

Mechanical Properties of Recycled Kenaf/Polyethylene Terephthalate (PET) Fiber Reinforced Polyoxymethylene (POM) Hybrid Composite

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ABSTRACT: Environmental concerns have attracted researchers to study the recycling of composite materials and thermoplastics due to the desire not to waste materials and reduce disposal of scraps that may eventually pollute the environment. The main objective of this article is to study the effect of recycling on the mechanical properties of kenaf fiber/PET reinforced POM hybrid composite. The virgin hybrid composite was produced by compression molding and later subjected to mechanical testing. The scraps obtained after the mechanical testing were shredded, granulated and subjected to compression molding to produce samples for mechanical testing. Tensile strength of 27 MPa was obtained and (after second recycling process) which is lower compared to 73.8 MPa obtained for the virgin hybrid composite. There was a significant increase in flexural modulus (4.7 GPa) compared to the virgin hybrid composite. The impact strength dropped to 4.3 J cm^{-1} as against 10.5 J cm^{-1} for the virgin hybrid composite. The results of thermal degradation showed about 80% weight loss for kenaf fiber between 300 and 350°C . The weight loss may be due to the degradation of cellulose and hemicellulose content of the fiber. The percentage water absorption of the recycled composite dropped by about 80% compared to the virgin hybrid composite. © 2013 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2014**, *131*, 39831.

KEYWORDS: degradation; thermogravimetric analysis; recycling; composites

Received 1 June 2013; accepted 8 August 2013

DOI: 10.1002/app.39831

INTRODUCTION

The current trend in the search for green environmentally friendly materials has attracted the attention of researchers to develop recyclable composite materials. One of the methods employed to achieve this aim is to replace the synthetic reinforcing materials such as glass fibers with natural fibers. The significance of using natural fibers cannot be overemphasized. The natural fibers are lighter than synthetic fibers and therefore reduce costs and processing time.^{1,2} The advantages of using natural fibers include weight savings by about 50% and reduced costs by ~30% while still remain environmentally friendly.³ Composites based on thermoset polymers are often regarded as impossible to recycle due to their crosslinked three dimensional structure. Their attractive mechanical properties have made them useful for large volume production in the automotive industry. On the other hand, thermoplastics are characterized as long chain polymers with semicrystalline or amorphous structure. They have high toughness, resistance to chemical attack and are recyclable. One of the advantages of using thermoplastic polymers is that they can be heated and rapidly cooled without changes in their microstructure. The choice of hybrid composite in this study was based on good mechanical and thermal properties that cannot be achieved with single fiber reinforced com-

posite. The properties are high stiffness, high strength, light weight, good wear resistance and low coefficient of thermal expansion. In a hybrid composite, one of the fibers will have a higher elastic modulus than the other. The high elastic modulus fiber will provide the stiffness and load bearing quality while the low modulus fiber makes the composite more damage tolerant and keeps the material cost low.

Recycled thermoplastic composite materials show degradation in mechanical properties and the degradation depends on the processing parameters. During recycling, the molecular weight of the polymers is reduced due to mechanical and thermal decomposition. As a result of these, the degraded polymer chains become less effective in linking to one another. The melting temperature of the material is reduced, the impact strength diminished, and the melt flow index increases.⁴

The common processes used for recycling of composite materials involved size reduction using slitting rollers, hydraulic shears, and shredders and then followed by post reduction using Granulators. The Granulator is expected to reduce the size of the composite to about 2–6 mm in diameter. The palletized material can then be injection or compression molded to produce the final part required.

The mechanical properties of recycled kenaf fiber and rice hulls (RH) reinforced polypropylene (PP) were investigated.⁵ The study conducted revealed that the flexural modulus of the PP/RH composite reduced by 10% after second recycling while the flexural modulus remains unchanged. A reduction in flexural strength of about 5% was observed for PP/kenaf composite after second recycling with an increase of about 20% in flexural modulus.

Duigo et al.⁶ studied the mechanical properties of flax fiber reinforced poly-L-lactide (PLLA) composite using an injection molding process. The results obtained showed that repeated injection molding cycles influenced the geometry of the reinforcement, mechanical properties and thermal behavior of the composite. Tensile stress, elastic modulus, and strain at break reduced due to fiber damage during the recycling process which resulted to poor fiber–matrix interfacial bonding. The decrease in tensile strength and modulus was similarly observed when PP was used as the matrix.⁷

Recycled wood floor from furniture making are recently been used as reinforcements with recycled thermoplastic materials to produce door stiles, rails and window lineal. The water absorption and thickness swelling of the recycled wood floor reinforced PP composite was investigated.⁸ The mixture of sawdust and recycled PP was used to produce the composite. The process used was melt compounding and injection molding. The results obtained from the investigation revealed that samples produced with lower content of wood floor showed low water absorption and thickness swelling compared to those with a high content of wood floor. Samples produced using compatibilizer showed lower thickness swelling compared to those without compatibilizer.

The mechanical properties of wood plastic composites using wood floor, recycled PP, polyethylene (PE) and virgin thermoplastics (PP and PE) materials were also investigated using melt blending and injection molding process. The results indicate that the mechanical properties of composites with PP are significantly higher than those with PE. The tensile strength and modulus of the composite decreased with increasing recycled plastic loading, while the impact strength increases with increase plastic content.⁹

The thermal stability of natural fiber composites during recycling can be studied using thermogravimetric analysis (TGA). The degradation of the natural fibers usually occurred in two stages: the first is a low temperature degradation which is from 220 to 250°C and the second is a high temperature degradation which is from 280 to 400°C. The low temperature degradation is associated with the degradation of hemicellulose content of the fiber while the high temperature is associated with degradation of lignin. The investigations conducted on high temperature thermal degradation of some natural fibers revealed that about 60% losses in mechanical properties were observed.^{10,11} Some researchers have worked on improving the thermal stability of natural fiber composites by grafting the fibers with monomers. The results showed a significant improvement since lignin can react with monomers.^{12,13}

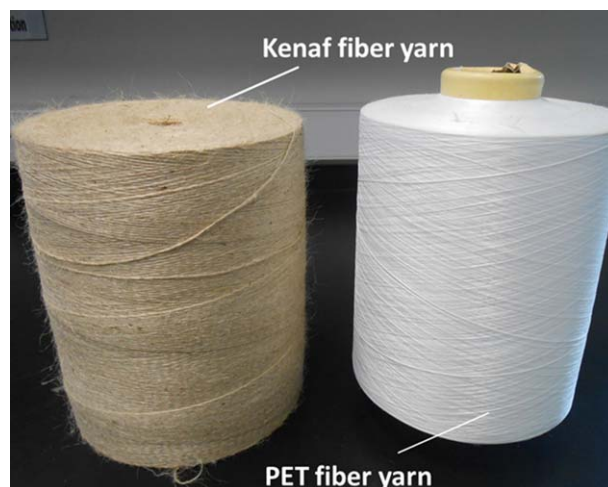


Figure 1. Kenaf fiber and PET fiber yarn. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

The main objective of this article is to study the mechanical properties of recycled POM/kenaf/PET hybrid composite and also to study the effect of recycling on the thermal degradation of the composite and its constituents.

MATERIALS AND METHODS

Material Selection and Equipment

A continuous kenaf yarn with a diameter of about 1 mm and PET yarn with a diameter of about 0.3 mm were selected as reinforcements to produce the hybrid composite. Figure 1 shows the kenaf and PET fiber used for the production of the hybrid composite. Acetal copolymer (POM) with melt flow index (MFI) of 9 g/10 min was used as the matrix. The selection of kenaf fiber was based on availability, good mechanical properties, low cost, and thermal stability compared to other locally available natural fibers like Oil palm and Coconut fiber. The thermal degradation of kenaf fiber begins at a temperature of about 200°C which makes it suitable as reinforcement in thermoplastic matrix with a processing temperature between 180 and 240°C. The selection of PET fiber was based on its thermal stability with POM in a composite and having good mechanical properties. The PET fiber does not melt during the production of the composite due to its high melting temperature (265°C) which is above the processing temperature (200°C) of the composite. A good interfacial bonding between the fibers and the matrix is best achieved at a processing temperature of 200°C without leading to the degradation of the kenaf and PET fibers. The mechanical properties of the kenaf and PET fiber remain unchanged even after the production of the composite. The selection of POM as the matrix was based on its good mechanical and physical properties compared to the locally available thermoplastics. Its high strength, good resistance to fatigue and good resistance to environmental attack may make it suitable for outdoor application in the automotive industry.

Tensile, flexural, and impact testing molds designed according to ATSM D638, D790, and D6110-10 were used for the fabrication of the virgin and recycled hybrid composite. A compression molding machine manufactured by Carver, USA with a

maximum processing temperature of 400°C was used alongside a domestic cooling fan to support the cooling process. A Universal Testing Machine (UTM) was used to test the material for tensile and flexural strength. A universal impact testing machine was used for Charpy impact test on all samples produced. A 500 g digital weight balance was used to take the weight of the reinforcements and the samples during production and water absorption test.

Production of Virgin Hybrid Composite

The mold was prepared by scraping the surfaces and the edges of the mold cavity to remove stains of other materials that may mix with the composite during production. The POM pellets were charged into the mold and were allowed to preheat at 200°C for 5 min inside the compression molding machine. The mold was then compressed at 200°C with a pressure of 130 MPa for 15 min due to low melt flow index of the matrix. The mold was further cooled to 50°C before it was discharged from the machine. The POM sheets (2-mm thick) produced was used as layers to produce the hybrid composite. The continuous kenaf and PET yarn were placed in between layers of POM inside the mold before taking into the compression molding machine. The weight percentage of the POM/Kenaf/PET was maintained at 80/10/10 for all test samples. Increasing the fiber content accelerates the degradation of composites during recycling by reducing the molecular weight of the polymer matrix.¹⁴ The high fiber content may restrict chain mobility in the matrix due to fiber–matrix interactions, therefore low fiber content was used in this study. The reduction in molecular weight can adversely affect the mechanical properties of the composite after recycling.

Production of the Recycled Hybrid Composite

After subjecting the virgin composite to mechanical testing and environmental degradation, the degraded samples were crushed and palletized in a Granulator to a diameter of about 5 mm. The palletized composite was compression molded at 200°C at a pressure of 150 MPa for 15 min. The mold was allowed to cool to 80°C under pressure before it was discharged from the compression molding machine. The composite was further recycled after subjecting it to mechanical testing and environmental degradation using the same production process stated above.

Mechanical Testing, Water Absorption, and Thermal Degradation

Tensile and flexural test of all samples were conducted according to ASTM D638 and D790, respectively.¹⁵ A notched Charpy impact test was conducted according to ASTM D6110-10 for testing of reinforced thermoplastic composite. The water absorption test was carried out according to the specification given in ASTM D579-99.¹⁶ An average of five samples were used to compute the final result of all individual tests carried out. Thermal degradation analysis of each constituent material (POM, kenaf, and PET) and the recycled composite was carried out to determine percentage weight loss as a function of temperature and time. Each sample was heated from 29 to 850°C for 40 min and the results of the degradation were recorded.

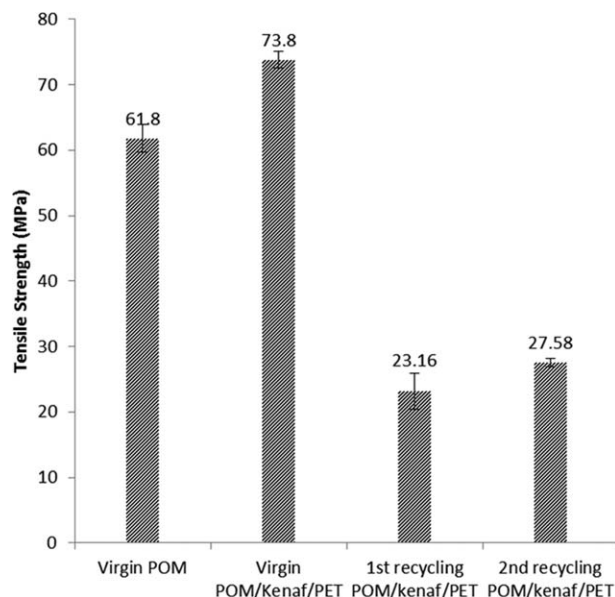


Figure 2. Tensile strength of the virgin and recycled hybrid composites.

RESULTS AND DISCUSSION

Tensile Strength and Elastic Modulus

Figures 2 and 3 show a drop in tensile strength and modulus after first and second recycling compared to the virgin hybrid composite. This drop may be due to fiber damage during recycling which can cause reduction in fiber length with high risk of fiber debonding.¹⁷ Repeated recycling of the hybrid composite has shown to influence the fiber geometry in the composite which may affect the mechanics of load transfer between the fibers and the matrix.¹⁸ The tensile properties do not only depend on the properties of the fiber and the matrix, but also on the interfacial bonding between the reinforcing fibers and the thermoplastic matrix.¹⁹ The stress transfer at the interphase requires good interfacial adhesion. To achieve good interfacial bonding, the surface energy of the reinforcing fiber should be equal or greater than the surface energy of the matrix. Figure 4

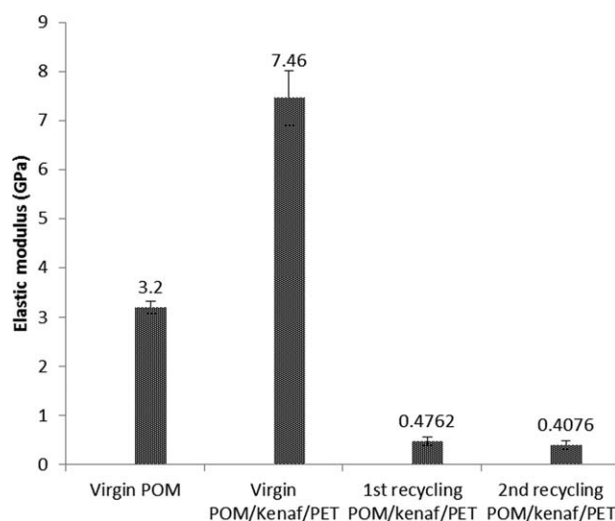


Figure 3. Elastic moduli of the virgin and recycled hybrid composites.

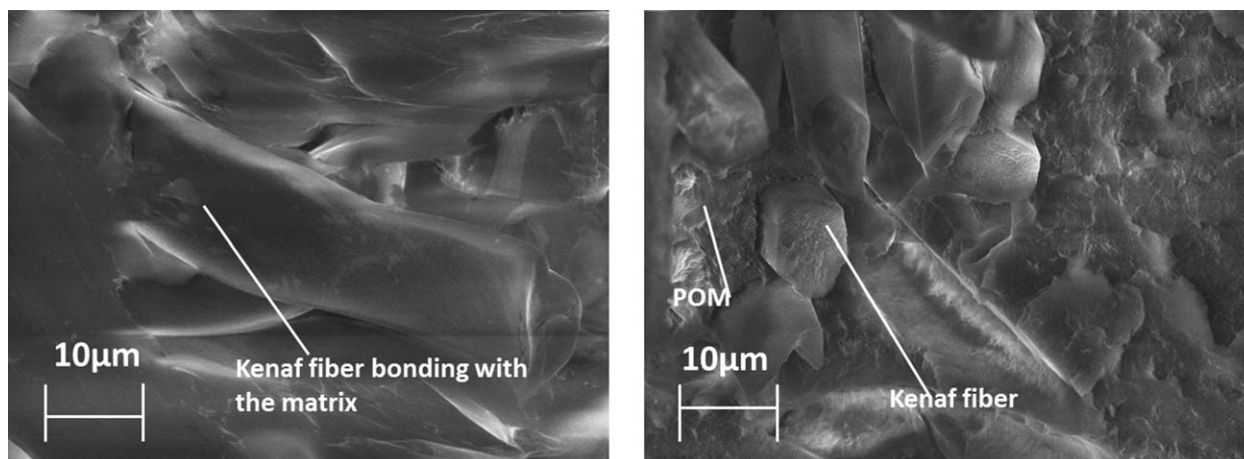


Figure 4. FESEM micrograph showing POM-kenaf bonding in the virgin hybrid composite.

shows the interfacial bonding between kenaf fiber and POM in the virgin hybrid composite. The good interfacial bonding at the interphase between kenaf and POM was due to high surface energy of the kenaf fiber with respect to POM. Figure 5 also shows a good bonding at the interphase between PET fiber and POM. The significant increase in tensile properties of the virgin

hybrid composite was due to good interfacial tension between the fibers and the matrix as shown in the FESEM micrograph. However, the poor interfacial bonding at the interphase between POM/kenaf and POM/PET in the recycled hybrid composite can be observed in Figure 6. The degradation of the reinforcements during recycling resulted in poor interfacial bonding

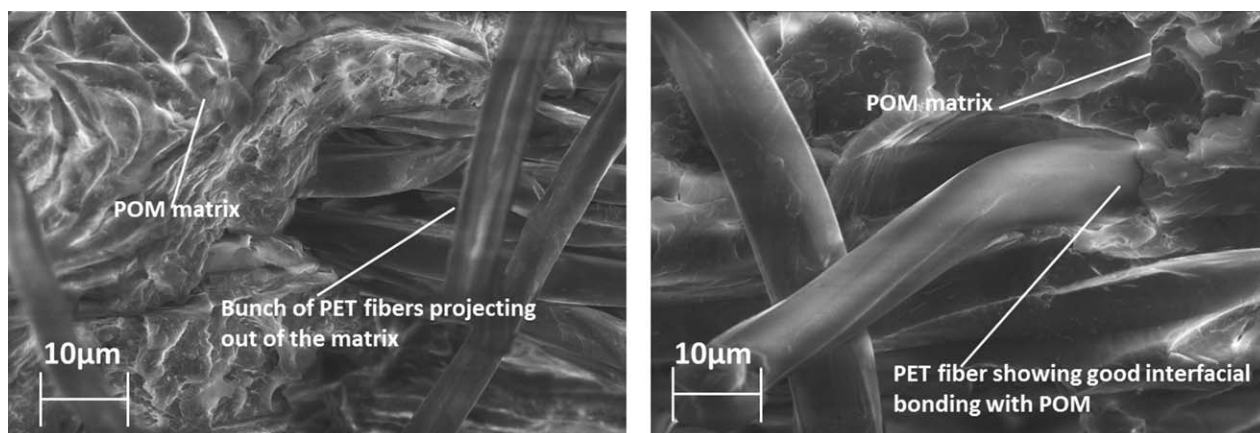


Figure 5. FESEM micrograph showing POM-PET bonding in the virgin hybrid composite.

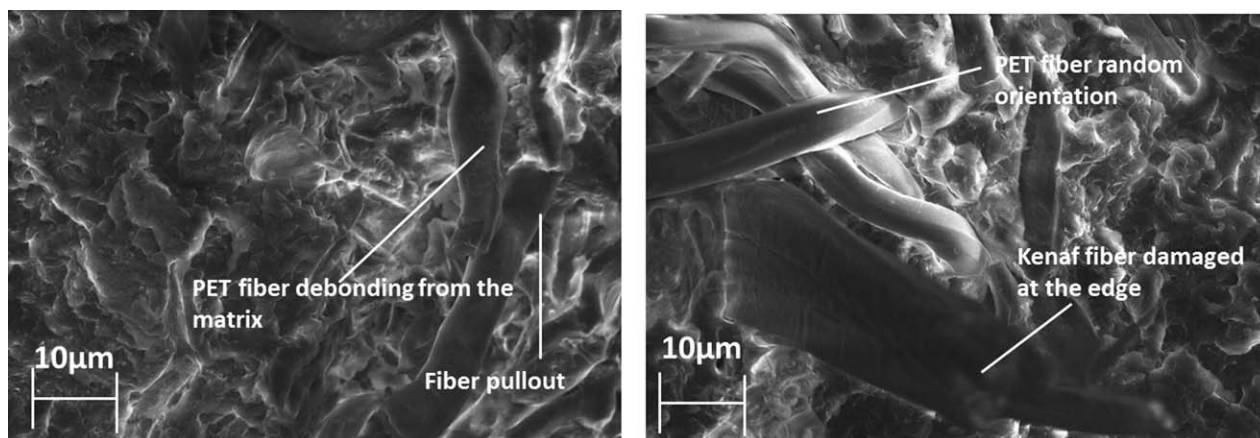


Figure 6. FESEM micrograph showing POM-kenaf-PET orientation in the recycled hybrid composite.

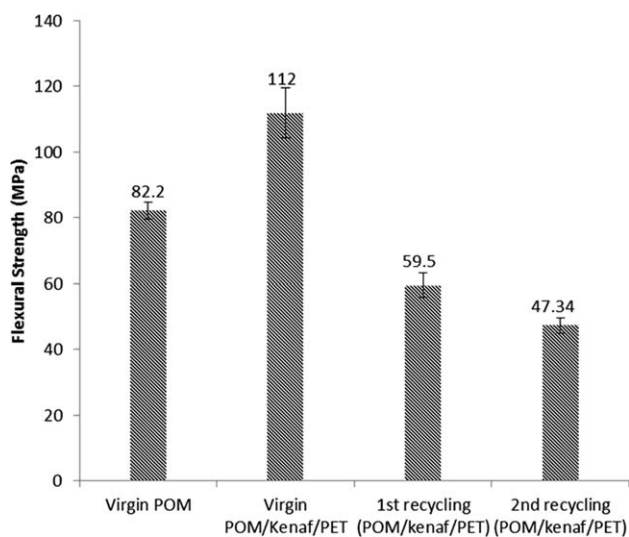


Figure 7. Flexural strength of the virgin and recycled hybrid composite.

between the fibers and the matrix. The poor interfacial bonding at the interphase may be due to the reduction in surface energy of the reinforcements during recycling.

Flexural Strength and Modulus

The flexural strength of the recycled hybrid composite dropped by about 100% with respect to the virgin hybrid composite as shown in Figure 7. The flexural modulus increases to 3.4 GPa after the first recycling and further increased to 4.7 GPa after the second recycling as shown in Figure 8. The drop in flexural strength and the significant increase in flexural modulus may be attributed to large reductions in fiber length and aspect ratio during recycling. The large reduction in fiber length may be due to shear stresses developed during the compression molding process. The interaction of the reinforcements with the mold cavity also accelerates degradation of the composite.

Impact Strength

Figure 9 shows the impact strength of the virgin and the recycled hybrid composite. The recycled hybrid composite showed a brittle type of failure during the impact test. Impact strength of 4.3 J cm⁻¹ was obtained which is lower compared

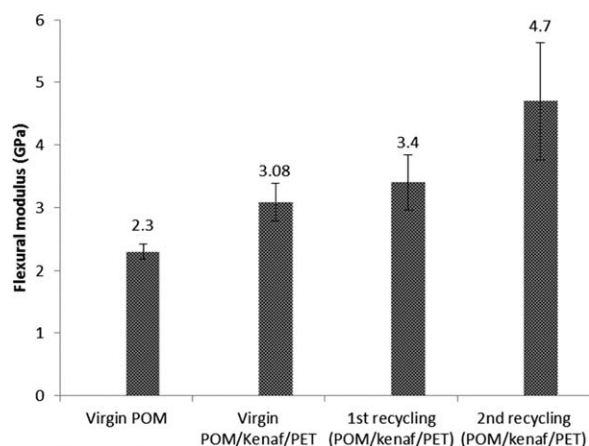


Figure 8. Flexural moduli of the virgin and recycled hybrid composite.

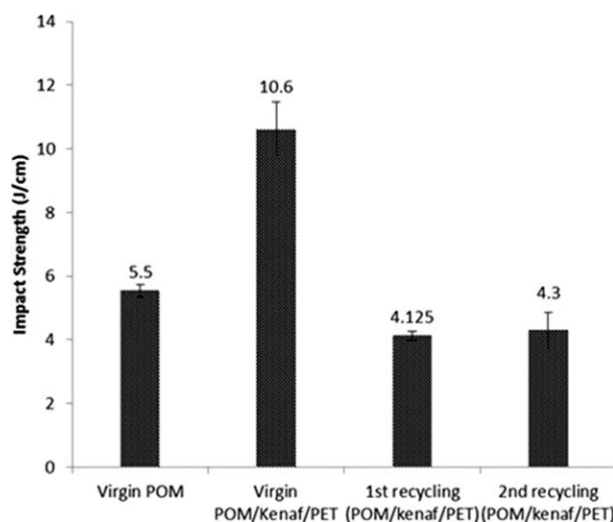


Figure 9. Impact strength of the virgin and recycled hybrid composite.

to 10.6 J cm⁻¹ for the virgin hybrid composite. Repeated recycling has increased the composite brittleness and reduced its impact strength. The poor interfacial tension between the fibers and the matrix also contributed to low impact strength of the recycled composites.²⁰

Thermal Degradation

Figure 10 shows the percentage weight loss of POM, kenaf, and PET fiber as a function of temperature before recycling. About 15% weight of kenaf fiber has degraded at 200°C and about 80% weight loss was observed between 300 and 350°C which may be due to the decomposition of lignin and cellulose content of the fiber.²¹ The weight loss of POM was observed to be similar to PET fiber until 350°C. The weight loss of PET fiber has increased by about 70% between 400 and 450°C with respect to POM as shown in Figure 9. This may be due to the reduction in molecular weight during recycling. The degradation curve in Figure 11 showed the percentage weight loss of the virgin and the recycled hybrid composite. The degradation of the recycled hybrid composite started earlier to that of the

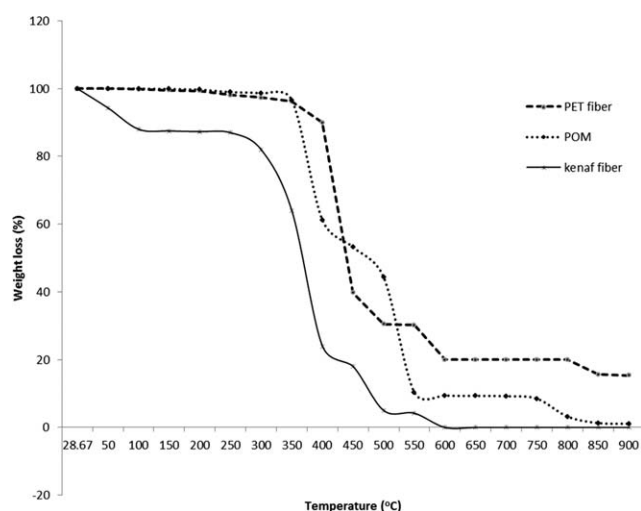


Figure 10. Thermal degradation curve for kenaf, PET and POM.

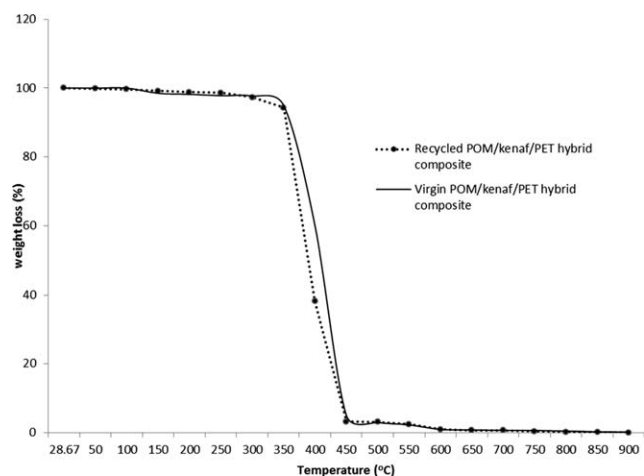


Figure 11. Thermal degradation curve for the virgin and recycled hybrid composite.

virgin hybrid composite. The recycled hybrid composite showed a weight loss of about 90% between 350 and 400°C compared to the virgin hybrid composite with a weight loss of about 90% between 400 and 450°C. The mechanical and thermal degradation of the constituent materials during recycling accounted for early weight loss in the recycled composite. The depolymerisation of macromolecular chains caused a reduction in molecular weight in the matrix and contributed to the degradation of the recycled composite.²² The mechanical degradation may be due to the interaction of the matrix with the mold during the compression molding process.²³

Water Absorption and Density

The percentage water absorption of the recycled composite dropped below that of the virgin hybrid composite (6.7–0.55%) as shown in Figure 12. This may be due to the degradation of the cellulose and hemicellulose content of the kenaf fiber during recycling. The cellulose is a hydrophilic polymer that contains a number of hydroxyl groups. The hydroxyl group reacts with hydrogen bond of water molecules which results in high mois-

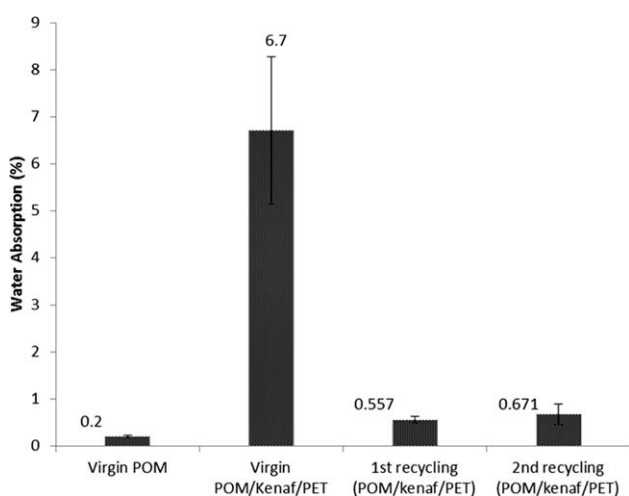


Figure 12. Water absorption results of the virgin and recycled hybrid composite.

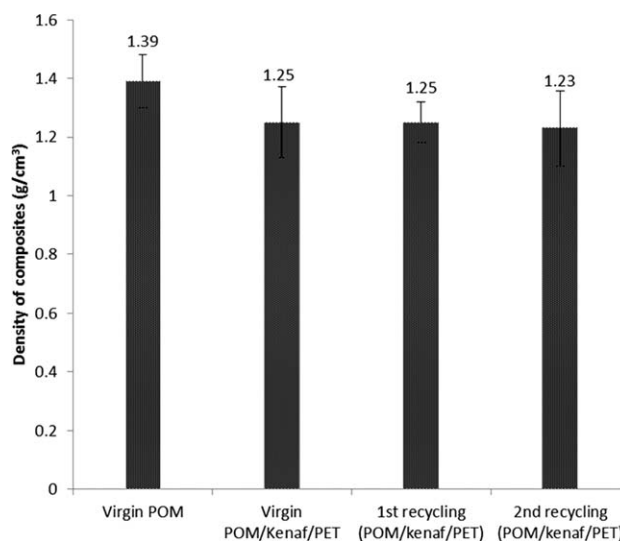


Figure 13. Density of the virgin and the recycled hybrid composite.

ture uptake in the composite.²⁴ This explained why the degradation of the cellulose and the hemicellulose during recycling may reduce the moisture uptake in the recycled hybrid composite. This process of moisture uptake does not take place in thermoplastics as PET fiber and POM are hydrophobic. Reduction in fiber length during the recycling process and reduction in aspect ratio may also influence moisture absorption. The density of the recycled composite remains unchanged after the first recycling but it reduced further to 1.23 g cm⁻³ after the second recycling as shown in Figure 13. The reduction in density after the second recycling may be due to mechanical degradation of kenaf fiber during recycling.

CONCLUSION

The process of recycling the hybrid composite resulted to fiber damage, reduction in fiber length, poor fiber–matrix interfacial bonding and thermal degradation of the reinforcements. Tensile strength of 27 MPa was obtained after second recycling. The poor interfacial bonding between the fibers and the matrix accounted for the drop in tensile strength and elastic modulus. A flexural strength of 47.34 MPa showed about 100% drop compared to the virgin hybrid composite. The microstructure of the recycled composite showed the random distribution of the fibers in the matrix and poor fiber–matrix interfacial tension. Thermal degradation results showed about 80% weight loss for kenaf fiber between 300 and 350°C. The mechanical and thermal degradation of the constituent materials during the first and second recycling process accounted for low impact strength in the composite. The degradation of cellulose and lignin in kenaf fiber during recycling resulted to low moisture uptake in the recycled hybrid composite.

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