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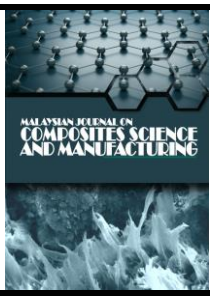

Thermo-Mechanical, Structural, and Biodegradability Properties of Water Hyacinth and Sheep Wool Fiber Reinforced Hybrid Polypropylene Composites

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ABSTRACT

Hybrid fiber reinforcements can incorporate a wider array of qualities than single fiber reinforcement. Instead of synthetic fibers, applying plant and animal-based organic materials as reinforcement in polymer matrix offers certain benefits, such as low price, greater availability, and better biodegradability. In the present study, water hyacinth fiber and sheep wool fiber reinforced polypropylene hybrid composites were fabricated at three different (5, 10, and 15 wt.%) fiber loading. The effect of fiber loading on thermo-mechanical, structural, and biodegradability properties was subsequently investigated. The manufacturing process of the composite material was carried out with utmost consideration for biodegradability, since polypropylene, the primary constituent, is not inherently biodegradable. The insertion of fibers into the polypropylene matrix showed variance in properties of different aspects. The tensile strength of the composites displayed a downward trajectory (from 25 to 10 MPa) with a 15% increase in fiber loading due to voids, and fiber dispersion, while impact strength exhibited an opposite trend (from 25 to 32 J/m). Except for hardness, all the mechanical properties degraded slightly after the employment of the reinforcement. Fourier transform infrared spectroscopic analysis revealed the movement of typical peaks and the appearance of new peaks demonstrating the bonding between the fiber and the matrix. Thermogravimetric analysis showed that the thermal degradation temperature of the composites improved at maximum fiber loading. On the other hand, the goal of achieving biodegradability has been succeeded by the implementation of a combination of plant and animal-based fibers as biodegradability of the manufactured composites thrives with increasing fiber content for the presence of cellulosic bonds, as evident from the FTIR spectrum. Even though some properties of the hybrid composite declined slightly with increasing fiber loading, the other characteristics, including service temperature and biodegradability experienced a prospective advancement. Hence, the 15% fiber-loaded composite was found to be a potential candidate in terms of slightly high temperature and environment-friendly applications.

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1. Introduction

Hybrid fiber-reinforced composites, which include various reinforcements in a single matrix structure, provide a lot of flexibility for customizing a variety of qualities [1]. In addition to having a high strength-to-weight ratio, fiber-reinforced polymer composites also have remarkable durability, flexural strength, and impact resistance [2]. Composite materials have been used in various manufacturing sectors, including construction, aerospace, automotive, biomedical, and many more [3]. Due to the severe environmental contamination caused by the consumption of plastics, there has been a surge in interest in using biodegradable polymers [4]. One of the most widely used plastics in markets is polypropylene (PP), which has benefits in terms of cost, recyclable potential, and technical criteria like greater heat stability.

The mechanical properties of synthetic polymers are now improved by mixing them with a variety of reinforcing fillers to get the desired qualities for practical use [5-7]. Research is being done to substitute synthetic fibers with lignocellulosic fibers as reinforcing fillers [8, 9]. The lignocellulosic fibers (rice husk, coir, jute, abaca, water hyacinth, etc.) are lightweight, reduce machine wear, and are readily accessible, sustainable, and affordable. Additionally, they are biodegradable and do not leave behind hazardous residues [10-12]. Lignocellulosic polymeric composites are made at an affordable price. As a result, these composites have received considerable interest and are playing an ever-growing part in creating a wide range of affordable, lightweight, environmentally friendly composites [13].

In the corresponding study, water hyacinth fiber and sheep wool fiber were used as the reinforcing materials since they both are readily available in the Indian sub-continent, especially Bangladesh, and have a minimal effect on the environment because of their biodegradable properties [14-16]. Although studies have been conducted on the individual effects of water hyacinth fiber and sheep wool fiber on polypropylene composites, no research has been found on their combined impact in enhancing such composites' thermo-mechanical, structural, or biodegradable properties. For example, a recent study done by Kulkarni *et al.* [17], focused solely on sheep wool-based polymer composites, where the addition of sheep wool fiber reinforcement leads to a significant increase in tensile strength, with improvements of up to 25% compared to unmodified polymers. The natural thermal stability of sheep wool fibers also enhances thermal resistance, as demonstrated by a 15°C increase in the onset degradation temperature. On the other hand, other studies were done by Varghese *et al.* [18] and Mahardika *et al.* [19] examined the effects of water hyacinth fibers in isolation. According to a research study examining the thermo-mechanical and morphological properties of water hyacinth (WH) fiber-reinforced polypropylene (PP) bio-composites, chemically treated WH fiber composites exhibited significant improvements in thermal stability and all mechanical properties, except for tensile strength, when compared to untreated composites [15]. However, the study did not specifically address the biodegradability of these composites. Another recent study investigated the potential of natural fibers, specifically pigeon pea stalk fibers and banana peel, as reinforcing agents in polypropylene composites. The study revealed that composites with the highest fiber loading, at 40 wt%, exhibited the highest level of biodegradation compared to composites with 10 and 20 wt% untreated fiber in polypropylene [20]. These findings suggest that using natural fibers in polypropylene composites could lead to more environmentally friendly materials with improved biodegradability. Compared to untreated fiber composites, maximum biodegradation was observed in treated fiber composites with the same amount of fiber loading.

This study aims to manufacture water hyacinth and sheep wool fiber-reinforced hybrid polypropylene composites and subsequently characterize those using thermomechanical testing, structural analysis, and biodegradability testing. The effect of the fiber loading on the mentioned properties of the corresponding composites was also reported.

2. Materials and Methods

2.1 Materials

Fibers from water hyacinth and sheep wool were mixed equally to reinforce the polypropylene matrix to fabricate the concerned hybrid composite. Polypropylene was blue in color and granular in size and its melting point was 130-170 ℃. Polypropylene and sheep wool were collected from the local market. Extraction of Water Hyacinth fiber was manual. Stems of water hyacinth were collected from local ponds. These were cut into small pieces and their peels were removed. Finally, fibers were extracted from the fibrous core of the stem. The entire process has been summarized in Figure 1.

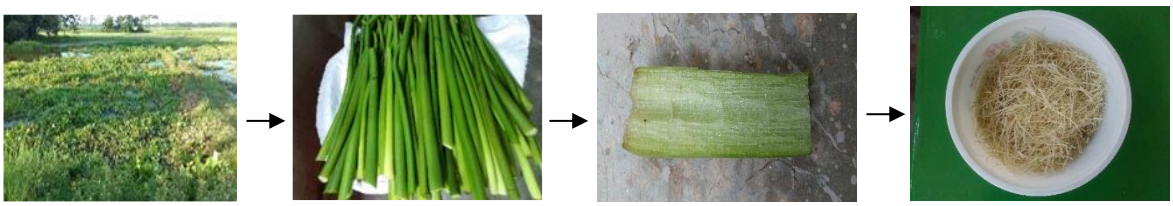

Fig. 1. Processing of water hyacinth fiber

2.2 Fabrication of Composites

Each of the fibers was cut into 3-5 mm size in length and measured in weight to get the perfect amount according to the required percentage. The amount of polypropylene was weighed at 95 gm, 90 gm, and 85 gm for 5 wt.%, 10 wt.%, and 15 wt.% fiber loadings, respectively. The mould surface was cleaned very carefully, and the mould-releasing agent was sprayed over the mould surface properly for easy removal and a good surface finish of the manufactured composite. After mould preparation, one layer of polypropylene was distributed uniformly throughout the lower mould. Then, the weighed fibers were distributed similarly on top of the polypropylene layer. The remaining polypropylene was distributed on top of the fiber, as shown in Figure 2 and was covered with the upper mould. The mould was placed in the hot press machine. The mixture of fibers and matrix was allowed to press at 30 kN pressure, and the temperature was initially raised to 150°C. When the temperature reached 150°C, it was held for 5-10 minutes. Afterwards, the temperature was raised to 165°C and then to 190°C and held there for 2-3 minutes. Finally, water was supplied to cool the mould to room temperature. After cooling, the pressure was released, and the specimen was carefully withdrawn from the mould.

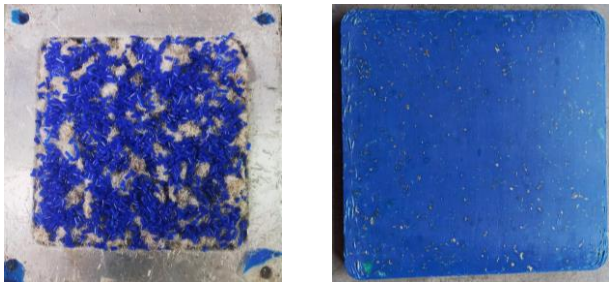

Fig. 2. Fabrication of Composites

2.3 Structural Analysis

Fourier Transform Infrared (FTIR) Spectroscopy was conducted to reveal the frequency peaks corresponding to different bonds lying within the sample. Details will be discussed in the results and discussion section.

2.4 Mechanical Testing

Tensile, flexural, hardness, and impact tests were conducted for each set of composites. Three samples from each set of composites with different fiber loading were prepared for each test. The average of the obtained results is reported in the later part of the corresponding study. Tensile tests were carried out according to ASTM D 638-01 using an Instron machine (system Id3369J8567, maximum capacity 50 kN). The three–point flexural test was performed using the same machine following ASTM D 790–98. The impact test of the composite was conducted using an impact tester MT 3016, and the specimen was prepared according to ASTM D 6110–97. Hardness was measured using a Shore durometer in its D scale.

2.5 Thermogravimetric Analysis

Thermogravimetric analysis was carried out in a thermo gravimetric analyzer of model TGA Q50 W/FMC, where nitrogen gas was used. Its temperature range is ambient +5 ℃ to 800 ℃, and the heating rate is 0.1 to 100 ℃/min. A temperature range of 25 ℃ to 500 ℃ and a constant heating rate of 10 ℃/min was used in the present research. TGA curves of each sample were obtained to understand better thermal degradation and the number of steps involved.

2.6 Biodegradability

The biodegradability behavior test was conducted to determine the biodegradability of the manufactured hybrid composites using compost soil. Polypropylene (PP) is manufactured from long chains of hydrocarbons impervious to breakdown by microbes. Biodegradability refers to the ability of a substance to decompose spontaneously in a nonhazardous process. Hence, PP cannot be referred to as biodegradable because it requires 20 to 30 years to break down and release poisons. One of the purposes of this study was to induce some biodegradability in PP by reinforcing it with natural fiber (Wool, WH fibers) along with modified mechanical properties.

In this test, ASTM D6003-96 was used as the standard method to determine the weight loss of manufactured composites. The composites' biodegradability can be evaluated by calculating the weight loss of the composites at 10-day intervals after burying them in natural compost soil for 30 days.

3. Results and Discussion

3.1 Tensile Properties

Tensile properties of the composite samples were measured for each fiber content (0, 5, 10, and 15 wt.%) with the help of stress/strain curves and respective equations. The wool and water hyacinth (WH) fiber ratio was set at 1:1. The tensile strength and Young's modulus values of the composites for different fiber loadings are shown in Figure 3.

The tensile strength decreased with an increase in fiber loading. As the fiber loading increased, the interfacial area between the fiber and matrix increased, which was weak because of the worsening interfacial bonding between the cellulose-based hydrophilic filler (WH fiber) and the hydrophobic polypropylene matrix [21]. This consequently decreased tensile strength. Also, not all WH fibers are glued by the matrix due to poor distribution of WH fiber in the matrix to enhance the tensile strength of the composite for further fiber content.

Young's Modulus has gradually decreased with increasing fiber content. This occurred due to a very low aspect ratio of the WH fibers. Such decline in the properties has been caused because of the formation of agglomerates, which block stress transfer [22]. Another factor that has contributed to this decline in strength and modulus is the increase of void in the composite during processing due to insufficient wetting of fibers [23]. The macrographic image in Figure 4, obtained from a test sample during preparation, is compelling evidence of voids present within the composite material. It is reasonable to assume that additional voids may exist that are not readily visible to the naked eye, such as those depicted in this illustration.

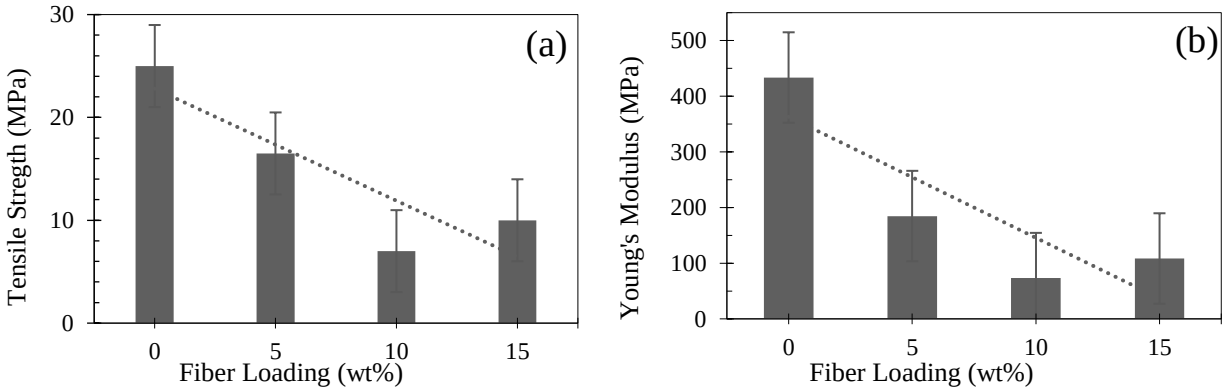

Fig. 3. Variation of (a) tensile strength and (b) Young's modulus against water hyacinth and sheep wool fiber loading

Fig. 4. Voids visible in macrograph

3.2 Flexural Properties

The flexural strength and flexural modulus of sheep wool and WH fiber-reinforced hybrid polypropylene composites are shown in Figure 5. Flexural strength decreased when fiber loading increased from 0 to 5%. Then flexural strength increased up to 10% fiber loading. Such a trend can

be explained through general composite theory, whichegards composites as brittle fibers and ductile–matrix systems [24]. If the fiber content is below its minimum volume faction, the stress on a composite may be high enough to break the fibers [25]. These broken fibers can be treated as voids, which reduce the strength of the composites. Later a significant decrease in flexural strength at 15% fiber represents an excess of the optimum fiber content that the composite can sustain for the proposed dimension. Due to the very high amount of fiber loading, fibers are exposed to the surface from the middle layer penetrating the polypropylene matrix layer and forming a bridge that connects both faces of the composite [26]. When the load is applied, this bridge promotes easy fracture leading to very low strength [27]. For the same reason, the Flexural modulus for 15% fiber loading decreased drastically from 10%. The initial increase in flexural modulus up to 10% represents the typical expectation of fiber loading.

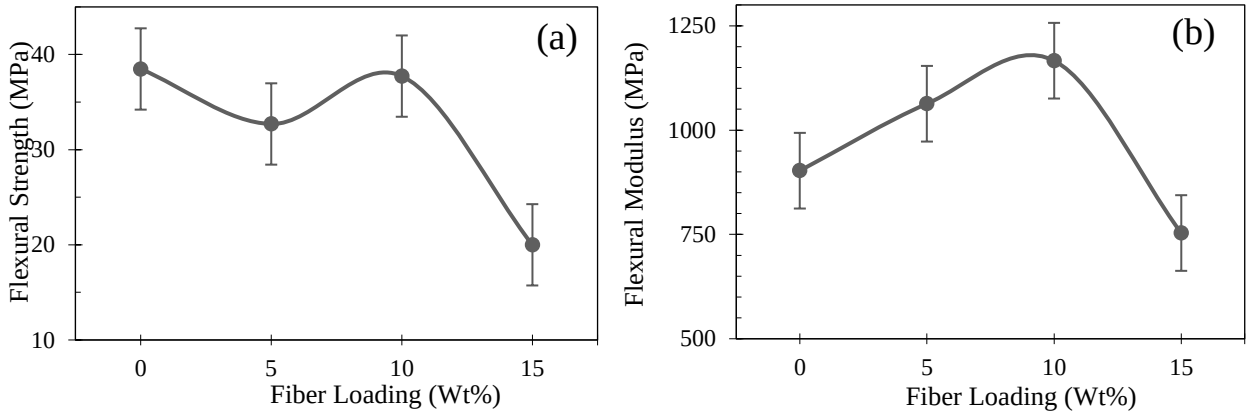

Fig. 5. Variation of (a) flexural strength and (b) flexural modulus against water hyacinth and sheep wool fiber loading

3.3 Impact Strength and Hardness Test Results

Figure 6 (a) shows the variation of impact strength against fiber loading. Due to the incorporation of fiber in the matrix, the void is reduced. Hence, more force is needed to pull out the fiber, increasing the Charpy impact strength [28, 29]. The effect of fiber loading on prepared composites' hardness is shown in Figure 6 (b). The hardness increases when the material's resistance to deformation increases. A composite's hardness depends on the filler's distribution into the matrix. Usually, a flexible matrix causes the resultant composites to exhibit lower hardness. Hardness is maximum at 5% fiber loading, then decreases to 15% fiber. Hardness increased to 5wt. % because incorporating fiber into the polypropylene matrix reduced its flexibility, resulting in a more rigid composite. With 10% and 15% fiber loading, hardness decreased because of the poor dispersion of the fiber into the matrix with more voids between the matrix and fiber.

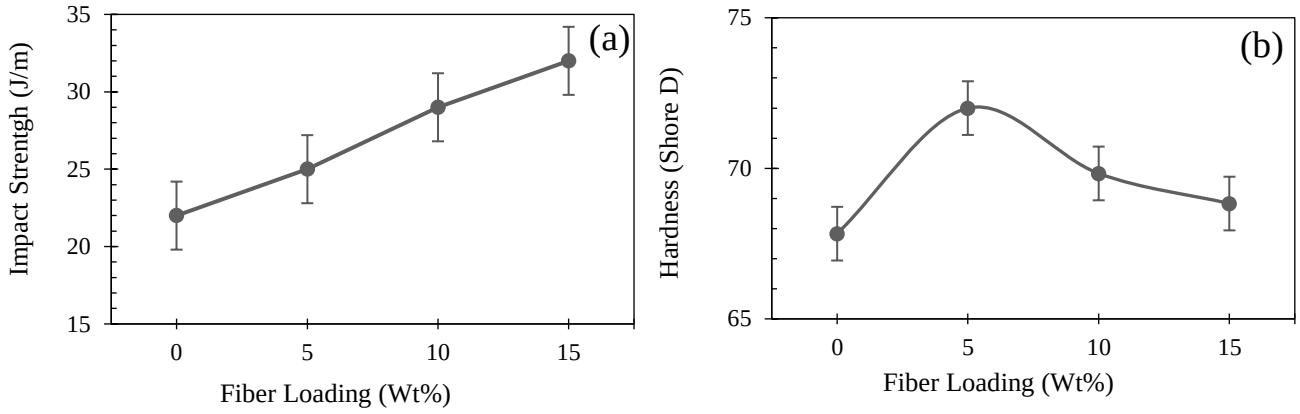

Fig. 6. Variation of (a) impact strength and (b) hardness against WH and sheep wool fiber loading

As overall mechanical property degraded significantly from 10% fiber loading, the maximum fiber loading for the study was limited to an optimum level since the further increase of fiber content would only worsen the mechanical properties of the composite.

3.4 Thermal Properties

For each of the prepared composites, one set of TGA samples was provided for analysis and the results are represented in Figure 7. It is evident that with the addition of fiber, thermal degradation commences at a higher temperature than the unreinforced polymer. The sample containing no fiber began to degrade at 200°C, while the initiation of degradation at 5% fiber loading was 230°C, further increased to 250°C at larger addition of reinforcements. Furthermore, the termination temperature of degradation also shifted to a higher value. The degradation ending temperature of the sample containing 0% fiber was found to be 450°C, reaching a maximum of 525°C for 15% loading. Thus, by the addition of reinforcement, the thermal stability of these composites has improved.

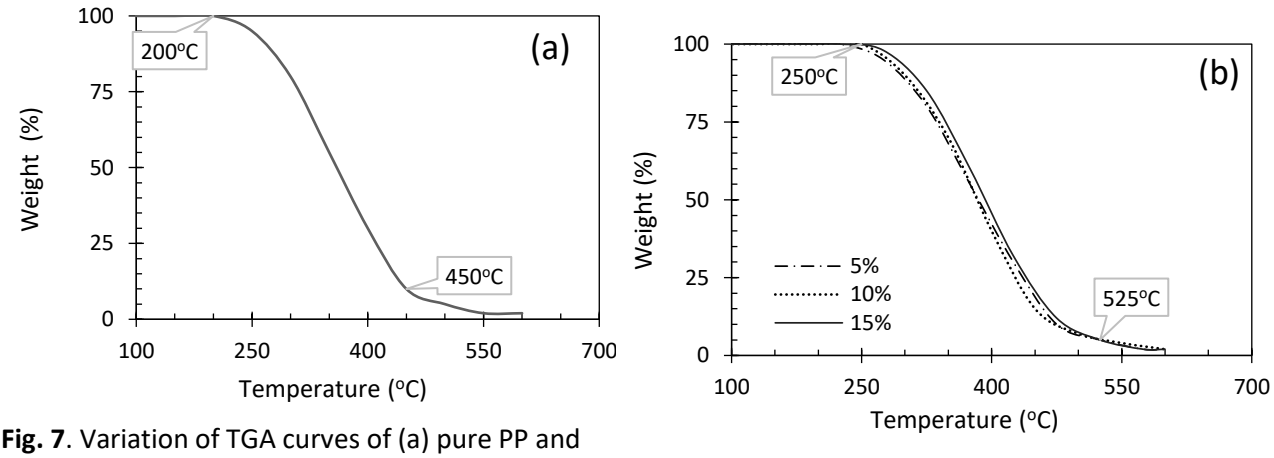

Fig. 7. Variation of TGA curves of (a) pure PP and
(b) Water hyacinth and sheep wool fiber reinforced PP composites

3.5 Biodegradability Properties

The weights of the samples of each type at 10-day intervals are represented in Figure 8. It is very much expected that the higher the fiber content, the larger degradation will take place since the amount of plastic (PP) decreases with increasing fiber content. This is also evident here as the weight of 15% fiber-reinforced composite decreased at the fastest pace to the lowest value compared to 10% and 5% reinforcement.

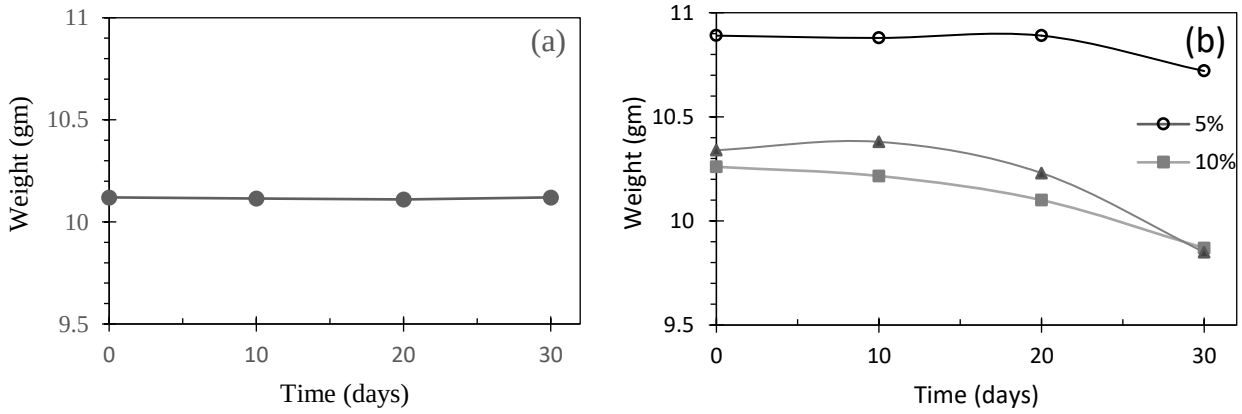

Fig. 8. Variation of biodegradation of (a) pure PP and (b) Water hyacinth and sheep wool fiber composites against time

3.6 Structural Analysis

The FTIR spectrum of the PP matrix composites with 15 wt. % fiber loading (Wool: WH fiber = 1: 1) is shown in Figure 9. The spectra of other composites showed a similar trend. The spectra of all samples showed characteristic C–H stretching vibration around 2915 cm^{-1}. Three vibrational bands are unique to all sets of composites at 2847, 1461, and 1017 cm^{-1}, with the former band attributed to C-H stretching and O-H stretching bond structure that contains a functional group of alkanes (cellulose and lignin) and carboxylic acids.

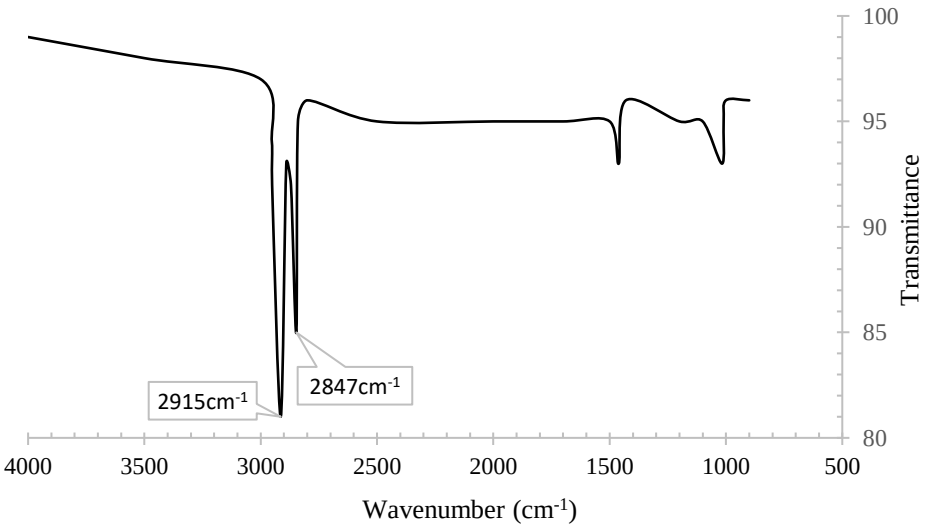

Fig. 9. FTIR spectra of 15% WH and sheep wool fiber reinforced PP composites

The peak at 1461 cm^{-1} is characterized as a C-H bending bond from the functional group of alkanes (cellulose, hemicellulose, and lignin). The peak at 1017 cm^{-1} is characterized as a C-O stretching bond structure from the functional group of alcohol (cellulose, hemicellulose, and lignin), carboxylic acids, esters, and ethers.

4. Conclusions

Water hyacinth fiber and sheep wool fiber reinforced polypropylene composites were created in the current work with three different fiber loadings (5, 10, and 15 wt.%). The impact of fiber loading on structural, thermos-mechanical, and biodegradable characteristics was later examined. The impact strength of the composites showed a tendency in the opposite direction from tensile strength, which decreased as fiber loading increased. After adding reinforcement, all mechanical properties except hardness slightly deteriorated. The bonding between the fiber and the matrix was demonstrated by the migration of recognizable peaks and the emergence of new peaks during the Fourier transform infrared spectroscopy examination. At maximal fiber loading, the thermal degradation temperature of the composites improved. On the other hand, combining fibers derived from plants and animals significantly increased biodegradability. Since the mechanical properties kept degrading drastically with increasing fiber content, the maximum fiber loading was optimized at 15%. Although some mechanical characteristics deteriorated at 15%, the degradation was within a considerable limit. Therefore, the 15% fiber-loaded composite was discovered to be a suitable contender for applications involving moderately high temperatures and environmental friendliness.

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