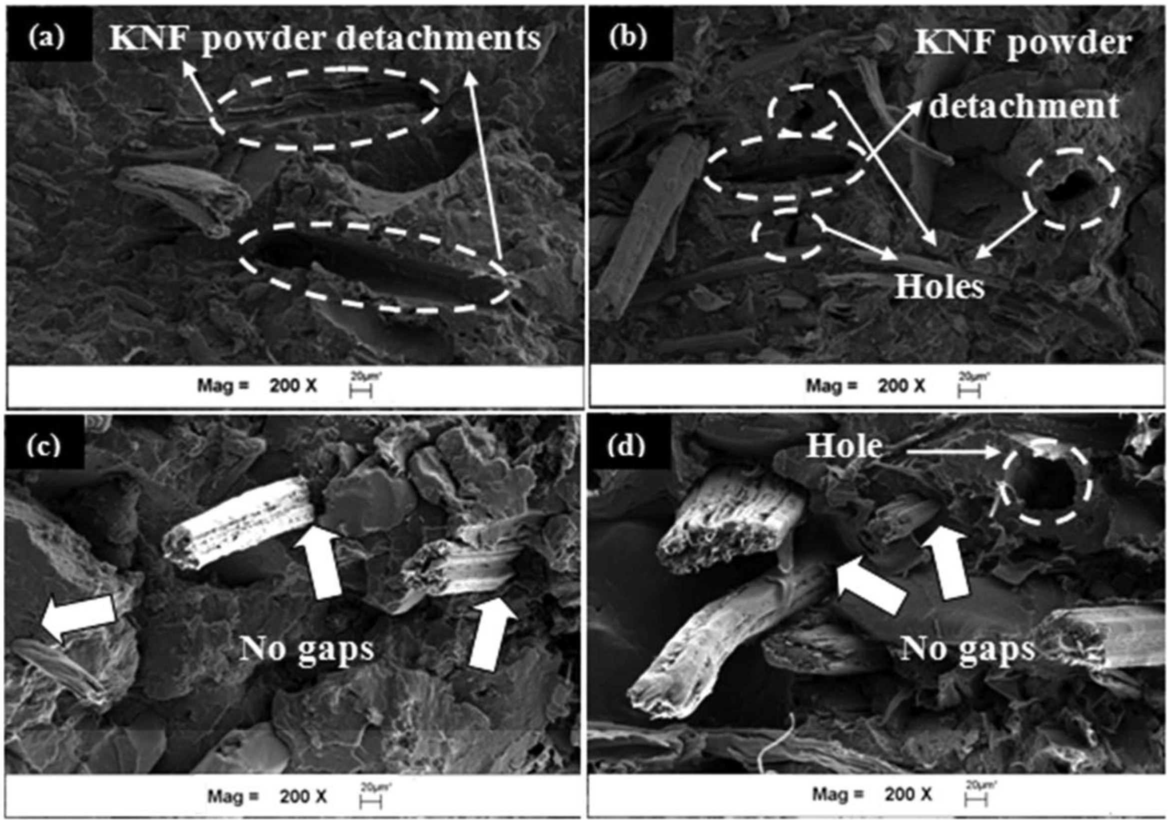

Figure 5. SEM micrographs of tensile fractured surfaces of (a) 10 phr of KNFp-filled PP/WTD composites, (b) 20 phr of KNFp-filled PP/WTD composites, (c) 10 phr of KNFs-filled PP/WTD composites, and (d) 20 phr of KNFs-filled PP/WTD composites; at a magnification of ×200.

filled PP/WTD composites was due to higher fiber aspect ratio (i.e., larger fiber length) of KNFs compared with that of KNFp.[21,23,24] Similar findings were reported by Migneault et al.[21] where the tensile modulus of elasticity and modulus of rupture were higher with larger wood fiber length in wood fiber/high-density polyethylene composites.

Figure 4 shows the elongation at break of KNFp-filled and KNFs-filled PP/WTD composites with increasing KNF loadings. The elongation at break was found to decrease with an increasing KNF loading for both types of composites. The addition of either KNFp or KNFs into the PP/WTD matrices led to an increase in the composite's stiffness, which consequently reduced its toughness. The reason for the reduction in elongation at break, with increasing KNF loading, may have been due to the decreases in the deformability of the rigid interface between the KNF and the PP/WTD matrices. Ismail et al.[20] reported a similar manner where the elongation at break shows a decrement of 52% upon increasing rice husk powder content of 30 phr for PP/recycled acrylonitrile butadiene rubber/rice husk powder composites.

At similar KNF loadings, KNFs-filled PP/WTD composites exhibited a lower elongation at break compared with KNFp-

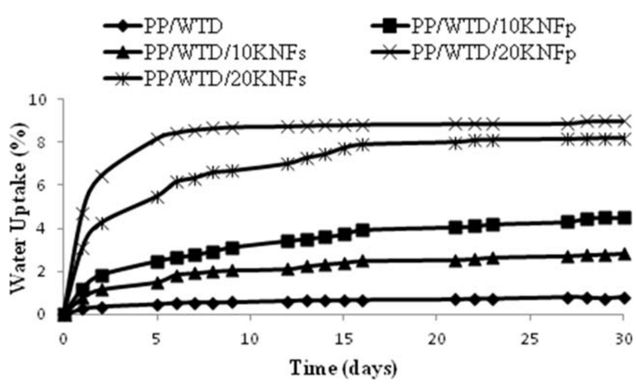

Figure 6. Water uptake of KNFp-filled and KNFs-filled PP/WTD composites at various KNF loadings.

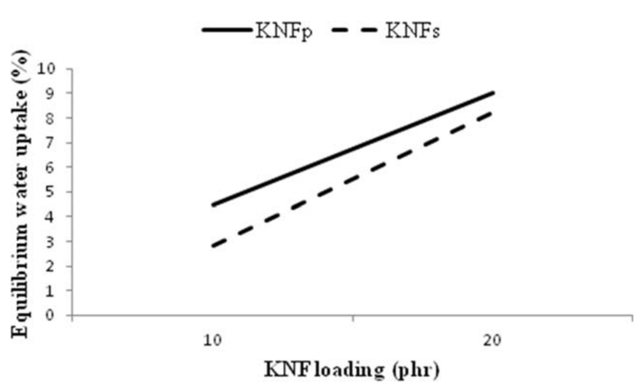

Figure 7. Comparison of equilibrium water uptake of KNFp-filled and KNFs-filled PP/WTD composites at 10 and 20 phr of KNF loading.

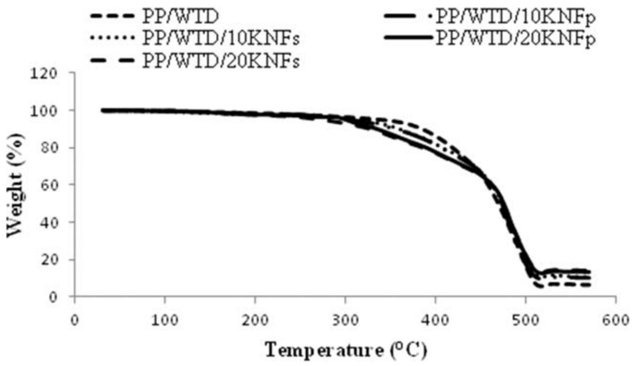

Figure 8. Thermogravimetric analysis thermogram of KNFp-filled and KNFs-filled PP/WTD composites.

filled PP/WTD composites. Again, as explained earlier, this observation may have been due to a higher aspect ratio of KNFs (i.e., larger fiber length), which resulted in higher composite stiffness (higher tensile modulus) and subsequently decreases the deformability of the composites.[21]

Morphological Properties

Scanning electron microscopy (SEM) micrographs of the fractured surface for KNFp-filled PP/WTD composites at 10 and 20 phr KNF loadings are shown in Figure 5(a,b). The interfacial adhesion between KNFp and PP/WTD matrices is poor as can be seen from the KNFp detachment sites and holes presence upon stress application. It was clearly seen in Figure 5(b) that more KNFp detachment sites and holes are present due to the uneven distribution of KNFp throughout PP/WTD matrices. This observation confirmed that higher KNF loading results in weaker interfacial adhesion between the KNFp and PP/WTD matrices, proven by the poor tensile strength at higher KNF loading.

SEM micrographs of the fractured surface for KNFs-filled PP/WTD composites at 10 and 20 phr KNF loadings are shown in Figure 5(c,d). It is clear that KNFs are strongly adhering to PP/WTD matrices with no big voids at the interface between them. The interfacial adhesion between KNFs and PP/WTD matrix is better compared with KNFp and PP/WTD matrix, which can be seen by fewer holes and fiber pull-out. This is due to the large aspect ratio of KNFs that gives higher tensile properties of KNFs-filled PP/WTD composites compared with KNFp-filled PP/WTD composites.[21,25]

Water Uptake

Figure 6 shows the water uptake of KNFp-filled and KNFs-filled PP/WTD composites with KNF loadings of 0, 10, and 20 phr. All composites showed an increase in water uptake with increasing immersion time and KNF loading. Initially, a sharp water uptake by all composites was seen, followed by a gradual increase until the equilibrium stage was reached at approximately 30 days. The presence of lumens, fine pores, hydrogen bonding sites in the natural fiber, the gaps and flaws at the interfaces, and the microvoids that formed in composites during the compounding process are all possible reasons for the water uptake in natural fiber polymer composites.[28] Thus, as KNF loadings increased, the increasing number of lumens, fine pores, hydrogen bonding sites, and microvoids in the KNF contributed to the increase in water uptake of the composites.

At similar KNF loadings, KNFs-filled PP/WTD composites exhibited a lower water uptake and equilibrium water uptake than KNFp-filled PP/WTD composites, as shown in Figures 6 and 7, respectively. This may have been due to a larger aspect ratio of KNFs that increased the interfacial interactions between the PP/WTD matrix and the KNFs and caused less water penetration into the composites. For this reason, a lower equilibrium water uptake was observed in PP/WTD composites filled with KNFs compared with composites filled with KNFp.

Thermal Properties

Figure 8 shows the thermogravimetric analysis thermogram of KNFp- and KNFs-filled PP/WTD composites. Table II shows the temperature at 50% weight loss, $T_{50\%}$, and the char residue of KNFp- and KNFs-filled PP/WTD composites.

It can be seen that $T_{50\%}$ and char residue increased with increasing KNF loading. A higher temperature was required to decompose composites with KNF, and increase in KNF loadings resulted in higher $T_{50\%}$ and char residue values. The formation of char prolongs the degradation of nondecomposed polymer by forming a barrier between the heat source and the polymeric material.[2,29] In general, a solid polymer will breakdown into small fragments when subjected to heat or elevated temperatures. When undergoing thermal decomposition, the lighter fragments will vaporize shortly after their formation, and heavier fragments may be subjected to further decomposition into lighter fragments for easier vaporization. However, not all polymers breakdown and vaporize completely, leaving no solid residues in the chamber. There is still the existence of some solid residue after heating, because not all fragments vaporize completely. The remaining solid residues can

Table II. TGA Parameters of KNFp-Filled and KNFs-Filled PP/WTD Composites

	Temperature at 50% weight loss, $T_{50\%}$ (°C)		Char residue (%)	
KNF loading (phr)	PP/WTD/KNFp composites	PP/WTD/KNFs composites	PP/WTD/KNFp composites	PP/WTD/KNFs composites
0	468	468	6.91	6.91
10	473	474	10.14	11.05
20	475	476	13.38	14.03

be carbonaceous (char), inorganic (either within the polymer structure or additive added), or a combination of both. In this study, KNFp or KNFs decomposed upon heating, leaving behind a carbonaceous material (char). When this char layer formed, it tended to form as a barrier to slow down the further thermal decomposition of the composites. Thus, as the char residue increased (i.e., the char layer became thicker), the heat flow to decompose the composites reduced. This occurred because when the thermal decomposition of a deeper layer of the material continues, the formation of volatiles inevitably has to pass through the char above to reach the surface. Hence, higher char residue indicates a higher thermal stability of composites.[28] However, at similar KNF loadings, the different forms of KNF do not show any significant effect on the thermal properties of KNF-filled PP/WTD composites, as can be seen from the similar $T_{50\%}$ and char residue values in Table II.

CONCLUSIONS

The stabilization torque, tensile modulus, and water uptake increased with increasing KNF loadings for both KNFp- and KNFs-filled PP/WTD composites. At similar KNF loadings, KNFs-filled PP/WTD composites give higher stabilization torque, tensile strength, and tensile modulus, but a lower elongation at break and water uptake compared with KNFp-filled PP/WTD composites. The thermal stability of KNFp- and KNFs-filled composites increased with increasing KNF loadings, but showed no significant increment at similar KNF loading. The different KNF forms had significant effect on the processing, tensile properties, and water uptake compared with thermal properties. SEM morphological study proves poor adhesion between KNFp and PP/WTD matrices, compared with the adhesion of KNFs and PP/WTD matrices, attributing to the higher properties of PP/WTD/KNFs composites. Both types of composites can be used to make low-cost interior car items, such as dashboards, door handles, and door panels.

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