Agro-fiber Reinforced Polymer Matrix Based Composite: Its Prospects in Bangladesh

A Thesis Submitted to the Department of Applied Chemistry and Chemical Engineering, University of Dhaka, in Partial Fulfilment of the Requirements for the Degree of Doctor of Philosophy (PhD)

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I dedicate this thesis to my parents and to my family

CANDIDATE'S DECLARATION

I, Pinku Poddar, Department of Applied Chemistry and Chemical Engineering, University of Dhaka, declare that the research work which is being presented in the PhD thesis, entitled, "**Agro-fiber Reinforced Polymer Matrix Based Composite: Its Prospects in Bangladesh**" is my own work and carried out by me under the supervision of Prof. Dr. A. M. Sarwaruddin Chowdhury, Professor (Grade-1), Department of Applied Chemistry and Chemical Engineering, Faculty of Engineering and Technology, University of Dhaka, Dhaka-1000, and Dr. Husna Parvin Nur, Ex-Director, Bangladesh Council of Scientific and Industrial Research (BCSIR), Dr. Qudrat-I-Khuda Road, Dhanmondi, Dhaka-1205. The matter embodied in this thesis has not been submitted for the award of any other Degree or Diploma of the university or other institute of higher learning.

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SUPERVISOR'S CERTIFICATE

This is to certify that the thesis entitled, "**Agro-fiber Reinforced Polymer Matrix Based Composite: Its Prospects in Bangladesh**" is being submitted by **Mr. Pinku Poddar** to the University of Dhaka, Dhaka-1000 for examination in fulfillment of the requirements for the award of the degree of Doctor of philosophy (PhD) in Applied Chemistry and Chemical Engineering, embodies the original research work carried out by him, registration number: 22/2014-2015 under our joint supervisions and guidance and has not been submitted in part or full for any degree of this or any other university. The work presented in this thesis is true and original.

It is also certified that the candidate **Mr. Pinku Poddar** fulfills all the requirements as laid down by the University for the purpose of submission of PhD thesis.

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ABSTRACT

Abstract

Due to the environmental concern, renewable materials have gained great desirability as reinforcement in polymer-fiber composites. Agro-fiber such as areca nut leaf sheath (ALS), coconut stem fibers (CSF) and coconut leaf sheath (CLS) short fiber reinforced polypropylene (PP) composites were produced by compression molding technique. Different formulated composites were prepared using 5, 10, 15, 20 and 25 wt % of ALS, CLS and CSF fibers. Water absorption property of the fibers were examined using water uptake test and evaluated strong hydrophilicity by gradually absorbing water until it reaches to a constant water absorption value. Fourier-transform Infrared Spectroscopy was used to identify structural feature of the composites and matrix material. Tensile strength, bending strength, tensile modulus, bending modulus and elongation-at-break etc. mechanical tests of the composites were investigated. Reinforcement property of fibers significantly enhanced the mechanical properties of the composites, and 10 wt% fiber based composites showed the optimum value of strength and stiffness. Chemical composition of the fibers was found using chemical treatment. Maximum α-cellulose contained ALS fiber was also used with LDPE matrix material and highest strength and other properties were showed for 5% fiber content. Again, alkali treatment and different doses of γ-irradiation was applied on ALS fiber, hybrid matrix (polyvinyl chloride & Polypropylene), and fire retardant chemicals were used with ALS fiber reinforced polypropylene composites. 1% nano ALS cellulose contained gelatin-PVA film showed highest mechanical properties. Thermal properties of the films and composites and degradation nature of the composites were analyzed. Presence of 1% nano ALS cellulose in gelatin-PVA film and addition of fibers in the composites lessen the thermal stability. Up to six months in soil medium at ambient condition, degradation test of the composites were investigated.

Keywords: Polypropylene, Low Density Polyethylene (LDPE), Composite, γ-irradiation, Mechanical properties.

LIST OF ABBREVIATIONS

ALS	Areca nut Leaf Sheath
CLS	Coconut Leaf Sheath
CSF	Coconut Stem Fiber
PP	Polypropylene
LDPE	Low Density Polyethylene
PVA	Polyvinyl Alcohol
PVC	Polyvinyl Chloride
BBS	Bangladesh Bureau of Statistics
FTIR	Fourier-transform Infrared Spectroscopy
ATR	Attenuated Total Reflectance
TS	Tensile Strength
BS	Bending Strength
TM	Tensile Modulus
BM	Bending Modulus
Eb	Elongation at break
SEM	Scanning Electron Microscopy
TGA	Thermo Gravimetric Analysis
DSC	Differential Scanning Calorimetry

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CHAPTER 1

1.1 Introduction

The world is now going towards natural things. Considering the environmental impact of natural products, the policies of environment of developed countries are increasing pressure on producers and researchers. Consequently, the fascination of polymer–based composites is growing where natural fibers are used as a reinforced material mostly owing to its renewable origin [1,2]. Polymer matrix composites are now-a-days finding widespread applications in chemical industries and many other areas as structural materials [3]. Recently, a great interest has been originated in the use of agro-fiber as substitute for synthetic fiber (e.g., carbon and glass) in the development of polymer matrix composites due to their more favorable properties such as low density, non abrasive, stiffness, high specific properties and non-brittleness. The environment as well as economic advantages such as biodegradability, abundance all over the world, employment generation, agricultural economy, cheap, power-efficient and application are also among its advantages [4,5]. Throughout the past two decades, natural fibers have been utilized in numerous applications including consumer goods, low-cost housing, automotive, aircraft, paper industries, textile, building and construction industries [6]. Extensive studies haves been carried out on lignocellulosic fibers based on sisal [7-9], pineapple [10-12], banana [13-15], bamboo [16], vakka [17], date [18], and oil palm empty fruit bunch fibers [19, 20] and the results represented that lignocellulosic fibers can be used as the effective reinforcing agents. This is because of its several advantages such as abundance, renewability, biodegradability, environmentally friendly, high strength-to-weight ratio, and low cost compared to their synthetic counterparts.

Areca palm (*Areca catechu,* Linn.) and coconut palm (*Cocos nucifera*, Linn.), valuable tropical resources for food and energy, are plenteous in the coastal districts of Bangladesh. The CSF, CLS, coir fibers are obtained from coconut palm and ALS fibers are generated from areca palm, are being used to make a variety of materials including carpets and mats. However, there are many other parts of the palm other than areca nut leaf sheath fibers, coir fibers, and coconut stem fibers from which fibers can be easily extracted but are not currently utilized as resources for materials in an organized way. In fact, all of these fibers are currently used as fuel materials in the rural area, which does not take advantage of their high length to diameter ratio. Some of these fibers occur in nature already in woven form e.g. leaf sheath [Figure 4.1(a), 4.1(b), 5.1(a) and 5.1(b)] and stem from e.g. stem fiber [Figure 2.1(c) and 6.1] which can be readily impregnated with plastics or cement. In order to find better utilization of CSF, CLS and ALS particularly as fillers or reinforcement in relatively low cost applications (where high strength or high modulus is not required) it is essential to assess the properties of fiber-reinforced composites. Hence, the processing of the naturally woven leaf sheath and stem fibers has been executed into short fibers for thermoplastic polymer reinforcement.

As such, sandwiching this fiber with thermoplastics could offer a strategy for producing advanced composites based on end-use requirements within a framework of low cost, recyclability, availability, bio-degradability and environmental consideration. Moreover, matrix materials are the most significant part of a composite. Polypropylene (PP) is an amorphous thermoplastic polymer and is widely utilized as engineering thermoplastic because of several essential and useful properties such as transparency, dimensional stability, flame resistance, high heat distortion temperature, and high impact strength. PP is also very acceptable for filling, reinforcing, and blending. PP with fibrous natural polymers of biomass origin is one of the most promising routes to produce natural-synthetic polymer composites [21-27]. In this research work, we present the fabrication of naturally areca nut leaf sheath as well as woven coconut leaf sheath and coconut stem fibers short fiber-reinforced polypropylene composites. The structural, mechanical, thermal and degradation properties of the composites are studied to find out its potential towards diversified applications ranging from automotive to consumer products within the environmental legal framework. Again chemical treatment, gamma irradiation, hybrid matrix (polyvinyl chloride & Polypropylene), another matrix material (LDPE) and fire retardant chemicals are used on comparatively better quality of fiber among three types of fibers for enhancing the composite quality. Nano ALS cellulose is also prepared and used as filler in the gelatin-PVA film. The aim is to find out the better quality of fiber and the optimum fiber volume fraction in the composite to maximize the mechanical, thermal and morphological properties, reveal trends of different properties after using different treatment.

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CHAPTER 2

Literature Survey, Aim of the Research Work and Prospects in Bangladesh

2.1 Literature survey

At first, composite term was used in 1500 B. C in the world. Long-lasting and strong constructions were prepared by Mesopotamian and Egyptians manufacturers and builders using a mixture of straw and mud. Straw was commonly used to provide reinforcement in the ancient composite products such as boats and pottery. Concrete structure was started using different types of mortars and lime in 25 B. C. Manufacturers, builders, engineers and artisans were tried from the first time to develop the usages of the composites materials in a various field.

Composite bows were firstly prepared using cattle tendons, horns, bamboo, wood and silk bonded with resinous materials by Mongols in 1200 AD. Composite item was developed after using chemical ingredients in 1870-1890. Polymerization process was applied and synthetic resins were converted liquid state to solid state in a cross-linked molecular structure. Actual development of composite materials was started after the development of plastic materials (Kamrun N. Keya and *et al.*, 2019).

In the plastics industry remarkable amounts of fillers and reinforcements are being consistently applied every year. The utilization of these supplements in plastics is probably to increase with the initiation of advanced coupling agents as well as upgrade compounding technology that allow the utilization of excessive reinforcement or filler content (Katz and Milewski, 1987).Polystyrene, phenolic, vinyl and polyester etc plastic materials were firstly applied as reinforced in 1900. Using synthetic chemicals the plastic Bakelite was prepared and performed better comparatively other composite materials. But in case of structural utilization, plastic materials could not able to provide sufficient strength and toughness of the structure. At first, Owens Corning introduced the glass fiber in 1935 and applied in the fiber reinforced polymer (FRP) industry. Fiberglass with plastic materials produced a surprising strong structure. In 1936, UPR (Unsaturated Polyester

Resins) were patented and different higher quality resins were started to utilize from 1938 (May-Pat A. and *et al.*, 2013 and Uddin N, 2013). In 1992, an outstanding review on the technology of short fiber composite by Milewski comprises a variation of causes that outcome in complications correlated with the properties of the composites. In case of the different properties of the fiber matrix composites, interaction between fiber and matrix, dispersion of fibers, hydrophilic nature of fibers and hydrophobic nature of matrix materials play an important role. Nowadays, composites are classified into two categories such as green composite and partly eco-friendly composites. When all constituents of composites e.g. reinforced and matrix materials are originated from renewable sources is called green composite. Man-made fiber or synthetic fiber with bio-polymer matrix material and natural fiber with petroleum derived non biodegradable matrix material based composites are known as partly eco-friendly composites. Whereas both composites are called bio-composites and scientists are more concentrated on it (Pecas, P. and *et al.*, 2018).

Nowadays, composite materials are utilized in various aspects as well as dependency of people on it is gradually increased day by day. Natural fibers are very cost effective, renewable, strong, eco-friendly, biodegradable, sustainable and it has good properties than that of synthetic fibers (May-Pat A. and *et al.*, 2013). ALS, CLS, CSF, jute, sugarcane, sisal, hemp, kenaf, banana, coir, bamboo, flax etc. natural fibers are renewable, recyclable, nonabrasive and provide better mechanical properties compared to synthetic fibers (Nguong CW and *et al.*, 2013). Natural fibers are suitable to use as reinforced materials in both thermoplastic and thermosetting composites. Epoxy, polyester, unsaturated polyester resin and phenolic etc. thermosetting resins are widely used for producing composite materials and natural fibers showed better performance.

Due to good characteristics e.g. low density, high strength and ecological advantages more attention are growing on natural fibers among researchers, students, academicians and also in manufacturers as well as industry. Again, because of the non-carcinogenic and bio-degradable properties of natural fibers based composites, the uses of these composites are increasing day by day. About 2.1 billion US dollar natural fiber reinforced composites used in different industry and it is expected 10% of the use of natural fibers in composite sectors will be increased after 5 years. Though natural fiber has a lot of advantages (e.g. low cost, low density, renewable etc.), it has some disadvantages such as higher moisture absorption property. For this reason chemical treatment is important to reduce moisture absorption. Consequently, the mechanical properties of the natural fiber based composites are significantly affected by several factors e.g. fiber-matrix adhesion, fiber length and fiber aspect ratio etc. (Kamrun N. Keya and *et al.*, 2019). UV and Gamma radiation is also used to increase the mechanical properties due to the intercross-linking between the neighboring cellulose molecules of natural fiber reinforced composites (Haydar U. Zaman and *et al.*, 2010). The researchers, academicians and R & D of natural fiber based composite manufacturers have been studying for thousands of years to develop the mechanical and thermal properties as well as to increase the acceptability and to solve the limitations of the composites.

2.2 Advantages of polypropylene (PP)

PP is an inexpensive material that largely provides a collaboration of significant mechanical, thermal, electrical, physical and chemical effects not appeared in any other thermoplastic. It possesses an outstanding degreasing agent, electrolytic affect and resistance to organic solvents. It exhibits minor impact strength, but its tensile property and working temperatures are higher to low or high density polyethylene. In fact, it is light in weight, against to staining, and has a low moisture absorption rate. Thus, this is a durable, semi-rigid material, heat-resistant, ideal for the transfer of hot liquids or gases. It is suggested for vacuum systems and where higher pressures and heats are encountered. It has outstanding resistance to alkalis and acids, but tinny aliphatic, aromatic and chlorinated solvent resistance.

2.3 Advantages of Low Density Polyethylene (LDPE)

The advantages of using LDPE are-

1. Low cost,
2. Impact resistant from -40°C to 90°C,

3. Moisture resistance,
4. Good chemical resistance,
5. Food grades obtainable,
6. Readily processed by all thermoplastic methods.

2.4 Classification of natural fibers

Natural Fibers

Plant

- **Stalk** e.g.: Straw (Cereal)
- **Grass** e.g.: Bagasse, Bamboo
- **Stem** e.g.: Jute
- **Fruit** e.g.: Coir
- **Leaf** e.g.: Banana, ALS, CLS, CSF
- **Root** e.g.: Broom root
- **Seed** e.g.: Cotton

Animal

- **Hair**
- **Wool**
- **Silk**

2.5 Agro-fibers used in this research work

Coconut Leaf Sheath, Coconut Stem Fiber and Areca nut Leaf Sheath are used in our research work.

(a) (b) (c)

Figure 2.1: (a) ALS, (b) CLS, (c) CSF.

2.6 Composition of natural fibers

Natural fibers mainly contain cellulose. Carbon (44.44%), hydrogen (6.17%), and oxygen (49.39%) are present in cellulose. The chemical formula of cellulose is

$(C_6H_{10}O_5)_n$ where 'n' represents the number of glucose groups and meaning of 'n' is the degree of polymerization. The chemical structure of cellulose is

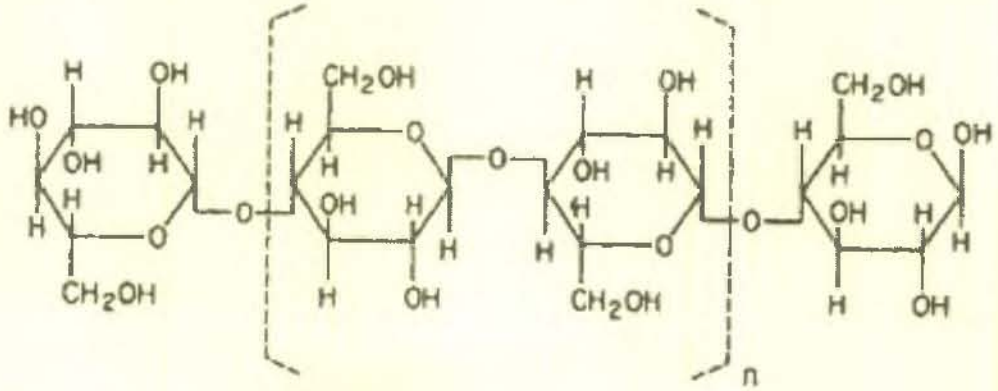

Figure 2.2: Chemical structure of cellulose.

Natural fibers also contain pectic matters, lignin, hemicelluloses, aqueous extract, fatty and waxy matters.

2.7 Benefits of applying natural fibers in composites

Natural fibers are used as reinforced materials in the composites. The benefits of applying natural fibers in composites are-

2.7 (a) Characteristics advantages

1. Renewable
2. Low density
3. Strong
4. Non abrasive
5. Lightweight
6. Sustainable
7. Resilient

2.7 (b) Ecological, social and economic advantages

1. Eco-friendly
2. Bio-degradable
3. Employment generation
4. Power-efficient
5. Agricultural economic
6. Cheap

2.8 Aim of the research work

Cocos nucifera and *Areca catechu* trees are frequently obtainable in the area of coastal in Bangladesh which provides enormous amount of stem fibers and leaf-sheath. These unusable and agro-waste fibers can be suitable elements in producing the composite materials and films from nanocrystalline cellulose. The ultimate aim of this research is to produce plastic like composite materials and renewable nanomaterial films with the help of these wastage materials which will be environmentally friendly usage in packaging, housing, shipping, aircraft, automotive, building and construction industries etc.

2.9 Prospects in Bangladesh

In Bangladesh, betel nut and coconut are cultivated almost all over the country whereas it is especially in the homesteads and farmlands of South-Eastern region (Nandi and *et al*., 2019). Weather conditions are favorable for the cultivation of *Areca catechu* and *Cocos nucifera* trees in most places in Bangladesh. For betel nut crop in 2017-2018 fiscal years, area of under garden and area of outside garden was 76,531.00 acres and 43,955.00 acres respectively (BBS 2018). Total area 1,20,486.00 acres has been considered for the betel nut crop. The average yield of areca nut leaf sheath fiber is about two-third of betel nut production of per tree per year. Total production of betel nut including inside & outside garden was 2,15,783.00 metric tons in 2017-2018 fiscal years (BBS 2018). Areca nut leaf sheath (ALS) fiber production was about 1,43,855.33 metric tons. According to Bangladesh Bureau of Statistics-2018, coconut was cultivated over 1,32,202.00 acres of land including area of under garden 10,365.00 acres and area of outside garden 1,21,837.00 acres which produces 4,66,975.00 metric tons of coconut in 2017-2018 fiscal years. Annual, average yield of coconut stem fiber and coconut leaf sheath is about 75% and 5% of total coconut production of per tree respectively. So, coconut stem fiber and coconut leaf sheath production was about 3,50,231.25 metric tons and 23,348.75 metric tons respectively in 2018.

Coconut leaf sheath, coconut stem fiber and areca nut leaf sheath are useful and abundant materials in nature. Generally, these agro-fibers are considered as agro-wastes which are randomly collected by villagers in rural area and used for fuel materials and most of the unused agro-fibers are become a source of environmental pollution and waste generation. These useful materials can be handled easily by applying technology and method. So, fibers can be collected properly and scientifically and used in agro-fiber based industries for multidimensional purposes. This sector would have a great impact on macro-economic circumstances like employment generation, poverty alleviation etc.

CHAPTER 3

Structural and Morphological Characterization of Coconut Stem Fiber (CSF), Coconut Leaf Sheath (CLS) and Areca nut Leaf sheath (ALS)

3.1 Abstract

Every year a large amount of agro waste materials are generated all over the world. Proper management of agro waste is a great challenge. At first, it is important to know the characteristics of the waste items as well as the natural fibers. A perceptible and morphological analysis for three types of fibers (coconut stem fiber, coconut leaf sheath and areca nut leaf sheath) has been investigated in this chapter. In addition, FTIR studies of the fibers were executed for detecting the structural characterization in the region between 4000-620 cm^{-1}. Chemical compositions of the fibers were investigated by chemical analysis. Morphological investigations were also done to identify the surface characteristics of the fibers.

3.2 Introduction

Agro-fibers are significantly being used as the reinforcements in composite materials. Its applicability has been dramatically increasing for hundreds of years [1]. Billion of tons of agricultural waste materials and agricultural crop residues are produced annually in the world. For fertilizer and household fuel purposes a little amount is used and major portion is burned and some amounts are allowed to rotten here and there. It is responsible carbon emission as well as air pollution. To overcome the situation, researchers, students, academicians and R & D sections of manufacturers are concentrated on it. The agro-fibers and agro waste can be used as reinforced material in both thermoplastic and thermoset composites and it will be economical and fruitful way to control environmental pollution. Comparatively natural fibers are better than synthetic or man-made fibers because of some properties such as low cost, renewable, biodegradable, eco-friendly, low density, light weight etc. [2-4]. The utilization of natural fiber based composites are increasing day by day due to the finished products are non-carcinogenic and biodegradable. Based on source, natural fibers are classified into two types such as plant fibers and animal fibers [5]. Coir, jute, sisal, kenaf fiber, flax, abaca, bamboo fiber, pineapple leaf fiber, bagasse, hemp etc natural fibers based composites are already utilized in different engineering purposes.

Coconut stem fiber, coconut leaf sheath and areca nut leaf sheath natural fibers are not used in composite material or any other engineering applications. Every year a large amount of these fibers are produced but a little amount is used for household fuel purposes and major unused portion is known as agricultural waste materials [6]. In this study, the structural and morphological properties of the fibers were investigated. These agricultural waste fibers can be used for preparing composite materials which will have a lot of engineering applications [7].

3.3 Experiment

3.3.1 Fibers collection

Coconut stem fibers and areca nut leaf sheath fibers were collected from coconut stem of *Cocos nucifera* trees (coconut tree) and *Areca catechu* (areca nut tree) respectively. Then the stem was soaked into water for two weeks to obtain coconut stem fibers and areca nut leaf sheath fibers. The water immersion made the coconut stem and areca nut leaf sheath soft and weakened the bond between the fiber and the resin and waxy materials, which eventually helped to peeling off the loosen fibers from the resinous materials. The peeled off fibers were cleansed using huge amount of fresh water and dried appropriately. Again, the coconut leaf sheaths fibers were collected from the coconut tree and which were found in mat form. Then the fibers were washed with fresh water and dried appropriately.

3.3.2 Chemical analysis

It is important to know the chemical composition of the fibers. At first, the hard portion was discarded and almost uniform fibers [8-9] were taken. Then the fibers were cleansed with sodium hydroxide and soap flake for 60 minutes and washed with fresh water and dried properly. For the identification of chemical composition following steps were followed.

At 60°C temperature, the dried fibers were heated with distilled water for 2 hours and the fibers were isolated by filtration. For constant weight the fibers were dried at 105°C in an oven. The quantity of aqueous extract in the fibers obtains from the calculation of loss of weight.

For 1 gm of dried fiber 100 ml benzene and alcohol was used maintaining 2: 1 ratio. Stirring intermittently and kept to stand for 10 hours. After 10 hours, the mixture was filtered and the fibers were separated from the solution. Then the separated fibers were cleansed several times with fresh benzene-alcohol mixture and with alcohol finally. The fibers were dried at 105°C in an oven for constant weight. The calculation of loss of weight during extraction represents the quantity of waxy matters and fatty matters in the fibers.

The de-waxed fibers were dried and mixed with 0.5% ammonium oxalate solution in a beaker maintaining 100 ml solution for per gm fiber. Then the fiber mixed solution heated for 72 hours in a heating mantle at 70-80°C. Hot distilled water was added all over the procedure to maintain the solution level constant. After 3 days, the fiber mixed solution was filtered properly and the fibers were cleansed perfectly using hot distilled water. Then the fibers were dried and evaluated the amount of pectic matters in the fibers obtains from the calculation of loss of weight.

The de-waxed and de-pectinised fibers were dried at 105°C in an oven and treated with 72% H_2SO_4 (15 ml for 1 gm fiber) at room temperature and stirring repeatedly. Then the mixture was diluted to 3% acid and kept to stand for two hours. The mixture was refluxed and allowed to stand for over-night then sintered funnel was used for filtration. The residual part of the sintered funnel dried at 105°C in an oven represents the quantity of lignin in the fibers.

By the treatment of bleaching agent, for instance sodium chlorite all non cellulosic parts of the fibers were separated but a combination of chlorite hemicelluloses and α-cellulose (combinedly called holo-cellulose) was obtained. For 1 gm of dried de-pectinised and de-waxed fibers, 80 ml 0.7% sodium chlorite solution ($P^H = 4$) was added and stirring the mixtures for 90 minutes at 90-95°C temperature. Then the buffer solution ($P^H=4$) of acetic acid and sodium acetate was added into the chlorite solution to keep the P^H value constant maintaining 1 ml buffer solution for 10 ml of chlorite solution. The solution was filtered and the fibers cleansed properly with fresh water and 2% sodium meta-bisulphite solution was added and stirred the mixture for 15 minutes. Again filtered and cleansed perfectly and the chlorite holo-cellulose was obtained in the sintered funnel and dried at 105°C till constant weight was attained. For every 1 gm of fiber, 100 ml of 24% KOH solution was mixed with the dried chlorite holo-cellulose and allowed to stand for 4 hours with stirring intermittently. Consequently, α-cellulose remained unresolved but hemicellulose went into solution. After filtration, α-cellulose was separated and cleansed with 2% acetic acid solution perfectly and finally washed with fresh water and dried properly. Thus the quantity of α-cellulose is obtained and the quantity of hemicellulose is achieved after the deduction of the weight of α-cellulose from the holo-cellulose.

3.3.3 Fourier Transform Infrared Spectroscopy analysis

ALS, CLS and CSF were investigated by Fourier Transform Infrared Spectroscopy with an attachment of attenuated total reflectance (ATR) accessory. Using a Perkin Elmer instrument the spectra were recorded in the wave number ranges of 4,000-620 cm^{-1}.

3.3.4 Surface morphology

For the morphological analysis of the fibers, SEM is an effective technique. Characteristics morphology of fibers and their shapes are important for using as reinforced materials. The morphology of fiber surface plays an effective role for better interaction between fibers and matrix which exhibits superior tensile properties. Hitachi S-4000 model field emission scanning electron microscope was used to investigate the surfaces of the ALS, CLS and CSF fibers operating at 5 kV.

3.4 Results and Discussion

Table 3.1 represents the chemical composition of the CLS, CSF and ALS fibers. As shown in Table 3.1, the highest amount of cellulose (α – cellulose and Hemicellulose) found for ALS fiber compare with other fibers and it was about

75%. However, CLS fiber contained comparatively maximum amount of lignin. Again, aqueous portion of CSF was highest compare with another fiber.

Table 3.1: Chemical composition of Coconut leaf sheath (CLS), Coconut stem fiber (CSF) and Areca nut leaf sheath (ALS).

Sl. No.	Name	%		
		CSF	CLS	ALS
1	Aqueous Extract	5.52	0.53	0.72
2	Pectic matters	1.75	0.880	1.15
3	Fatty and waxy matters	5.91	2.86	5.06
4	Lignin	22.54	27.97	19.59
5	α – cellulose	46.26	59.39	66.08
6	Hemicellulose	18.02	8.45	7.40
	Total	100	100	100

3.4.1 Structural characterization

The functional groups of the fibers were investigated applying FTIR analysis. Figure 3.1, 3.2 and 3.3 appeared the FTIR record of ALS, CLS and CSF fibers respectively where the bands at around 1734 cm^{-1} and 1248 cm^{-1} corresponded to hemicelluloses [10] and the bands at around 3435 cm^{-1} and 2930 cm^{-1} corresponded to α-cellulose whereas the persisting bands belonged to lignin. The bands of CSF and CLS fibers at around 1734 cm^{-1} and 1248 cm^{-1} are strongly sharp than that of ALS fiber. It may be due to the presence of higher amount of hemicelluloses in the CSF and CLS fibers. However, the bands of CSF and CLS fiber at around 3435 cm^{-1} and 2930 cm^{-1} are not strongly sharp compare with ALS fiber. In this region ALS fiber represents more sharp bands and it may be due to the presence of comparatively higher amount of α-cellulose.

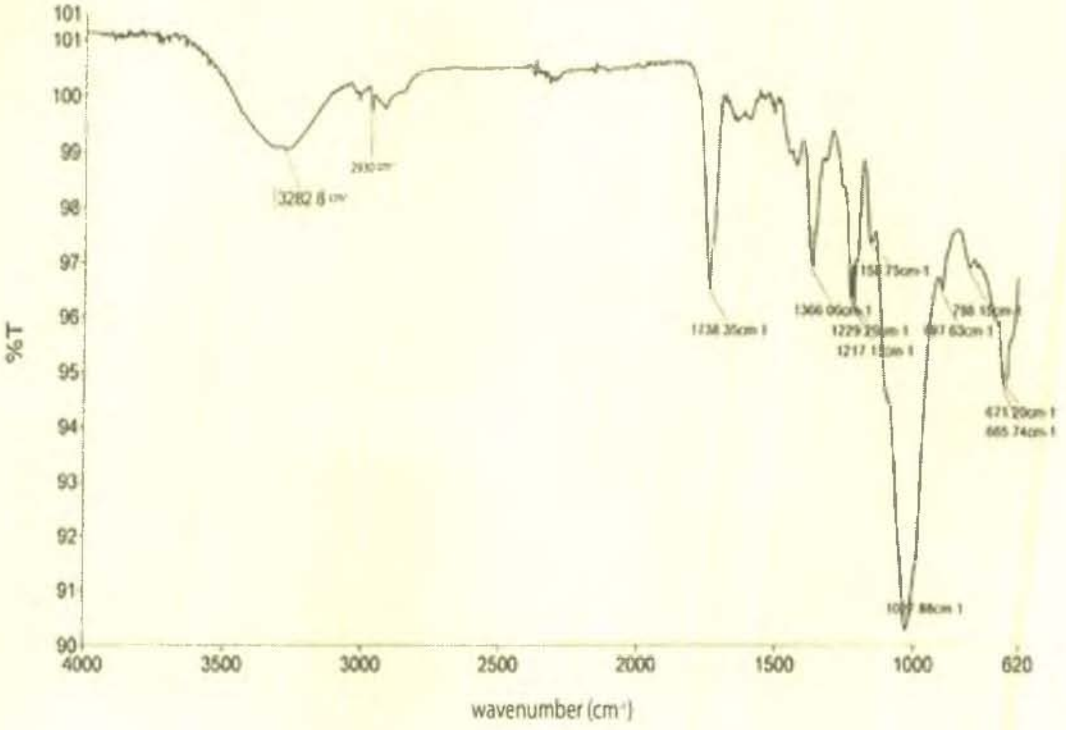

Figure 3.1: FTIR of ALS Fiber.

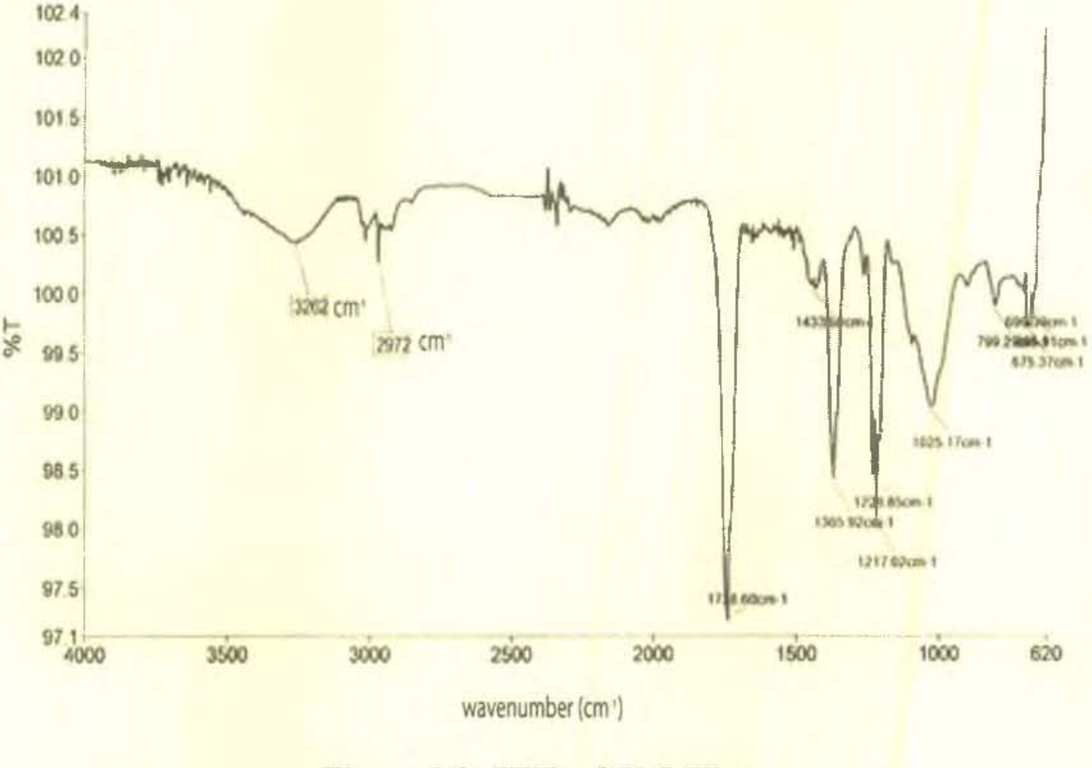

Figure 3.2: FTIR of CLS Fiber.

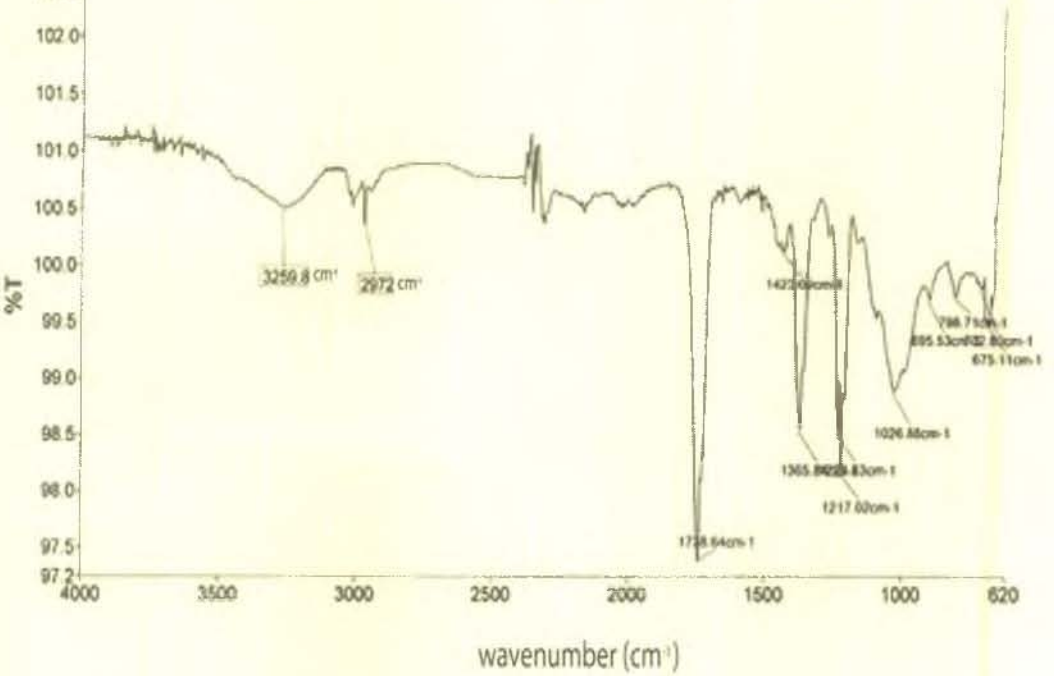

Figure 3.3: FTIR of CSF.

3.4.2 Scanning Electron Microscopic analysis

Surface properties of every fiber are identical as well as morphology is also different. The morphological images of ALS, CLS and CSF are showed in the Figures 3.4, 3.5 and 3.6 respectively and from the interpretation of the figures it is observed that Figure 3.4 is more fibrous than that of the other fibers. It may be due to the presence of comparatively lower amount of lignin in the ALS fiber and which will be responsible for better mechanical properties.

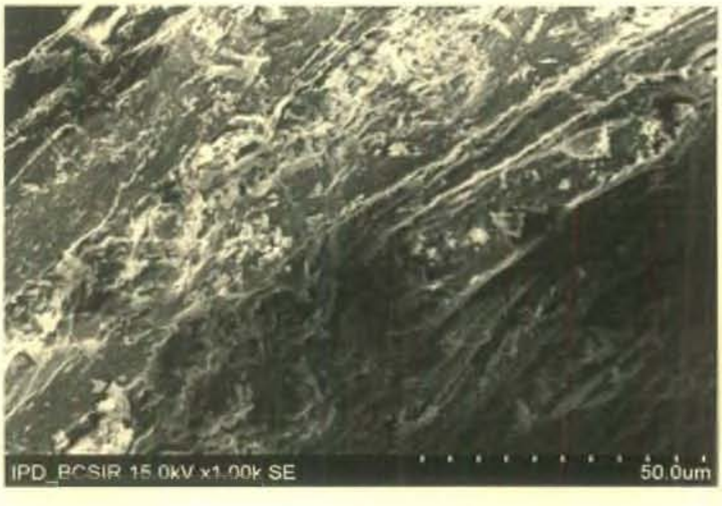

Figure 3.4: ALS Fiber.

Figure 3.5: CLS Fiber.

Figure 3.6: CSF Fiber.

3.5 Conclusion

Some agricultural waste fibers such as CSF, CLS and ALS were examined by chemically and morphologically. Structural characterizations of the fibers were analyzed by FTIR. After chemical analysis, it was found that ALS fiber contained highest amount of α – cellulose and lowest quantity of lignin than that of the other fibers. It will appear better adhesion between the ALS fiber and matrix materials. Morphological studies indicated that ALS fiber was more fibrous and some gaps were found in the coconut stem fiber. Chemical compositions of the fibers were confirmed by FTIR analysis.

3.6 References

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CHAPTER 4

Study of Short Areca nut Leaf Sheath Fiber Reinforced Polypropylene Composites: Mechanical, Thermal and Morphological Analysis

4.1 Abstract

Natural fibers from agro wastes can be utilized as reinforced materials in the composites. Areca nut leaf sheath fibers were used with polypropylene to produce composite materials by compression molding technique. The chemical constituents such as, lignin, α-cellulose and hemicellulose of the fiber was analyzed. Different percentages of ALS fiber (5 to 25 wt%) were used for preparing composites. The mechanical, structural, morphological and thermal properties of the ALS fiber based polypropylene composites were investigated. The water uptake was done to investigate the hydrophilic nature of the composites. In the soil medium, soil degradation test of the composites were performed up to 24 weeks. 10% fiber reinforced composites showed the maximum tensile strength and bending strength and it was considered an optimum fiber content.

4.2 Introduction

Because of the renewable and biodegradable source of agro-fibers, utilization of natural fibers as reinforcing material to produce composites is increasing day by day. People are concern about environment and always trying to choose environment friendly item. PP, relatively a low-cost polymer, offers superior physical, chemical, mechanical, thermal and electrical properties compared to its counterpart thermoplastic polymers [1]. For example, PP shows higher tensile strength and enjoys a higher working temperature compared to low or high density polyethylene [2].

In Bangladesh, *Areca catechu* trees are abundant in the coastal areas - southern and western parts of the country. Beyond the obtaining its fruit, areca nut - an essential element often chewed along with the betel leaf - this tree generates enormous leaf-sheath, which is generally used as fuel for domestic cooking by poor rural people.

Recently, this leaf-sheath has gained interest as a potential candidate for fabricating composite materials for a variety of applications, including interior design, and automobiles, among others [3]. Each year, large amounts of agro waste materials or agricultural crop residues are generated all over the world but very minute amount of them are utilized. Small portion of the agro waste materials are used for fertilizer preparation and for cooking purposes as a household fuel. The major unused agro waste materials are responsible air pollution, water pollution and overall environmental pollution. Researchers, students, academicians and related authorities are always trying to solve the environmental problem as well as to find out the fruitful way about the utilization of the agricultural waste materials [4-7]. Agro waste materials can be used as reinforcement in the composite materials and it is accepted by the researchers and which will be helpful to control the environmental pollution [8-11]. Natural fibers are comparatively better than the synthetic fibers. Utilization of natural fibers is increasing day by day instead of synthetic fibers due to the awareness and consciousness about environment and health are growing in the mind of people.

Natural fibers are renewable, biodegradable and eco-friendly in nature. Besides these the fibers are strong, cheap, sustainable, low density and light weight which increased the acceptance level of natural fibers than that of synthetic fibers. Natural fiber based composites are non-carcinogenic and have no health hazards. Consequently, researchers are more concentrated on natural fiber based composites [12-19]. Abaca, bamboo fiber, bagasse, pineapple leaf fiber, flax, hemp, coir, sisal, kenaf fiber, jute etc. natural fibers are widely used as reinforced materials in the composites. Natural fibers are also classified into two categories such as plant fibers and animal fibers. Plant fibers are renewable, biodegradable and low cost raw materials for composite preparation. Again these fibers are light in weight than that of carbon and glass fibers. Light weight and low density of the natural fibers are beneficial in the different manufacturing sectors. Due to the availability, good mechanical and thermal insulation properties, natural fibers reinforced composites are used various construction fields [20-21].

Areca nut leaf sheath fiber is a natural plant fiber which is produced from areca nut leaf sheath of *Areca catechu* (areca nut plant). Areca nut leaf sheath is one type of agro waste material causes environmental pollution. In the previous years, ALS fibers are not collected and not used in the composite fields. In this study, we would like to utilize the areca nut leaf sheath due to the biodegradability, renewable, eco-friendly, sustainable, low cost, light weight etc. properties of the fibers which will enrich the composite engineering fields on the other hand it will be helpful to control the environmental pollution. ALS fibers reinforced composites are environmentally friendly and can be used for packaging, building, construction and automobile [22-26] etc.

4.3 Experimental

4.3.1 Materials

From Polyolefin Company Private Ltd. of Singapore, polypropylene was purchased. To impart actual and homogeneous adhesion between fibers and matrix, particle size should be smaller. To get small particle (50-60 μm) of PP (figure 4.1, d), using grinder PP granules were grinded. Areca nut leaf sheath (figure 4.1, a) was soaked into water for 15 days to obtain areca nut leaf sheath fibers (figure 4.1, b). The water immersion made the areca nut leaf sheath soft and weakened the bond between the fiber and the resin and waxy materials, which eventually helped to peeling off the loosen fibers from the resinous materials. The peeled off fibers were cleansed using huge amount of fresh and pure water and dried appropriately.

Using hand scissors, the fibers were cut into 2-3 mm size (Figure 4.1, c) and all dirt's were taken out from the fibers with the help of mesh. Again, the fibers were cleansed using fresh and pure water. Then the cleansed fibers were dried appropriately. Before the composites fabrication the fibers were vacuum-dried for 5 hours at 100°C temperature.

(a) (b)

(c) (d)

Figure 4.1: Images of areca nut leaf sheath (a), ALS fiber (b), chopped fiber (c) and PP powder (d).

4.3.2 Chemical configuration of ALS fiber

Areca nut leaf sheath fiber contains α – cellulose, hemicellulose, pectic matters, aqueous extract, lignin and fatty and waxy matters and the amount of these components are shown in the Table 4.1. 7.40% hemicelluloses, 19.59% lignin and 66.08% α – cellulose are present in the fiber. Small particles of cellulose of areca nut leaf sheath fibers are cemented and surrounded together by hemicelluloses and lignin.

Table 4.1: Chemical compositions of ALS fiber.

Sl. No.	Name	%
1	α - cellulose	66.08
2	Lignin	19.59
3	Hemicellulose	7.40
4	Pectic matters	1.15
5	Fatty and waxy matters	5.06
6	Aqueous extract	0.72
	Total	100

4.3.3 Composite preparation

Compression molding was used to make the PP matrix-based composites. A family of five formulations of ALS fiber (5%, 10%, 15%, 20% and 25% fiber) reinforced PP composites was prepared by varying the fiber content as shown in the Table 4.2. All formulations were made using moulds in the heat press under 5 bar consolidation pressure and 190°C temperature for 5 min. The model of heat press was 3856, Carver, INC, USA. After 5 minutes, in a separate press the moulds were cooled down to room temperature under 5 bar pressure. The prepared composite is shown in the Figure 4.2.

Table 4.2: The composition of the different composites.

Formulation no.	Weight percentage of ALS and PP in different composites
F1	5% ALS + 95% PP
F2	10% ALS + 90% PP
F3	15% ALS + 85% PP
F4	20% ALS + 80% PP
F5	25% ALS + 75% PP

Figure 4.2: Finished product (composite).

4.3.4 TS and BS test

Using an electro-mechanical testing machine, mechanical properties of the composites (F1- F5) were performed under a maximum load capacity of 5kN with a gauge length of 20 mm and crosshead speed of 10 mm/min. But, in case of bending properties, the crosshead speed was set at 10 mm/min with a span distance of 40 mm. Tensile and bending tests were performed following DIN 53455 and DIN 53452 standards methods, respectively. Five specimens for each sample were examined in the instrument.

4.3.5 Water uptake

Water uptake analysis was done by soaking the pre-weighed dried F1-F5 composites in the distilled water at 25°C temperature. After recording the initial dry weight, the samples were soaked into water and kept for a fixed period and then removed from water and wiped out the excess surface water using tissue paper, and the wet weight (final weight) was taken. The equation of calculation of water uptake is - Water uptake $= \frac{W_f - W_i}{W_i} \cdot 100$ .... (1)

Where, W_i is the initial weight of the samples, and W_f is the final weight of the samples after fixed time.

4.3.6 Degradation tests of the composites

Cellulosic material undergoes deterioration when buried in moisturized soil (at least 25%). To investigate soil degradation profile of the composites, F1-F5 composites were carried out for six months burying the samples in humid soil. After set time point, the samples were taken out, cleansed with fresh water and dried for 8 h at 80°C temperature. The variation of TS was intermittently observed in order to compute the degradable nature of the composites in this humid condition.

4.3.7 Fourier Transform Infrared Spectroscopy analysis

100% Polypropylene and F2 composite were investigated by Fourier Transform Infrared Spectroscopy with an attachment of ATR accessory. The spectra were recorded in the wave number ranges of 4,000-600 cm^{-1} using a Perkin Elmer instrument.

4.3.8 TGA and DSC analysis

In the material of the composites, the physical changes are occurred due to the effects of temperature [27]. The thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) of the PP and the optimized composites were performed to find out the physical changes. The increasing rate of temperature was 20°C/min under inert nitrogen atmosphere and maximum temperature was 900°C.

4.3.9 SEM analysis

Surface morphology plays an important role in case of composite type materials. For the morphological analysis of the composites, SEM is an effective technique. The strong interaction between fibers and matrix exhibits superior tensile properties. The compatibility and distribution between the fibers and the matrix materials could be found using SEM analysis. S-4000 model of Hitachi brand Field emission scanning electron microscope was used to investigate the fracture surfaces of the tensile specimens of both F2 and F5 composite samples operating at 5 kV.

4.4 Results and Discussion

4.4.1 Mechanical features of the composites

The mechanical properties including, tensile strength, bending strength and elongation at break were calculated, and the results are presented in the Figures 4.3, 4.4 and 4.5. As observed from the Figure 4.3, the highest tensile strength (28.7MPa) was found for F2 formulation. The tensile strength values were progressively reduced with increasing fiber content beyond 10% (such as, 15, 20, and 25 wt% for F3, F4 and F5, respectively). The similar situation was found for bending strength. In the Figure 4.4, F2 composite appeared the highest value (46.9MPa). Interestingly, from F1 to F2 formulation, the value was enhanced, by contrast, the bending strength values were continuously decreased from F2 to F5.

Figure 4.5 clearly showed that percentage elongation at break (%) was decreased with increasing fiber content in the composites. The highest Eb% value (15.2) was obtained for F1 composite and it was 13.8, 11.6, 8.9 and 6.4 % for F2, F3, F4 and F5 composites respectively.

The ratio between fiber and matrix in the composites is very important and the amount of fiber present plays a great role to reinforce the mechanical properties. The highest tensile strength and bending strength was found for F2 formulation of 10% fiber with 90% PP due to the better fiber-matrix adhesion. A declining trend of TS and BS of the composites was appeared above 10 percentage fiber content in the composites, which can be ascribed to the fact that interaction between fiber and matrix increased from F1 to F2 formulations and gradually decreased from F3 to F5 formulations.

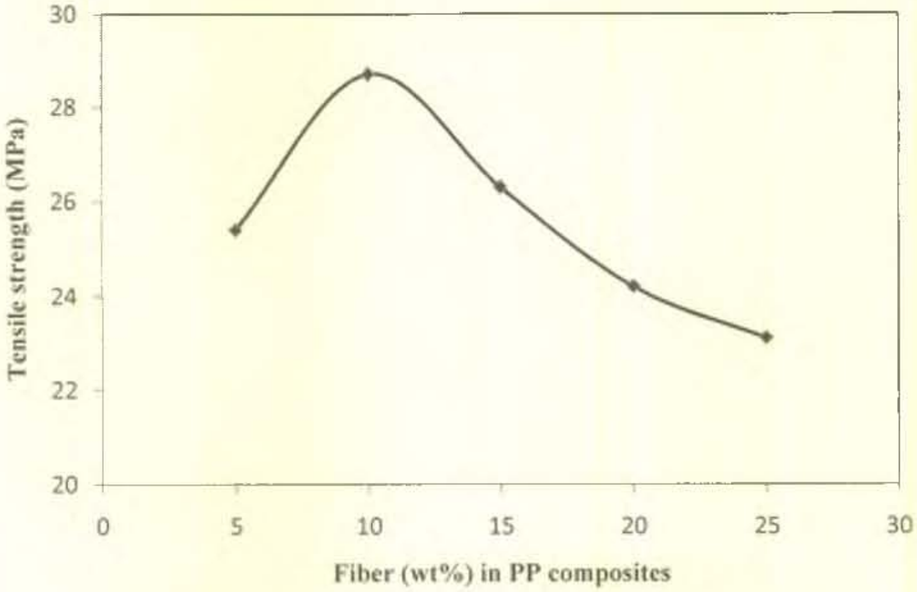

Figure 4.3: TS of different % of fiber content composites.

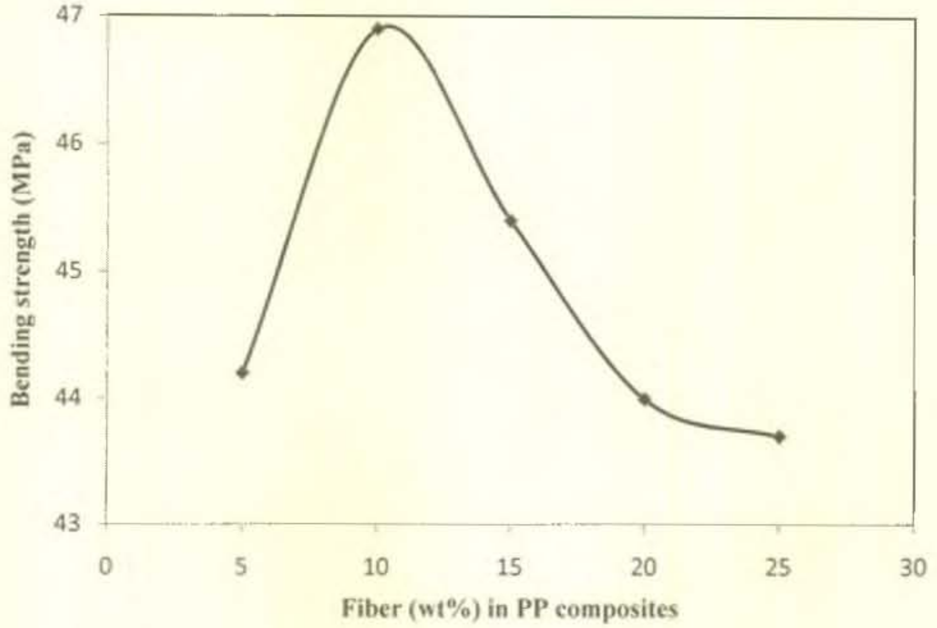

Figure 4.4: BS of different % of fiber content composites.

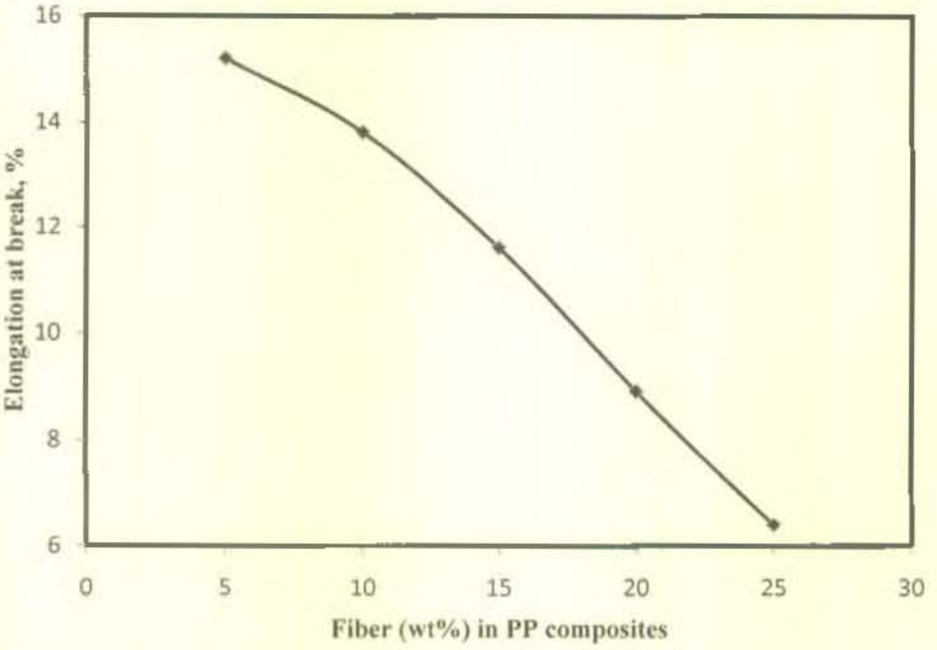

Figure 4.5: Eb of different % of fiber content composites.

4.4.2 Water absorption of the composites

Water absorption values of the F1, F2, F3, F4 and F5 formulated composites were computed. The hydrophilic nature of the fiber influences the water absorption capacity. The subtraction value of initial weight from the final weight indicates the water uptake value. The water uptake profile of all samples is presented in the figure 4.6. For all samples, water uptake initially was gradually increased over time and then reached to a plateau value. For F3, F4 and F5 samples, water uptake reached equilibrium value after 30 h, whereas, F1and F2 samples experienced equilibrium water uptake after 20 h of soaking. The lowest water uptake value was obtained for F1 sample (0.15%), while, the highest value was recorded for F5 sample (0.27%), both after a time period 60 h. The highest water absorption for F5 sample may be due to the higher amount of hydrophilic fiber content. On the other hand, F2 sample showed a nearly minimal level of water absorption (0.15%).

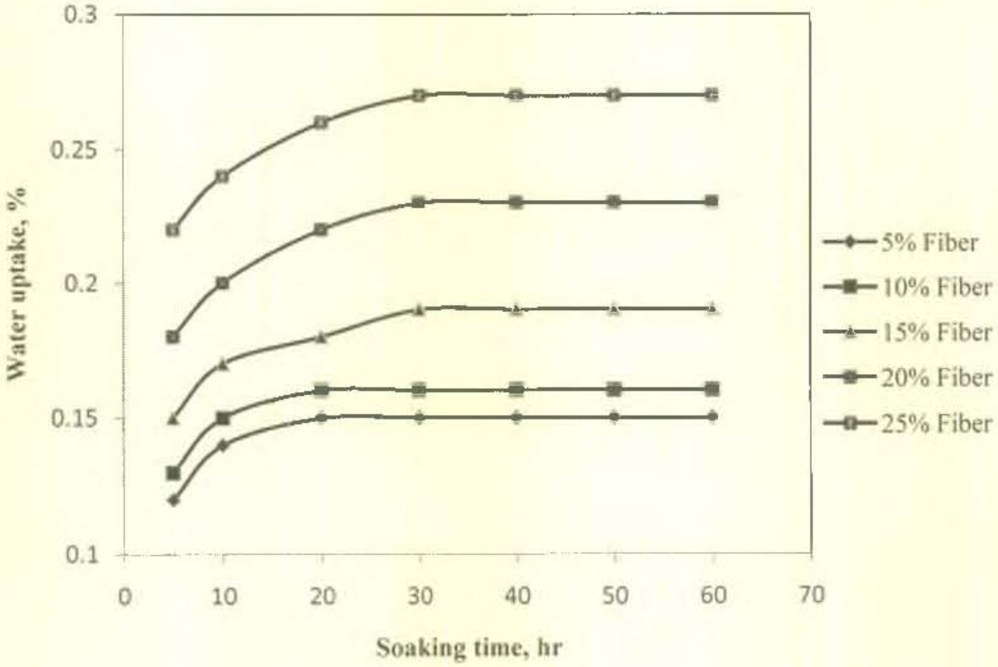

Figure 4.6: Water uptake profile of different composites against soaking time.

4.4.3 Soil burial test

A soil burial test was performed to investigate the degradation trend of the samples in soil media for up to 6 months. Figure 4.7 showed the Tensile Strength values against time (maximum 24 weeks). In case of all samples, Tensile Strength values were declined slowly with degradation time was observed from the above figure. At the end of soil burial experiment, F1, F2, F3, F4 and F5 samples reduced nearly 33, 20, 35, 37 and 42% of TS. It is observed that TS properties of the F2 formulated composites retained more compare to other formulated composites and it may be due to the better interaction between fiber and matrix in the F2 composite. Due to the hydrophilic property and biodegradable nature of cellulose, natural fiber reinforced composites materials degrade in the soil burial experiment as well as such degradation is responsible for the diminished mechanical properties of the composites over time [28].

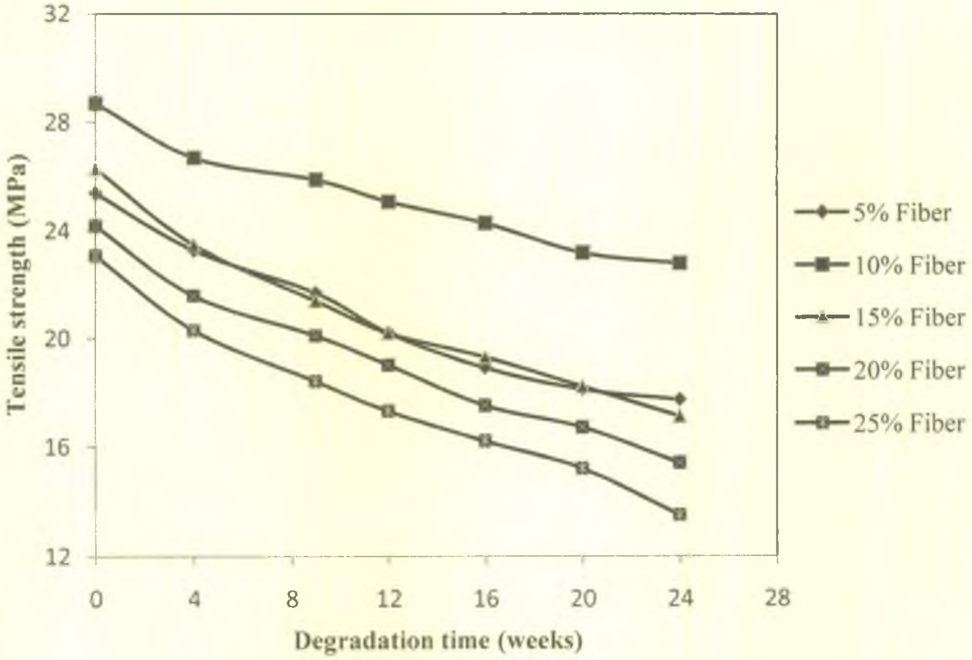

Figure 4.7: Degradation of TS of the composites in the soil medium.

4.4.4 FTIR analysis

FTIR analysis was carried out to calculate the change of surface composition of the optimized composite (F2). The FTIR spectra of pure polypropylene and F2 formulated (optimized) composite were shown in Figure 4.8 and Figure 4.9 respectively. As shown in Figure 4.8, the fingerprint region of pure polypropylene was observed from 1455-600 cm^{-1}, particularly at around 1455 and 997 cm^{-1} ascribed to C=C stretching and C-H bending vibrations, respectively. The peak observed at 2916 cm^{-1} assigned to C-H stretching vibration. In the Figure 4.9, a peak at 1741 cm^{-1} was evident which was assigned to C=O (carbonyl) stretching of acetyl groups of hemicellulose in the natural ALS fiber [29], along with characteristic peaks of polypropylene.

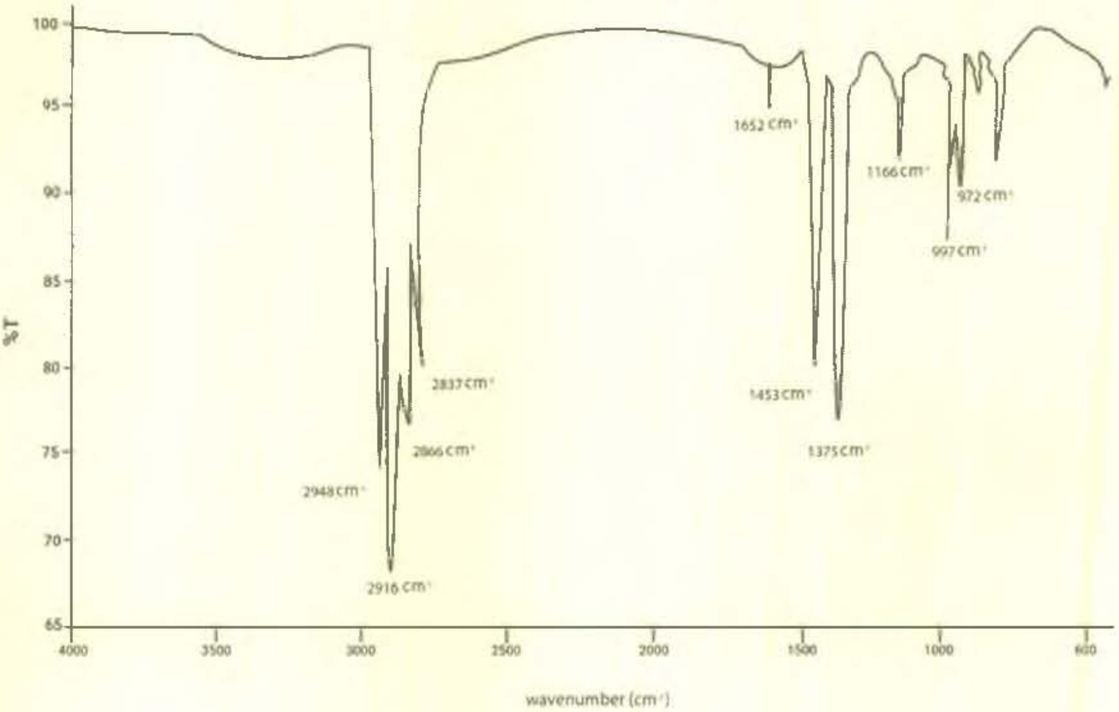

Figure 4.8: FTIR spectra of PP.

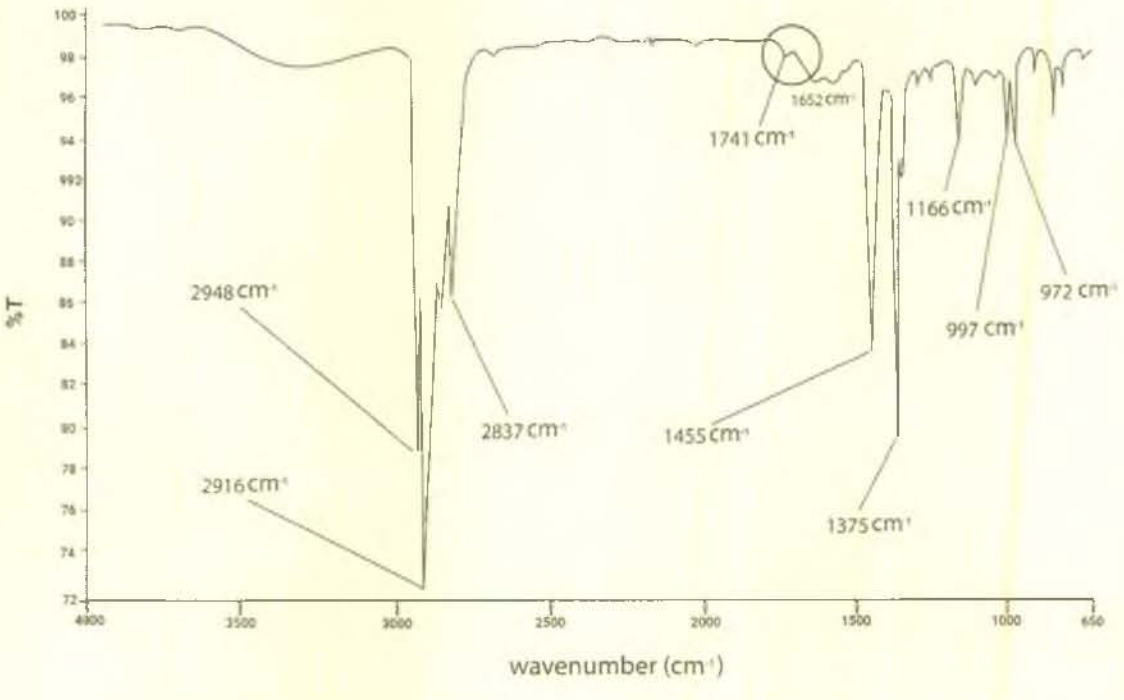

Figure 4.9: FTIR spectra of F2 composite.

4.4.5 TGA and DSC experiment

The thermal characteristics of pure polypropylene and F2 formulated composite were evaluated by TGA and DSC and presented in Figure 4.10 and Figure 4.11 respectively. As shown in Figure 4.10, the degradation process of polypropylene started at about 380°C and at 500°C temperature, 98.73% mass change was completed. However, in case of composite material, the decomposition of the composite started earlier (280°C) compared to the polypropylene matrix and 10.38% degradation was completed at 420°C temperature which might be due to the interaction between fiber and matrix material of the composites (Figure 4.11). DSC scans revealed the profile of melting characteristics of matrix and composite materials. Figure 4.10 and 4.11 showed that the DSC scans of the polypropylene matrix and optimized composite respectively and revealed that endothermic reactions were occurred for matrix as well as composite materials and 477.2°C and 477.6°C heat consumed respectively.

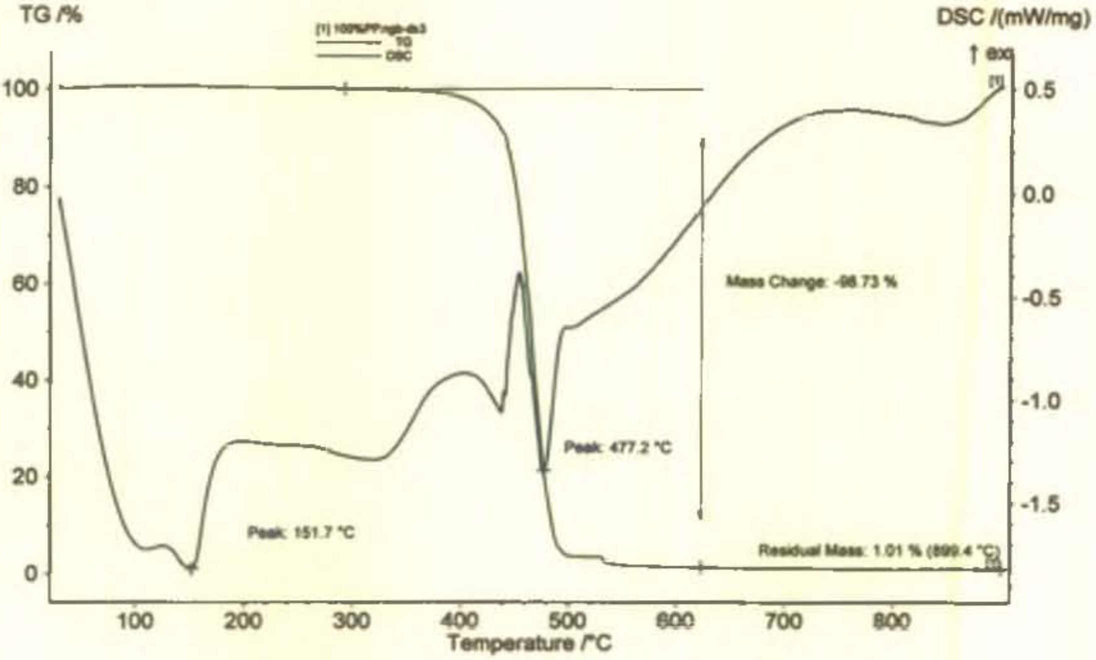

Figure 4.10: TGA and DSC of polypropylene.

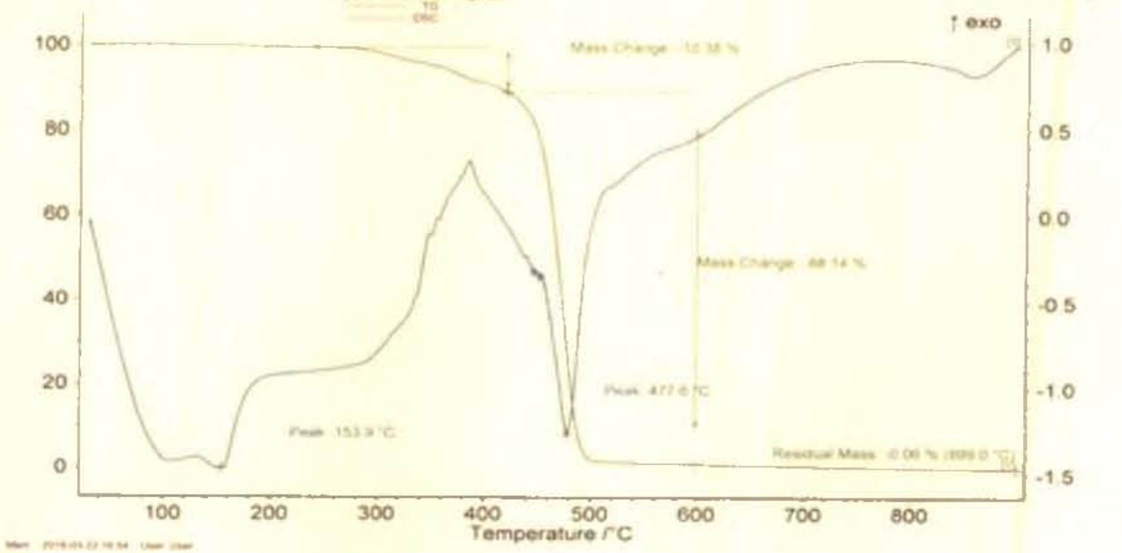

Figure 4.11: TGA and DSC of F2 composite.

4.4.6 Scanning Electron Microscopic test

Morphology properties of F2 and F5 formulated composites were analyzed by SEM (Figure 4.12 and Figure 4.13). SEM examinations showed that adhesion between fiber and matrix of the both composites was not similar. Adhesion between fiber and matrix increased for 5% to 10% fiber content but decreased for above 10% of fiber content in the composites. Consequently, some gaps were observed for 25% fiber formulated (F5) composite. Whereas no gaps was observed between fiber and matrix for 10% fiber formulated (F2) composite, indicating that a better fiber-matrix adhesion was occurred, and which ultimately lead to higher mechanical properties.

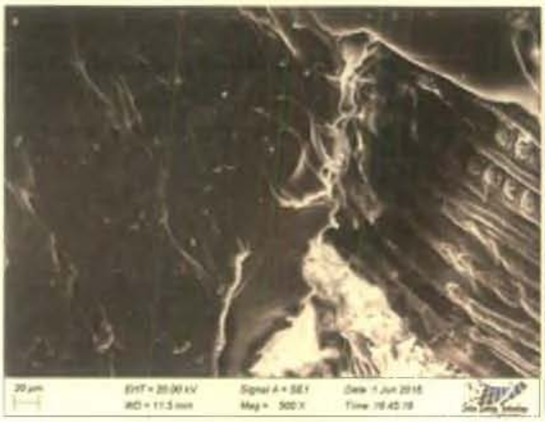

Figure 4.12: F2 composite.

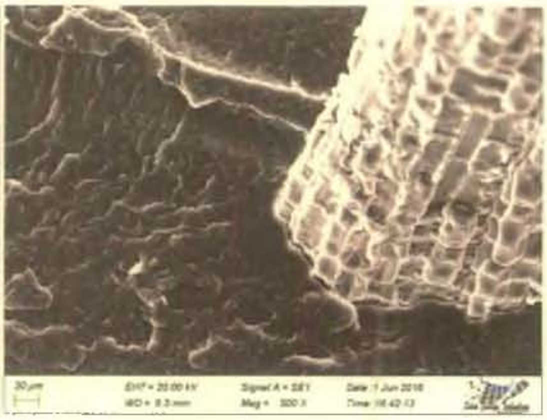

Figure 4.13: F5 composite.

4.5 Conclusion

ALS short fibers reinforced PP composites were produced using compression molding technique. The effect of fiber content in the composites was evaluated via water uptake, mechanical, morphological, thermal and soil degradation test. The 10% fiber content was found to be optimum which leads to better mechanical properties. SEM images indicated that higher fiber content, beyond 10% fiber content, introduced poor fiber-matrix adhesion, which eventually was responsible for declining trend of mechanical properties above optimum fiber content. Soil degradation test also revealed better resistance to soil degradation of F2 sample over degradation time. Based upon the above results, these new composites could be used in many applications including, packaging, furniture, housing and aviation, shipping and automotive sectors.

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CHAPTER 5

Preparation and Characterization of Naturally Woven Coconut Leaf Sheath (CLS) Fiber Reinforced Polypropylene Matrix Based Composites

5.1 Abstract

In polymer matrix composites, renewable materials have gained outstanding attractiveness as reinforcement. By compression molding naturally woven coconut leaf sheath (CLS) fiber-reinforced polypropylene (PP) composites were produced. Different weight percentages of CLS fiber (5, 10, 15, 20 and 25 wt%) were used for preparing different composites . Hydrophilic nature of the CLS fiber was assessed utilizing water uptake test and revealed strong hydrophilicity by gradually absorbing water until it reaches to a constant water uptake value of 150%. Structural characteristic of the composites were established by infrared spectrophotometer. Mechanical performance was analyzed applying tensile and bending tests. Fiber reinforcement importantly intensified the mechanical properties of the composites, and the optimum improvement of strength and stiffness was achieved for the 10% fiber content. Moreover, thermal properties, and degradation character of the composites were analyzed. Addition of fiber decreased the thermal stability of the composites. Degradation test of the composites were executed up to six months in soil medium at ambient condition and appeared slow degradation of mechanical properties over time.

5.2 Introduction

World is now going towards natural things. Environmental polices of developed countries are intensifying the pressure on producers to observe the environmental effect of their products. Therefore, the interest in utilizing natural fibers as a reinforcement of polymer–based composites is increasing mainly owing to its renewable origin (Mohanty and *et al.*, 2000). Polymer matrix composites are finding widespread applications in chemical industry and many other areas as structural materials (Mittal and *et al.*, 2016). In current years, a significant attentiveness has been generated in the use of agro-fiber as substitute of synthetic fiber (e.g., carbon and glass) in the evolution of polymer matrix composites owing to their more favorable properties such as low density, non abrasive, stiffness, high specific properties, non brittleness, and environment as well as economic advantages such as biodegradable, abundant all over the world, generate rural jobs, non-food/agricultural based economy, low cost, low energy consumption and utilization (Riccieri and *et al.*, 1999; Woodhams and *et al.*, 1984). Throughout the past two decades, natural fibers have been applied in many applications and industries including in consumer goods, low-cost housing, automotive, aircraft, paper industries, textile as well as building and construction industries replacing (Vaidya and *et al.*, 2008). Extensive studies has been carried out on lignocellulosic fibers such as sisal (Kuruvilla and *et al.*, 1996; Oksman and *et al.*, 2001) jute (De Albuquerque and *et al.*, 2000), pineapple (Abdullah-Al-Kafi and *et al.*, 2006; Wisittanawata, S. Thanawanb and T. Amornsakchai, 2014; George and *et al.*, 1998), banana (Srinivasana and *et al.*, 2014; Benítez and *et al.*, 2013; Pothan and *et al.*, 1997), bamboo (Kabir and *et al.*, 2014), vakka (Murali Mohan and *et al.*, 2007), date (Mohanty, S. N. Das, H. C. Das and S. K. Swain, 2013) and oil palm empty fruit bunch fibers (Rozman and *et al.*, 1999; Abdul and *et al.*, 2007), and the results have shown that lignocellulosic fibers can be used as an effective reinforcing agents because of its several advantages compared to their synthetic counterparts.

Mentionable here that coconut palm (*Cocos nucifera*, Linn.), a valuable tropical assets for food and energy, is plenteous in the coastal districts of Bangladesh. The coir fibers, obtained from coconut palm, are being used to make a diversity of materials containing carpets and mats. However, there are plenty of other parts of the palm other than coir from which fibers can be taken out but are not recently utilized as assets for materials in a systematized method. In fact, all of these fibers are currently used as fuel in the rural area, a utilization which does not take superiority of their high length to diameter ratio. Some of these fibers occur in nature so far in woven form e.g. leaf sheath (Figure 5.1 (a) and b), which can be readily infused with plastics or cement. In order to find better utilization of coconut leaf sheath (CLS), especially as fillers or reinforcement in comparatively low cost applications (where high strength or high modulus is not required) it is required to assess the properties of fiber-reinforced composites.

As such, sandwiching this fiber with thermoplastics could offer a strategy for producing advanced composites based on end-use prerequisite within a framework of low cost, recyclability, availability, bio-degradability and environmental consideration. Moreover, thermoplastic matrix materials are the most principal part of a composite. Polypropylene (PP) is an amorphous thermoplastic polymer and are extensively using as engineering thermoplastic because it possesses various essential and effective properties such as transparency, dimensional stability, flame resistance, high heat distortion temperature, and high impact strength. In addition, PP is extremely satisfactory for filling, reinforcing, and blending. PP with fibrous natural polymers of biomass origin is one of the most favorable routes to generate natural-synthetic polymer composites [Dey and *et al.*, 2011; Garcia and *et al.*, 2005; Karmaker and G. Hinrichsen, 1999; Khan and *et al.*, 2001; Bledzk and *et al.*, 1999; Wambua and *et al.*, 2003; Czvikovszky, 1995). Taken as a whole, in this paper, we present the fabrication of naturally woven coconut leaf sheath short fiber-reinforced polypropylene composites. The structural, mechanical, thermal and degradation characters of the composites are studied to observe the potentiality towards diversified application within the environmental legal framework.

5.3 Experiment

5.3.1 Materials

PP was procured from Polyolefin Company Private Ltd., Singapore and was grinded to small particles (50-60 μm) for appropriate and homogeneous matrix-fiber adhesion. Coconut leaf sheath fibers were collected from the rural area of Bangladesh and chopped into small pieces (2-3 mm) using hand scissors (Figure 5.1(c)) and all dirt's were taken out from the fibers with the help of mesh. Before the composites fabrication the fibers were vacuum-dried at 100°C for 5 hours.

5.3.2 Composite preparation

The PP matrix based unidirectional composites were prepared by compression molding. At first, with the help of cleaning agent, the mould was cleansed. Then the dried CLS short 10 wt% fibers with PP were used to prepare composites according to the ratio of fiber : PP = 5:95, 10:90, 15:85, 20:80 and 25:75. All formulations were made using moulds in the heat press under 5 bar consolidation pressure and190°C temperature for 5 min. The model of heat press was 3856, Carver, INC, USA. After 5 minutes, in a separate press the moulds were cooled down to room temperature under 5 bar pressure. Figure 5.1 (d) shows the final composite product.

Table 5.1: The composition and mechanical properties of the different composites.

Formulation No.	**Weight percentage of PP and CLS in different composites**	**Mechanical properties***				
		Tensile strength (MPa)	**Tensile modulus (MPa)**	**Bending strength (MPa)**	**Bending modulus (MPa)**	**Elongation at break (%)**
F1	95% PP + 5% CLS	10.81 ± 1.2	103 ± 11	35.20 ± 3.5	220 ± 15	24.11 ± 2.3
F2	90% PP + 10% CLS	26.86 ± 2.3	278 ± 13	45.55 ± 2.6	525 ± 12	19.97 ± 3.5
F3	85% PP + 15% CLS	16.33 ± 2.5	204 ± 8.2	34.24 ± 2.7	386 ± 18	18.99 ± 2.6
F4	80% PP + 20% CLS	9.47 ± 1.5	109 ± 7.8	32.21 ± 1.1	209 ± 11	18.15 ± 1.8
F5	75% PP + 25% CLS	8.10 ± 1.0	96 ± 8.3	28.63 ± 1.3	182 ± 13	18.06 ± 2.1

*Data represent mean ± SD

Figure 5.1: (a) Coconut palm, arrows show woven leaf sheath, (b) Naturally woven coconut leaf sheath, (c) chopped coconut leaf sheath; and (d) chopped coconut leaf sheath-reinforced PP composite.

5.3.3 Water uptake of the CLS fiber

The water uptake of the CLS fibers was periodically monitored up to 10 days by soaking the pre-weighed dried samples in the distilled water at room temperature. The samples were removed time to time, lightly wiped to taken out surface water and weighed. The equation of calculation of water absorption is -

$$\text{Water absorption} = \frac{W_f - W_i}{W_i} \,.\, 100 \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

Where, W_i is the initial weight of the samples, and W_f is the final weight of the samples after fixed time.

5.3.4 FTIR analysis

The structural analysis of the PP and composites was conducted applying a Perkin Elmer Fourier transform infrared (FTIR) spectrometer with an attachment of attenuated total reflectance (ATR) accessory. The spectra were appeared over the range of 4000-600 cm^{-1} at a resolution of 4 cm^{-1}.

5.3.5 Mechanical properties of the composites

Using a universal testing machine (model H50 KS-0404, Hounsfield Series S, UK) with a load capacity of 5000 N at a crosshead speed of 10 mm/min and gauge length of 20 mm, the tensile properties of the composites (F1-F5) were measured according to DIN 53455. The three point bending tests were executed at a crosshead speed 10 mm/ min, and span distance 40 mm, according to DIN 53452. The average results of minimum five samples for each composite were reported.

5.3.6 Surface morphology

For the morphological analysis of the composites, SEM is an effective technique. Surface morphology plays an important role in case of composite type materials. The compatibility and distribution between the fibers and the matrix materials could be found using SEM analysis. The strong interaction between fibers and matrix exhibits superior tensile properties. Hitachi S-4000 model field emission scanning electron microscope was used to investigate the fracture surfaces of the tensile specimens of F2 and F5 composites operating at 5 kV.

5.3.7 Thermal analysis

The thermogravimetric analysis (TGA) and DSC of the PP and optimized composite were performed to find out the physical changes occurring in the material of the composites due to temperature effects (Monteiro and *et al.*, 2012). The increasing rate of temperature was 20°C/min under inert nitrogen atmosphere and maximum temperature was 900°C.

5.3.8 Degradation test of the optimized composite

The degradation test of the optimized composite was executed for six months burying the samples in humid soil whose moisture level was at least 25%. After set time point, the samples were taken out, cleansed with fresh water and dried at 80°C temperature for 8 h. The variation of tensile strength and tensile modulus (stiffness) was intermittently observed in order to compute the degradable nature of the composites in this humid condition.

5.4 Results and Discussion

5.4.1 Water uptake of the CLS fiber

The hydrophilic nature of the fiber influences the water retention capacity. Figure 5.2 shows the water uptake profile of the CLS fiber submerged in distilled water at room temperature for different time periods. Initially, the fiber gradually absorbed water and reached a plateau value of 150% after 8 days. It is well known that hydroxyl groups (-OH) present on the surface of the fibers are responsible for water absorption (Rahman and *et al.*, 2007). As shown in the Table 5.2, CLS contains about 70% cellulose (Poddar and *et al.*, 2016). The cellulose structure of CLS possesses a lot of –OH groups. These hydroxyl groups are responsible for the strong hydrophilic character of CLS. At this state, the cellulosic –OH groups are very reactive and they form strong hydrogen bonds being able to preserve water (Mishra and*et al.*, 2003).

Table 5.2: Chemical compositions of CLS fiber.

Components	**%**
Aqueous extract	0.53
Fatty and waxy matters	2.86
Pectic matters	0.80
Lignin	27.97
α - cellulose	59.39
Hemicellulose	8.45
Total	100.00

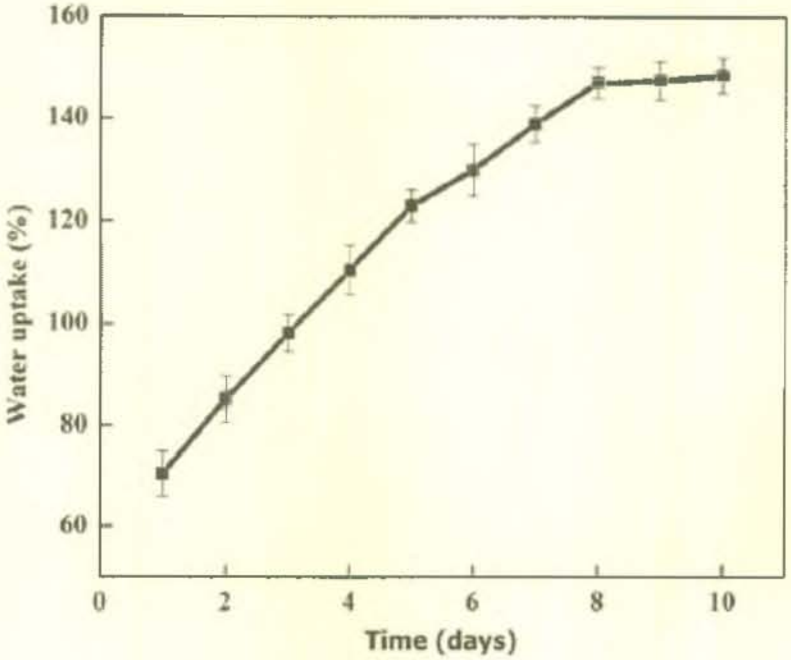

Figure 5.2: Water uptake profile of naturally woven coconut leaf sheath (CLS) soaked in distilled water for different time period.

5.4.2 Structural characterization

Naturally woven CLS short fiber-reinforced PP composites were fabricated varying the fiber content from 5 to 25 wt%, and the five formulations, namely, F1, F2, F3, F4 and F5 were presented in the Table 5.1. The FTIR spectra of PP and F2 composite were illustrated in Figure 5.3 and 5.4, respectively. As shown in Figure 5.3, the fingerprint region of pure PP was observed from 1455-600 cm^{-1}, particularly 1455 and 997 cm^{-1}attributed to C=C stretching and C-H bending vibrations, respectively. The peak observed at 2916 cm^{-1} assigned to C-H stretching vibration. The spectrum of composite showed a peak at about 1740 cm^{-1} associated to carbonyl (C=O) stretching of acetyl groups of hemicellulose in the natural CLS fiber (Cantero and *et al*., 2003), along with characteristic peaks of PP (Figure 5.4).

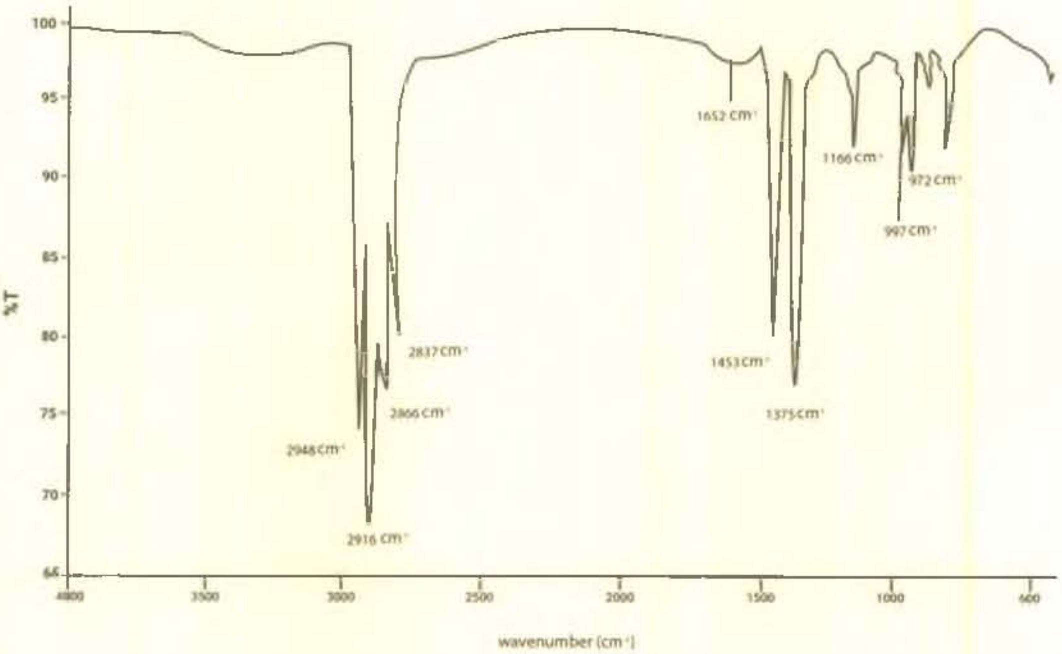

Figure 5.3: FTIR spectra of polypropylene.

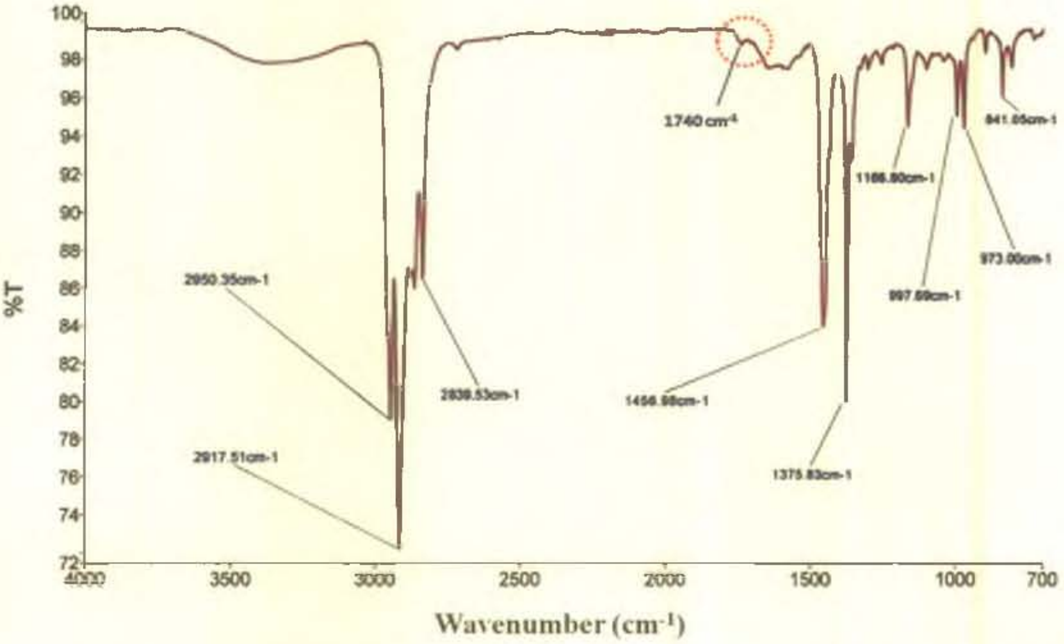

Figure 5.4: FTIR spectra of optimized composite F2 (90% PP +10% CLS).

5.4.3 Mechanical properties of the composites

Mechanical property of the composites is an important factor for the application of interests. As such, tensile and bending tests of the composites were done to measure the strength and stiffness of the composites. Figure 5.5 showed typical stress-strain curves for the composites. All composites exhibited non-linear stress-strain curves demonstrating ductile behavior. The tensile strength (TS), bending strength (BS) and elongation at break (Eb%) of the composites were presented in Figure 5.6 (a) and tensile modulus (TM) and bending modulus (BM) of the composites were presented in Figure 5.6(b) and corresponding data were shown in Table 5.1. It was found that fiber reinforcement occurred improving the strength and stiffness of the composites. The increase in fiber content from 5 to 10 wt% resulted in both increased tensile and bending strength. The highest TS and BS values were observed for F2 (90% PP + 10% CLS) and was considered as an optimized composite. Interestingly, higher amount of fiber content (>10 wt%) reduced TS and BS of the F3, F4 and F5 composites. From F1 to F2 formulation, the TS and BS values were enhanced, particularly significant increase of TS, while as, they were gradually declined from F3 to F5 formulations. As observed from Table 5.1 and Figure 5.6 (a), the highest TS and BS values of 26.86 MPa and 45.55 MPa, respectively, was found for F2 formulation. Elongation at break (Eb%) is one of the significant mechanical properties of any engineering material. As a whole, the composite showed lower elongation at break for all formulations. This phenomenon might be owing to the low elongation at break (%) of the CLS fibers compared to the matrix PP. No appreciable dissimilarities were found with the variation of fiber content.

The significant enhancement in the tensile and bending modulus of the composites was obtained with increasing fiber content from 5 to 10 wt%, as shown in Figure 5.6 (b). Beyond 10 wt% fiber content, the composites exhibited a decreased modulus with loading up to 25 wt% fiber content. Likewise strength, F2 formulation showed the maximum TM and BM values of 278 MPa and 525 MPa, respectively. A 10 wt% fiber loading increased correspondingly 170 and 140% of TM and BM of the composite.

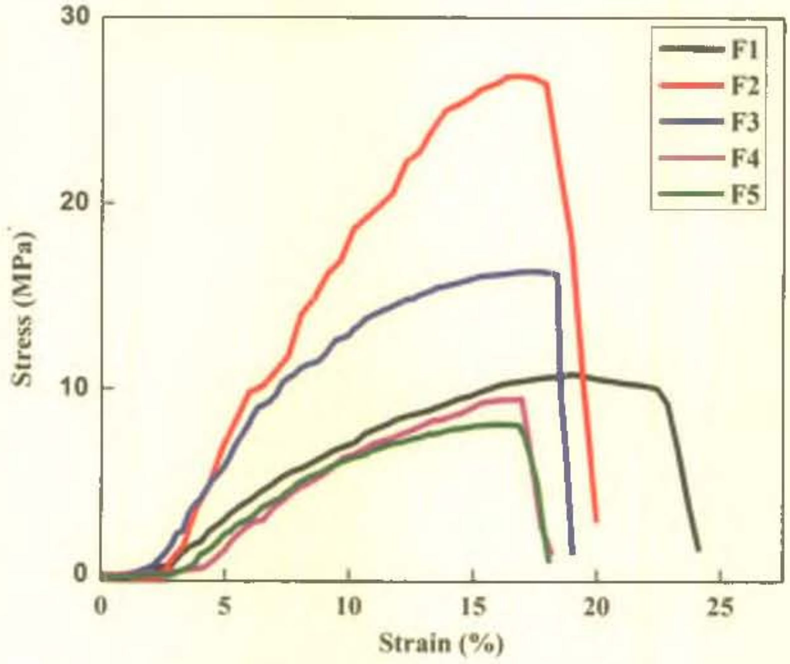

Figure 5.5: Representative stress-strain curves of the composites.

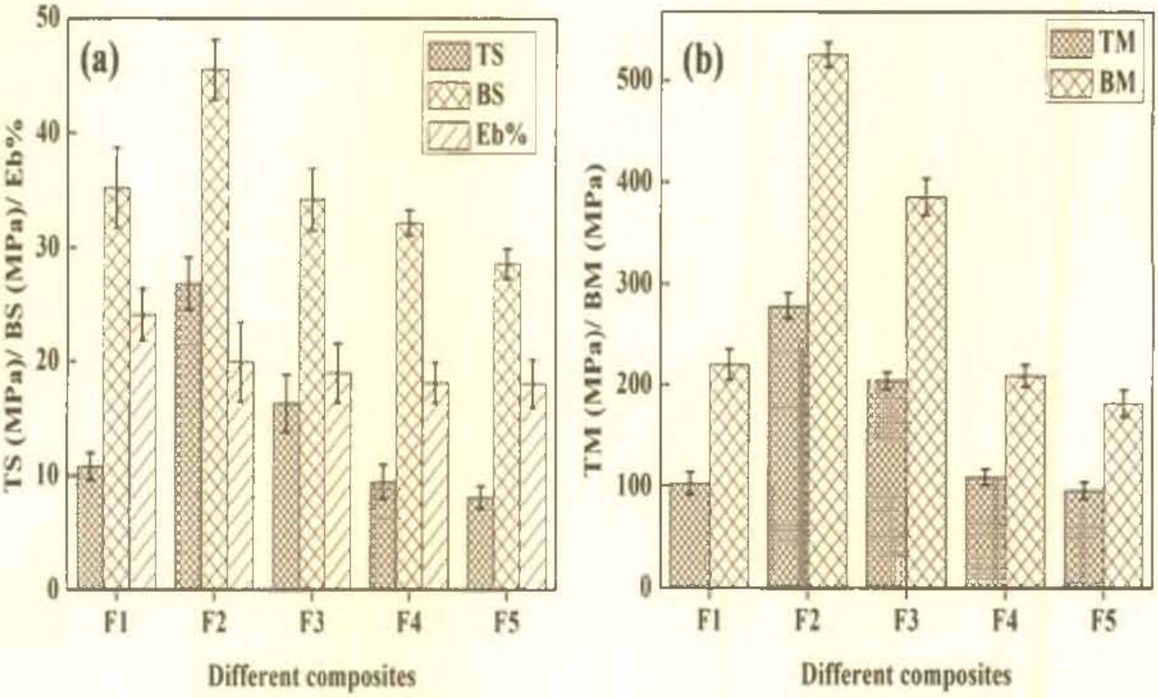

Figure 5.6: Mechanical properties (a) TS, BS and Eb and (b) TM and BM of the composites.

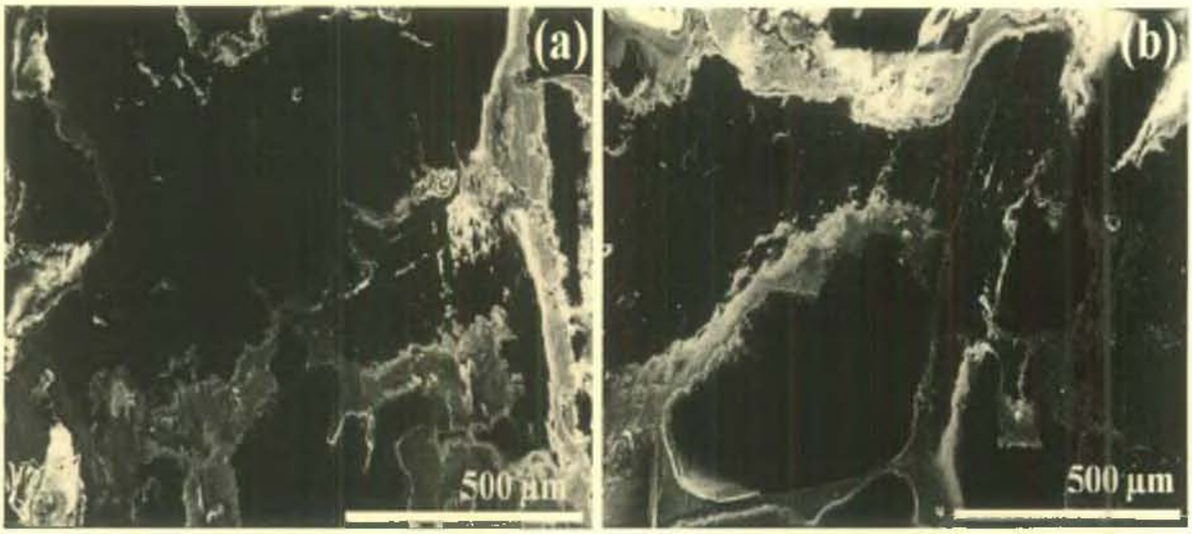

Figure 5.7: SEM images of (a) optimized composite F2 (90% PP +10% CLS) and (b) F5 composite (75% PP + 25% CLS).

It is expected that with the 10 wt% of fiber reinforcement, the fibers are capable to provide to the effectual stress transfer between fibers and the PP matrix and concomitantly increases the tensile strength. The improvement in the strength is attributable to the strong interactivity between fibers and the PP matrix, as evidenced by a scanning electron micrograph being taken on fracture surface of optimized composite containing 10 wt % fibers (Figure 5.7(a)). It is worthwhile to mention that the properties of hybrid composites depends not only the properties of individual reinforcing fibers but also the orientation and arrangements pattern of the fibers, fiber-matrix adhesion etc. The decreased strength and stiffness at higher CLS fiber content (15 to 25 wt%) could be ascribed to tinny adhesion between the fiber and the matrix, which encourages micro crack origination at the interface. A higher fiber content results in more (and probably larger) void origination through processing, which leads to micro crack origination under loading and therefore decreases the strength (Thwe and *et al.*, 2002). SEM images of fracture composite with 25 wt% fiber content showed larger fiber breakages and gaps as well as delamination suggesting the reduced matrix to fiber interactions and dispersion problems (Figure 5.7 (b)). On the other hand, at low fiber content (5 wt %), short fibers added may act as defects which can promote crack formation, hence reducing the strength (Soykeabkaew and *et al.*, 2004).

From the overall results of Figure 5.6 (a) and (b), it is concluded that 10 wt % is the optimum amount of fibers where TS, TM, BS and BM are recorded highest amongst the respective fiber composites.

5.4.4 Thermal properties of the composites

Thermal characteristics of the PP matrix (granules) and the optimized composite (10 wt % fiber) were evaluated by TGA and DSC. The thermograms of both PP granules and optimized composite were presented in Figure 5.8 and 5.9, respectively. As shown in Figure 5.8, the degradation process of PP started at about 380°C and at 500°C temperature 98.73% mass change was finished. However, the decomposition of the composite started earlier compared to the PP matrix which might be ascribed to the interaction between fiber and matrix material (Figure 5.9). The addition of fibers decreased the thermal strength of the composites. As seen from the thermogravimetric curve of composite, the mass change started at 320°C.

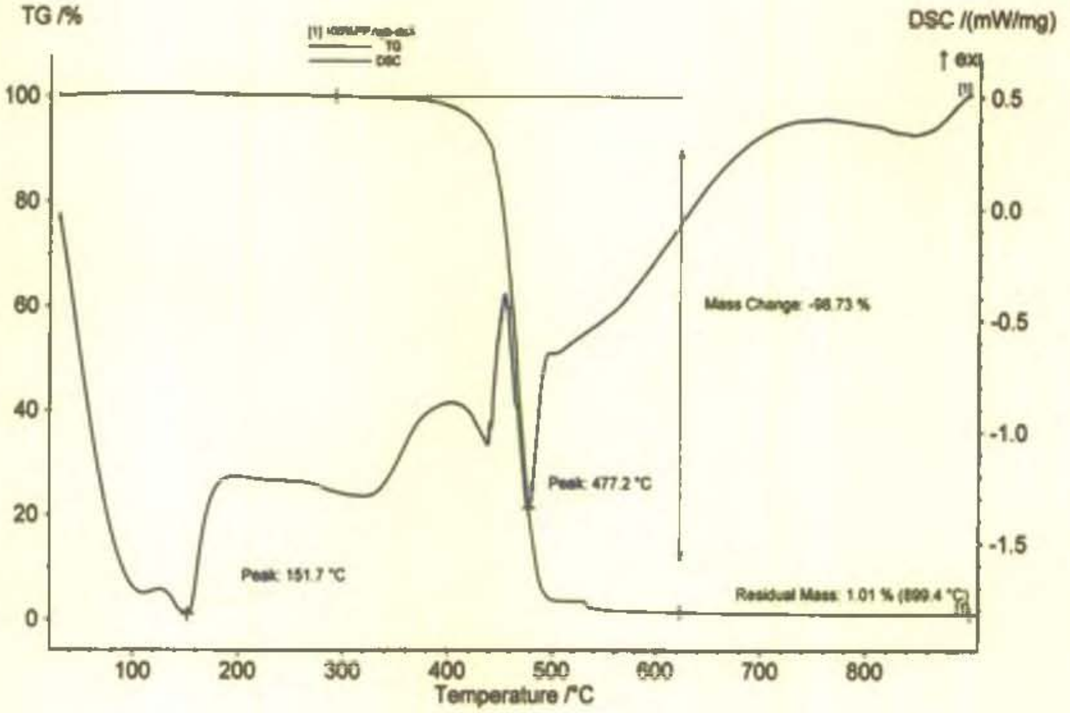

Figure 5.8: TGA and DSC of polypropylene.

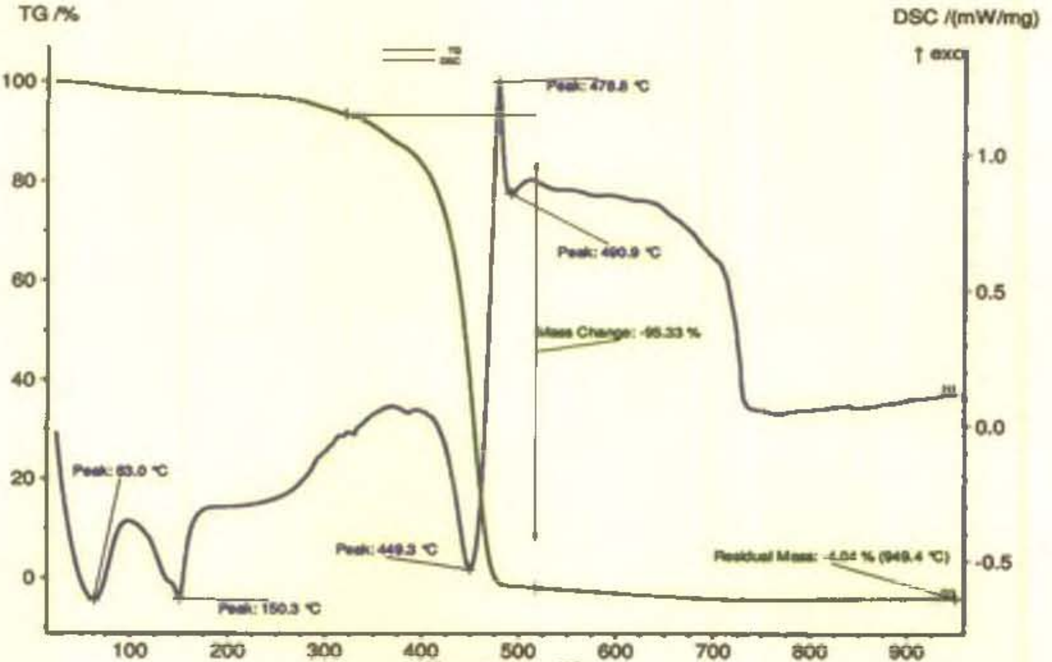

Figure 5.9: TGA and DSC of optimized composite F2 (90% PP +10% CLS).

Figure 5.8 and 5.9 revealed that the melting profile of composite did not change significantly. Only a little displacement was observed in respect to neat PP. The DSC scans of the PP matrix and composite showed peaks of melting at 151.7 and 150.3°C, respectively. The temperature peak of melting slightly decreased because of the influence of added fibers, showing an interaction between fiber and matrix.

5.4.5 Soil degradation of the optimized composite

To understand the detrimental effects of soil environment on the mechanical property of the composite, it is necessary to carry out the soil degradation test. The degradation test of the optimized composite was executed in the soil medium (high moisture content) for up to six months. Tensile strength (TS) and tensile modulus (TM) values were plotted against degradation time and were appeared in Figure 5.10 (a) and (b), respectively. It was observed that gradually TS and TM values were reduced over time, as expected. A slightly decrease of TM was observed compared to TS of the composite.

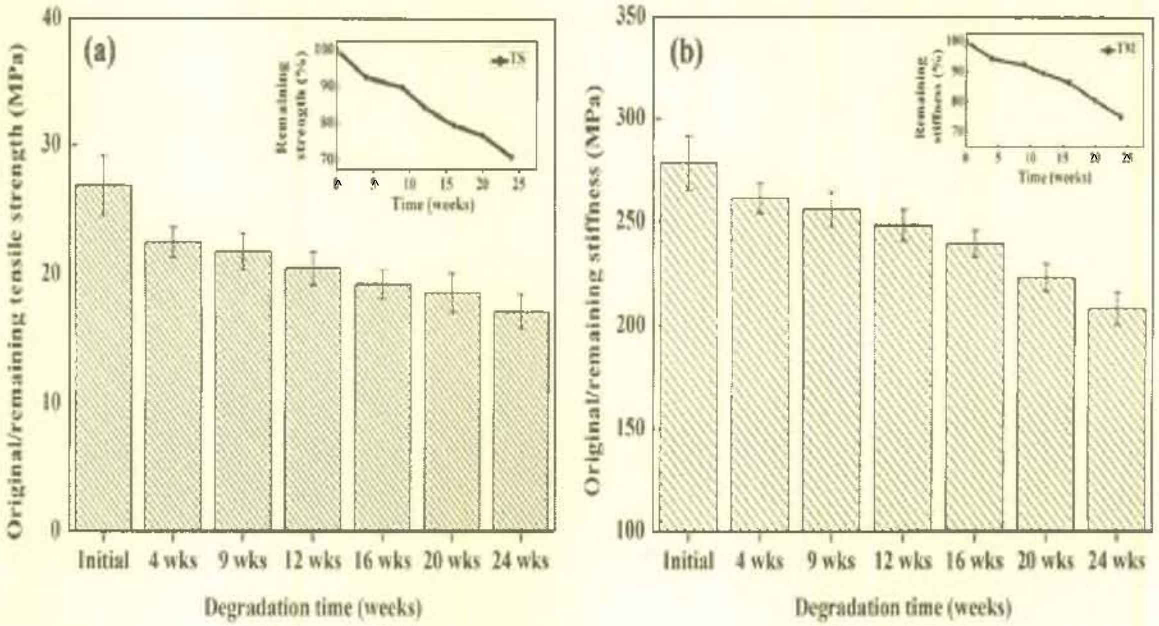

Figure 5.10: Degradation of (a) TS and (b) TM (stiffness) of the optimized composite F2 (90% PP +10% CLS) in the soil medium. (Inset images show decreasing pattern of tensile strength and tensile modulus as a function of time).

After six months of degradation in the soil, composite retained respectively, 70 and 75% of its initial tensile strength and modulus (as shown in the inset images of Figure 5.10 (a) and (b)). During immersion of the composite in the humid soil medium, water penetrated from the fiber edges of the composites and thus deteriorated the fibers steadily inside the composite as well as weakening of the bonding strengths between the fibers and matrix. In addition, bacterial attack could also responsible for the loss of the mechanical properties because biodegradable fibers are normally attacked by microbial action (Mohanty and *et al.*, 2000; Marasinghe and *et al.*, 1997).

5.5 Conclusion

The intention of this research was to evaluate the benefits of reinforcing polypropylene with naturally woven coconut leaf sheath short fibers. CLS short fiber-reinforced polypropylene composites were formulated by compression molding varying the fiber loading. Tensile and bending strength were intensified with enhancing the fiber content up to 10 wt% and this was considered as optimum fiber loading. Reinforcement decreased the thermal stability of the composites. The optimized composite retained about 70% of its initial strength and stiffness after six months of soil degradation. Hence, the composites might have a potential utilization where relatively lower strength is required.

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CHAPTER 6

Preparation and Characterization of Coconut Stem Fiber Reinforced Polypropylene Matrix Based Composites

6.1 Abstract

The intention of this research work is to investigate mechanical (tensile strength, bending strength, tensile modulus, bending modulus, elongation-at-break, thermal (TGA and DSC), morphological, water uptake and soil degradation at dissimilar compositions (5, 10, 15, 20 and 25 wt %) of coconut stem fiber reinforced polypropylene composites. 10% fiber based composite showed highest mechanical properties and others properties supported the results.

6.2 Introduction

The demand of eco friendly and biodegradable fibers reinforced thermosets [1-4] and thermoplastic [5] composites have increased owing to the development of people's living standards. Recently, a considerable demand has been originated in the utilization of agro-fiber as substitute of synthetic fiber (e.g., carbon and glass) in the evolution of polymer matrix composites owing to their more favorable properties for instance non abrasive, low density, stiffness, non brittleness, high specific properties, and environment as well as economic advantages such as abundant all over the world, biodegradable, originate rural jobs, non-food/agricultural based economy, low energy consumption, low cost and utilization [6-8].

In our country, *Cocos nucifera* trees (coconut tree) are highly obtainable in the coastal area which produces vast coconut stem fiber. This unusable item is used in this research work to produce biodegradable and environment friendly composite material. Different compositions (5, 10, 15, 20 and 25 wt %) of fibers are used to prepare reinforced polypropylene composites. Among these, 10% fiber based composite showed highest mechanical properties and FTIR, TGA-DSC, morphological, water uptake, soil degradation test of different composites were also performed.

6.3 Experimental

6.3.1 Materials

PP was procured from Polyolefin Company, Private Ltd., Singapore. To get actual and homogeneous adhesion between fibers and matrix, PP was grinded to get 50-60 μm particles. Coconut stem fibers (Figure 6.1) were collected from coconut stem of *Cocos nucifera* trees (coconut tree). Then the stem was soaked into water for two weeks to obtain coconut stem fibers. The water immersion made the coconut stem soft and weakened the bond between the fiber and the resin and waxy materials, which eventually helped to peeling off the loosen fibers from the resinous materials. The peeled off fibers were cleansed using huge amount of fresh water and dried appropriately. Using hand scissors, the fibers were cut into 2-3 mm size (Figure 6.2) and all dirt's were taken out from the fibers with the help of mesh. Before the composites fabrication the fibers were vacuum-dried at 100°C for 5 hours.

Figure 6.1: Coconut stem fiber.

Figure 6.2: Coconut stem fiber chips.

6.3.2 Chemical composition of Coconut Stem Fiber (CSF)

Coconut stem fiber (CSF) contains α – cellulose, fatty and waxy matters, lignin, hemicellulose, pectic matters and aqueous extract and the amount of these components are shown in the Table 6.1. 22.54% lignin, 46.26% α – cellulose and 18.02% hemicelluloses is present in the fiber. More importantly, small particles of cellulose of coconut stem fiber are cemented and surrounded together by hemicellulose and lignin.

Table 6.1: Chemical compositions of coconut stem fiber.

Sl. No.	Name	%
1	Aqueous Extract	5.52
2	Lignin	22.54
3	Pectic matters	1.75
4	Fatty and waxy matters	5.91
5	α - cellulose	46.26
6	Hemicellulose	18.02
	Total	100

6.3.3 Composite preparation

CSF reinforced PP matrix based composites were formulated by compression molding. Five formulations of coconut stem fiber (5%, 10%, 15%, 20% and 25% fiber) reinforced PP composites was prepared by varying the fiber content as shown in the Table 6.2. All formulations were made using moulds in the heat press under 5 bar consolidation pressure and 190°C temperature for 5 min. The model of heat press was 3856, Carver, INC, USA. After 5 minutes, in a separate press the moulds were cooled down to room temperature under 5 bar pressure. The prepared composite is shown in the Figure 6.3.

Figure 6.3: Coconut stem fiber and Polypropylene composite.

Table 6.2: The composition of the different composites.

Formulation no.	**CSF (wt %)**	**Polypropylene Powder (wt %)**
F1	5	95
F2	10	90
F3	15	85
F4	20	80
F5	25	75

6.3.4 Mechanical properties of the composites

The tensile tests of the composites were performed using an electro-mechanical testing machine under a maximum load capacity of 5kN with a gauge length of 20 mm and crosshead speed of 10 mm/min. But, in case of bending properties, the crosshead speed was set at 10 mm/min with a span distance of 40 mm. Tensile and bending tests were performed following DIN 53455 and DIN 53452 standards methods, respectively. Five specimens for each sample were examined in the instrument.

6.3.5 Water uptake

At room temperature (25°C), water uptake analysis was done by soaking the pre-weighed dried samples in the distilled water. The samples were removed time to time, lightly wiped to taken out surface water and weighed. The equation of calculation of water absorption is -

$$\text{Water absorption} = \frac{w_f - w_i}{w_i} \,.\, 100 \ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

Where, W_i is the initial weight of the samples, and W_f is the final weight of the samples after fixed time.

6.3.6 Degradation tests of the composites

To investigate soil degradation profile of the composites, all composite samples were buried in garden soil for 24 weeks. Cellulose undergoes deterioration when buried in moisturized soil (at least 25%). After set time point, the samples were taken out, cleansed with fresh water and dried at 80°C temperature for 8 h. The variation of TS was intermittently observed in order to compute the degradable nature of the composites in this humid condition.

6.3.7 FTIR analysis

The structural study of 100% Polypropylene and F2 composite were examined by Fourier Transform Infrared Spectroscopy with an attachment of ATR (attenuated total reflectance) accessory. Using a Perkin Elmer instrument, the spectra were recorded in the wave number ranges of 4,000-600 cm^{-1}.

6.3.8 TGA and DSC analysis

The thermogravimetric analysis (TGA) and DSC of the PP and optimized composite were performed to find out the physical changes occurring in the material of the composites due to temperature effects [9]. The increasing rate of temperature was 20°C/min under inert nitrogen atmosphere and maximum temperature was 900°C.

6.3.9 SEM investigation

The compatibility and distribution between the fibers and the matrix materials could be found using SEM analysis. For the morphological analysis of the composites, SEM is an effective technique. Surface morphology plays an important role in case of composite type materials. The strong interaction between fibers and matrix exhibits superior tensile properties. S-4000 model of Hitachi brand field emission scanning electron microscope was used to investigate the fracture surfaces of the tensile specimens of F2 and F5 composites operating at 5 kV.

6.4 Results and Discussion

6.4.1 Tensile and other mechanical properties of the composites

Tensile and bending tests of the composites were done to measure the strength and stiffness of the composites. Mechanical property of the composites is an important factor for the application of interests. All composites exhibited non-linear stress-strain curves demonstrating ductile behavior. The tensile strength (TS), bending strength (BS), tensile modulus (TM), bending modulus (BM) and elongation at break (Eb%) of the composites were presented in Figure 6.4, 6.5, 6.6 and 6.7 respectively. It was found that fiber reinforcement occurred improving the strength and stiffness of the composites. The increase in fiber content from 5 to 10 wt% resulted in both increased tensile and bending strength. The highest TS and BS values were observed for F2 (90% PP + 10% CSF) and was considered as an optimized composite. Interestingly, higher amount of fiber content (>10 wt%) reduced TS and BS of the F3, F4 and F5 composites. From F1 to F2 formulation, the TS and BS values were enhanced, while as, they were gradually declined from F3 to F5 formulations. As observed from Figure 6.4 and 6.5, the highest TS and BS values of 24.28MPa and 44.29MPa, respectively, was found for F2 formulation. Elongation at break (Eb%) is one of the significant mechanical properties of any engineering material. As a whole, the composite showed lower elongation at break for all formulations. This phenomenon might be owing to the low elongation at break (%) of the CSF fibers compared to the matrix PP. No appreciable dissimilarities were found with the variation of fiber content.

The significant enhancement in the tensile and bending modulus of the composites was obtained with increasing fiber content from 5 to 10 wt%, as shown in Figure 6.6. Beyond 10 wt% fiber content, the composites exhibited a decreased modulus with loading up to 25 wt% fiber content. Likewise strength, F2 formulation showed the maximum TM and BM values of 249MPa and 509MPa, respectively.

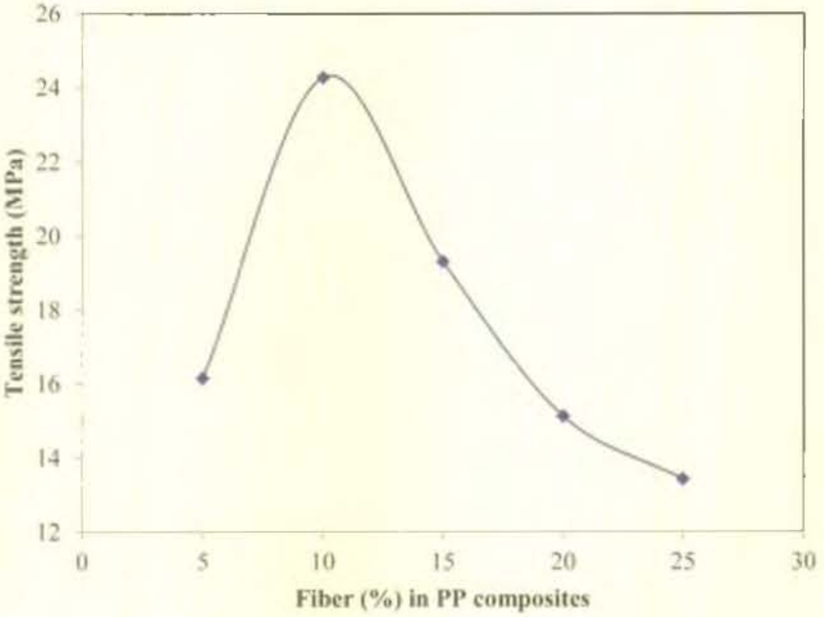

Figure 6.4: Tensile Strength of different % of CSF fiber.

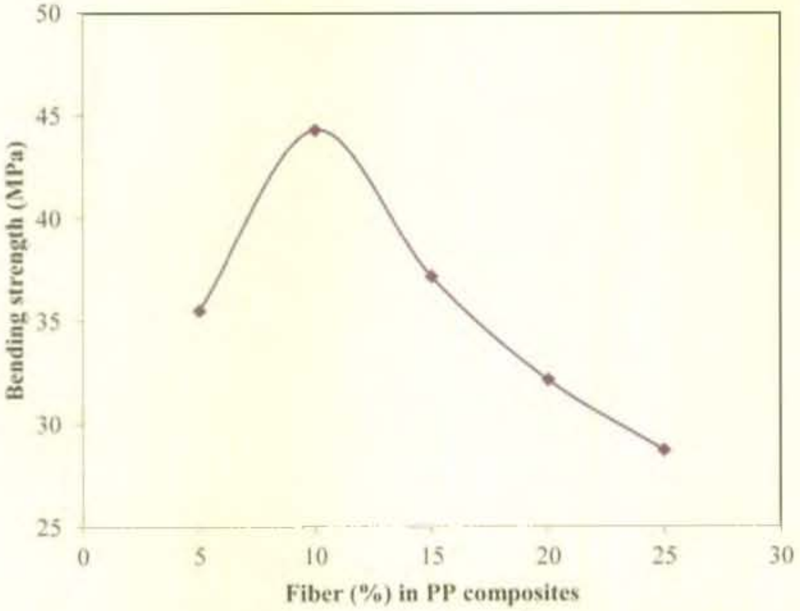

Figure 6.5: Bending Strength of different % of CSF fiber.

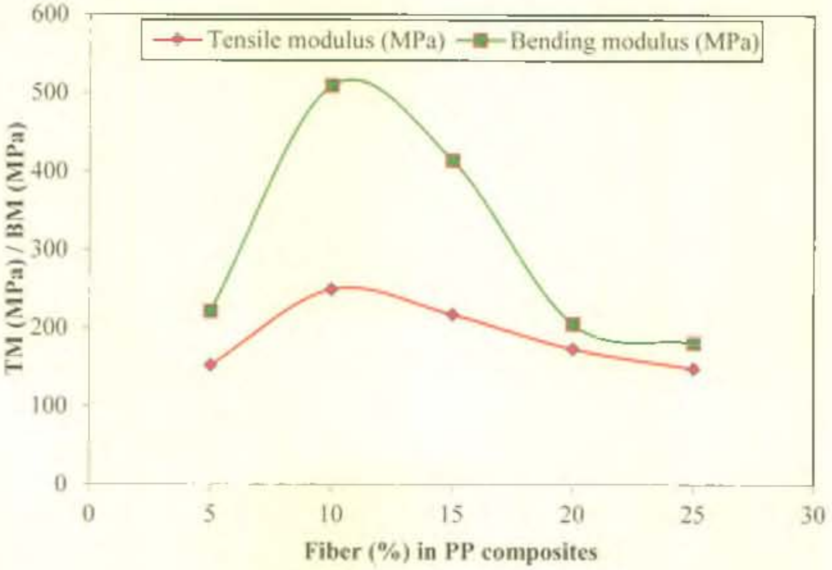

Figure 6.6: TM and BM of the composites.

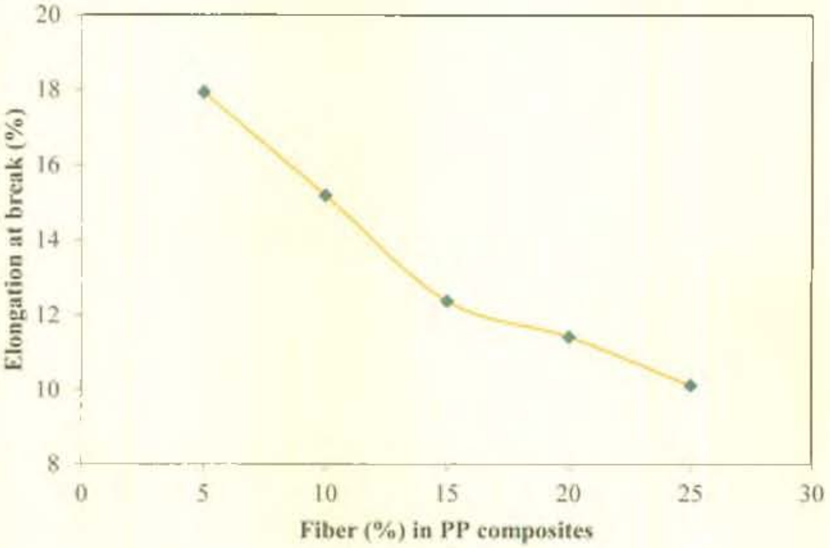

Figure 6.7: Eb of different % of CSF fiber.

6.4.2 Water absorption of the composites

The values of water uptake of the different formulated composites (F1, F2, F3, F4 and F5) were computed by submerging the samples in de-ionized water contained in a static glass beaker at room temperature. The subtraction value of initial weight from the final weight indicates the water uptake value. The hydrophilic nature of the fiber influences the water retention capacity. Figure 6.8 shows the water uptake profile of the composites. As shown in the Table 6.1, CSF contains about 65% cellulose [10]. The cellulose structure of CSF possesses a lot of –OH groups. These hydroxyl groups are responsible for the strong hydrophilic character of CSF. At this state, the cellulosic –OH groups are very reactive and they form strong hydrogen bonds being able to preserve water [11].

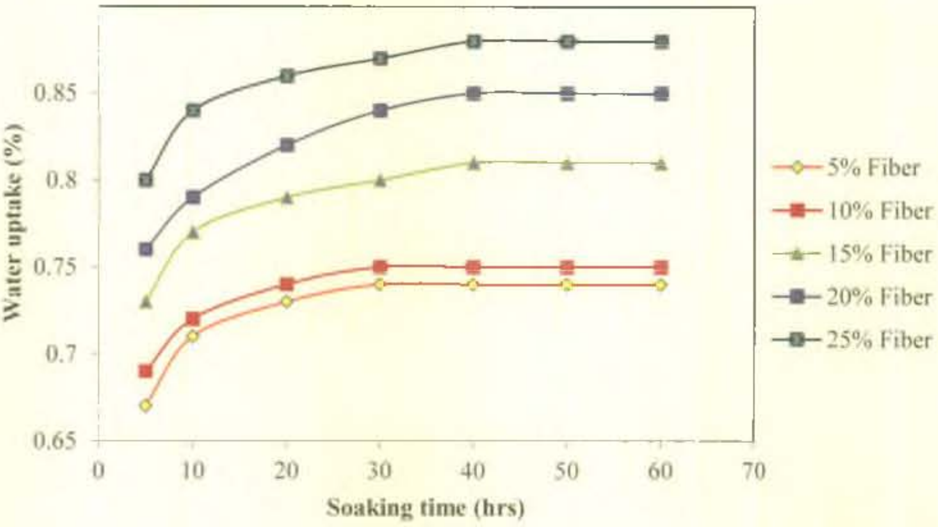

Figure 6.8: Water uptake (%) by composites in aqueous media at 25°C.

6.4.3 Degradation tests of the composites

Coconut stem fiber is a cellulose-based biodegradable fiber - absorbs water within a couple of minutes owing to its strong hydrophilic nature. Cellulose has a powerful trend to demean when buried in soil [12]. Soil degradation test was performed to investigate the degradation trend of the samples in soil media for up to 24 weeks. During soil degradation test, water penetrates through the cutting edges of the composites which lead to the degradation of cellulose in the fiber, and such degradation is responsible for the diminished mechanical properties of the composites over time. In addition, bacterial attack could also responsible for the loss of the mechanical properties because biodegradable fibers are normally attacked by microbial action [13].

After six months of degradation in the soil, F1, F2, F3, F4 and F5 samples reduced nearly 61, 33, 53, 66 and 74% of TS. Figure 6.9 showed the degradation profile of the composites. As observed from Figure 6.9, the degradation rate of TS of the optimized (F2) composite is the lowest (33%).

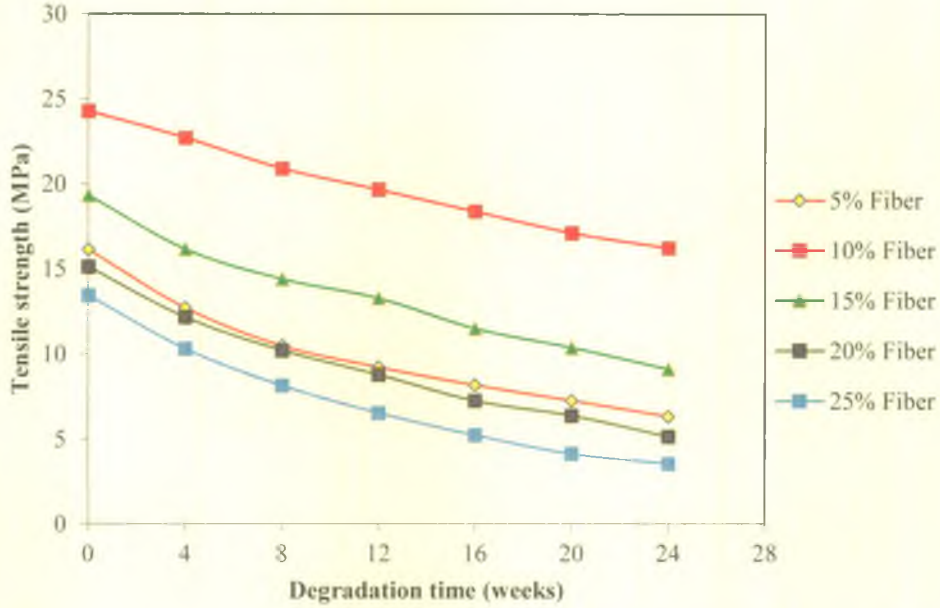

Figure 6.9: Degradation of TS of the different formulated composites in the soil medium.

6.4.4 Structural characterization

FTIR analysis was carried out to calculate the change of surface composition of the optimized composite (F2). The FTIR spectra of PP and F2 composite were illustrated in Figure 6.10 and 6.11, respectively. As shown in Figure 6.10, the fingerprint region of pure PP was observed from 1455-600 cm^{-1}, particularly 1455 and 997 cm^{-1} ascribed to C=C stretching and C-H bending vibrations, respectively. The peak observed at 2916 cm^{-1} assigned to C-H stretching vibration. The spectrum of composite appeared a peak at about 1742 cm^{-1} associated to carbonyl (C=O) stretching of acetyl groups of hemicellulose in the natural CSF fiber [14], along with characteristic peaks of polypropylene (Figure 6.11).

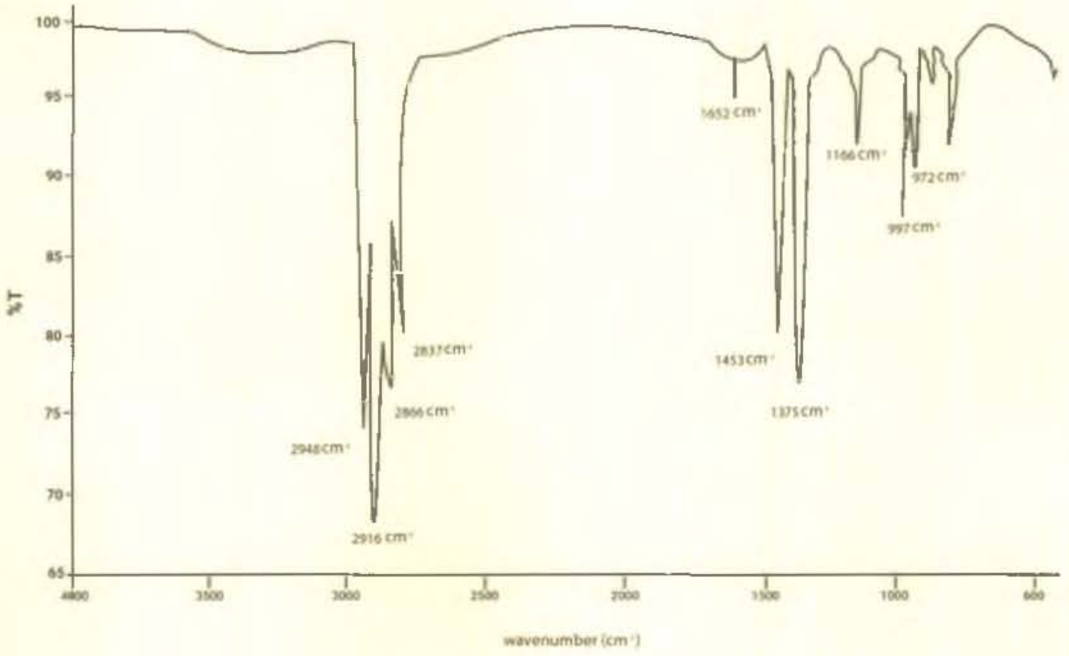

Figure 6.10: FTIR spectra of polypropylene.

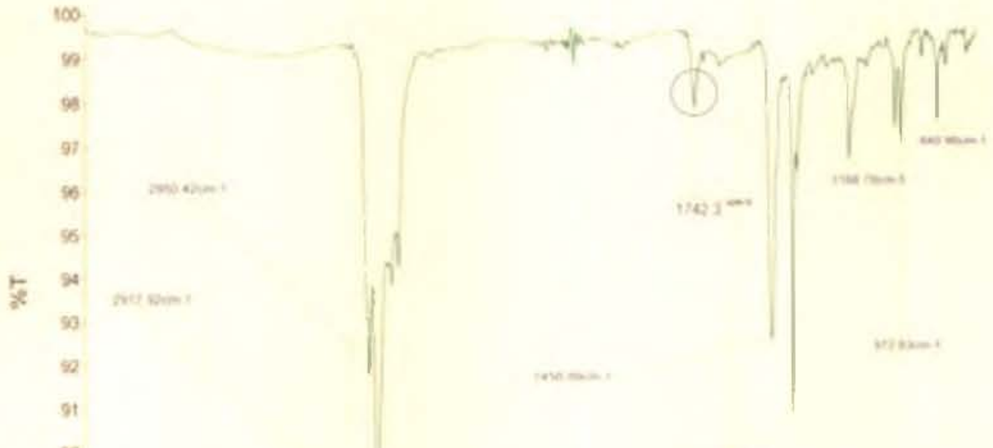

Figure 6.11: FTIR spectra of F2 (10% CSF +90% PP) composite.

6.4.5 TGA and DSC analysis

The thermal characteristics of both PP granules and optimized composite (F2) were introduced in Figure 6.12 and Figure 6.13 respectively. As shown in Figure 6.12, the degradation process of PP started at about 380°C and at 500°C temperature, 98.73% mass change was completed. However, the decomposition of the composite started earlier compared to the PP matrix which might be due to the interaction between fiber and matrix material of the composites (Figure 6.13).The addition of fibers decreased the thermal strength of the composites. As seen from the thermogravimetric curve of composite, the mass change started at 280°C.

DSC scans revealed the melting profile of matrix and composite. Figure 6.12 and Figure 6.13 revealed that the DSC scans of the PP matrix and composite showed peaks of melting at 151.7 and 151.4°C, respectively. The temperature peak of melting slightly decreased because of the influence of added fibers, showing an interaction between fiber and matrix.

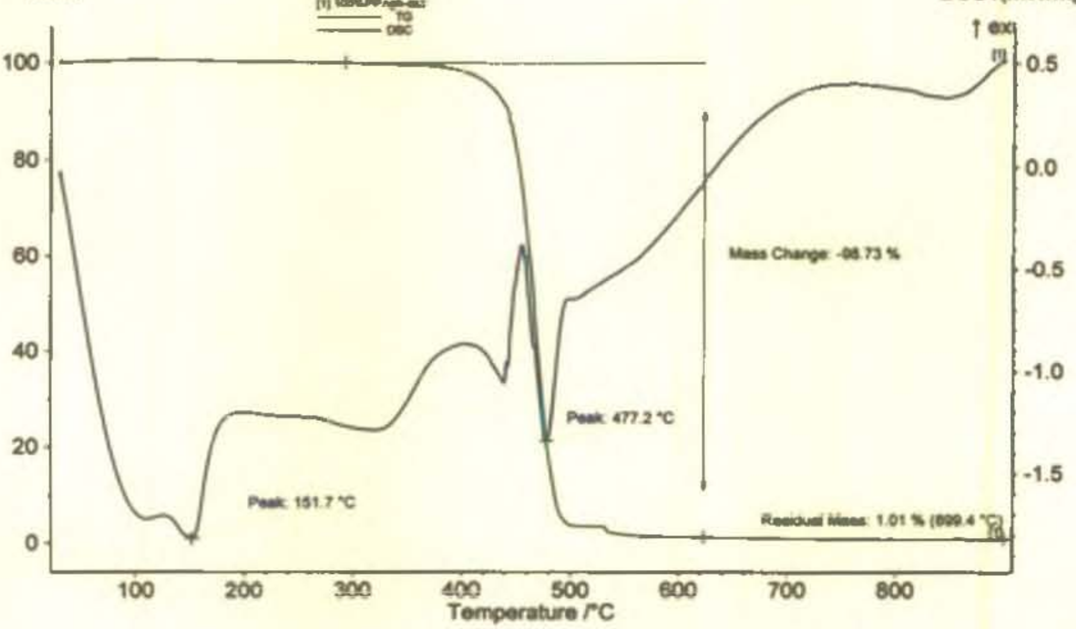

Figure 6.12: TGA and DSC of polypropylene.

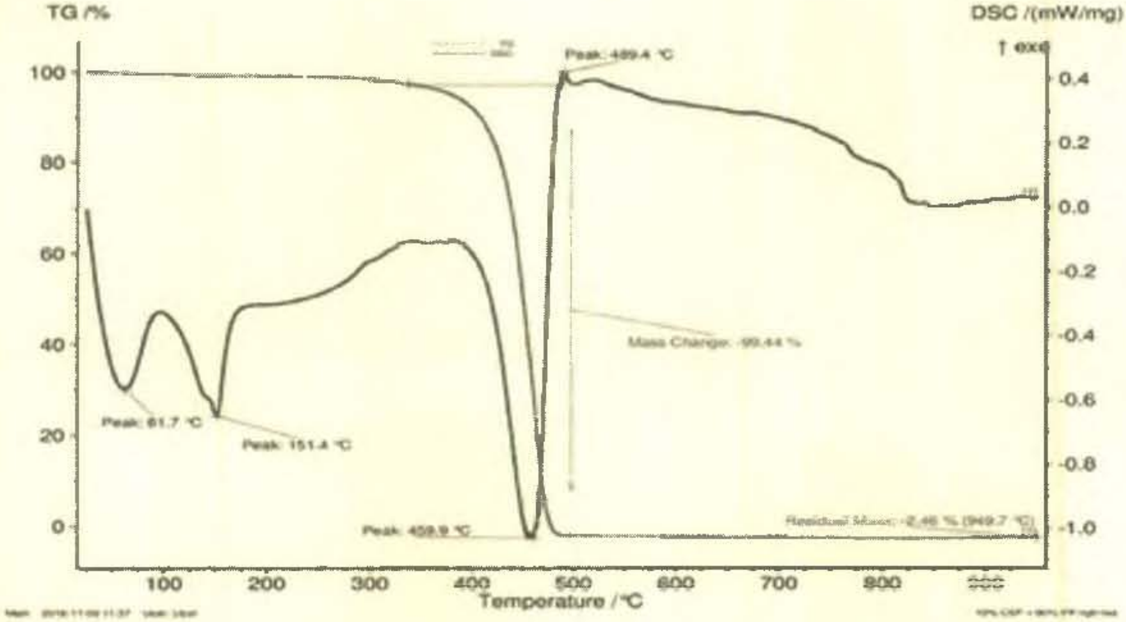

Figure 6.13: TGA and DSC of optimized composite F2 (90% PP +10% CSF).

6.4.6 Scanning Electron Microscopic (SEM) analysis

Morphological properties of coconut stem fibers reinforced polypropylene F2 (optimized composite) and F5 (overloaded fiber composite) composites were analyzed by SEM shown in Figure 6.14 and Figure 6.15 respectively. SEM examinations revealed that filler particles are well dispersed in the matrix material of optimized composite, as compared with the overloaded fiber composite. As shown in Figure 6.15, some voids are found where the fibers have pulled-out. These voids are responsible for weak interfacial bonding between fiber and matrix.

Figure 6.14: F2 composite

(10% CSF+90% PP).

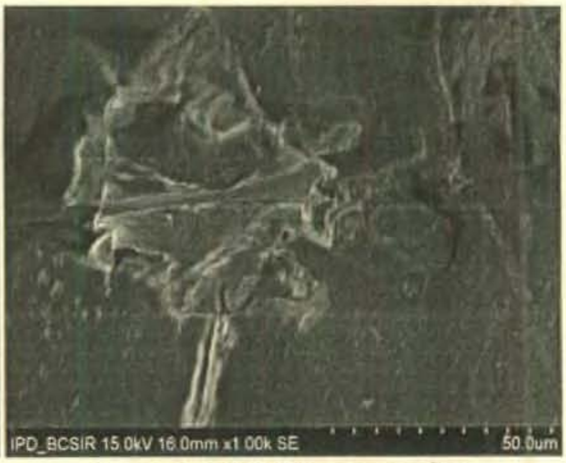

Figure 6.15: F5 composite

(25% CSF+75% PP).

6.5 Conclusion

The effect of coconut stem fiber content in the composites was evaluated via mechanical, morphological, water uptake, thermal and soil degradation test. The 10% coconut stem fiber content was found to be optimum which leads to better mechanical properties. Scanning Electron Microscopy images recommended that higher fiber content, beyond 10% fiber content, introduced poor fiber-matrix adhesion, which eventually was responsible for decreasing trend of mechanical properties above optimum fiber content. For all formulations, elongation at break (%) decreased might be due to the low elongation at break (%) of the CSF fibers compared to the matrix. Water uptake nature of F2 composite was nearly same of the F1 composite. Soil degradation test also indicated better resistance to soil degradation of F2 sample over degradation time. These composites would have a prospective application in the construction sectors where comparatively less strength is preferred.

6.6 References

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CHAPTER 7

Gamma Irradiated Areca nut Leaf-Sheath Fiber Reinforced Polypropylene Based Composites: Mechanical, Thermal and Morphological Analysis

7.1 Abstract

For the environmental concern natural fiber composite has a great demand. Due to renewable origin, the demand of natural fiber reinforcement of polymer based composites is growing day by day. Our present research work focus on preparation of γ- irradiated areca nut leaf-sheath (*areca catechu*) fiber reinforced thermoplastics composites as well as the study of their different properties. Areca nut leaf sheath fiber-matrix polypropylene (PP) composites were prepared by compression molding. Fiber reinforcement of the composites importantly increased the mechanical properties and the optimum improvement of strength and stiffness was achieved for the 10% fiber content. The water uptake character was done to investigate the hydrophilic nature of the composites. Moreover, degradation character and thermal properties of the composites were analyzed. Degradation test of the composites were executed up to six months in soil medium at ambient condition and appeared slow degradation of mechanical properties over time. Addition of fiber decreased the thermal stability of the composites. The structural and morphological features of the composites were also investigated.

7.2 Introduction

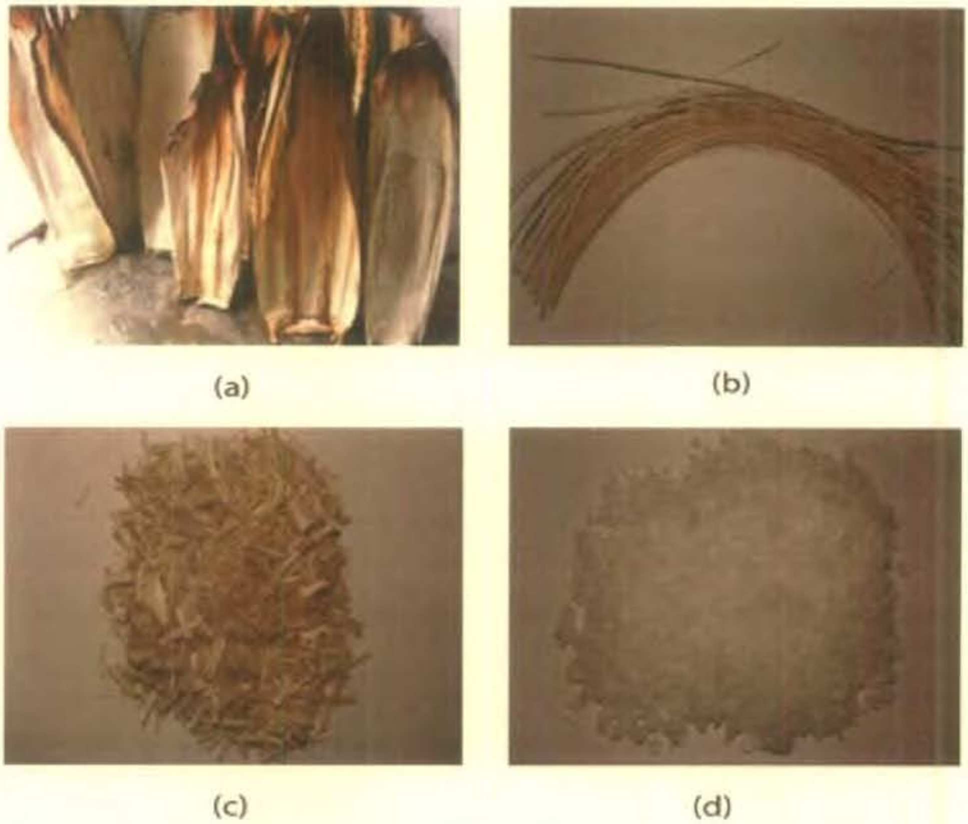
Outstanding mechanical, physical, chemical, thermal and electrical properties are observed in polypropylene but these are not found in any other thermoplastic material [1-2]. Compare with LDPE (low density polyethylene) or HDPE (high density polyethylene), its impact strength is lower, but tensile strength and working temperature is prevailing [3]. In the coastal area, *Areca catechu* trees are highly obtainable in our country which fabricates large amount areca nut leaf sheath [4]. Consider the environmental policies and renewable source, uses of natural fibers reinforced polymer matrix based composites are increasing. Uses of fillers and reinforcement materials generate from natural or renewable agricultural sources are superior both in case socio-economic characteristics and biodegradability property [5-8]. Agro-fiber reinforced polymer matrix based composites [9-12] create a new era in environmental perception.

Various types of natural fibers like areca nut leaf sheath (ALS), banana, oil palm, sisal, jute, sugarcane, wheat, flax straw, cotton, silk, coconut and bamboo which are extensively obtainable have demonstrated to be efficacious reinforcement in the thermoplastic and thermo-set matrix materials [13-20]. In this research work, our focal point on fabrication of γ- irradiated areca nut leaf-sheath (*areca catechu*) fiber reinforced polypropylene based composites as well as the study of their different properties and permutated application within environmental legal framework. These γ- irradiated areca nut leaf-sheath (*areca catechu*) fiber reinforced composites may be used in the constructive purposes [21], door and door frames [22-25], furniture and housing sectors.

7.3 Experimental

7.3.1 Materials

Polypropylene was purchased from Polyolefin Company, Private Ltd., Singapore. To impart actual and homogeneous adhesion between fibers and matrix, PP was grinded to get 50-60 μm particles (Figure 7.1, d). Areca nut leaf sheath (Figure 7.1, a) was collected from *Areca catechu* trees. Then the leaf sheath was soaked into water for 15 days to obtain areca nut leaf sheath fibers (Figure 7.1, b). The water immersion made the areca nut leaf sheath soft and weakened the bond between the fiber and the resin and waxy materials, which eventually helped to peeling off the loosen fibers from the resinous materials. The peeled off fibers were cleansed using huge amount of fresh water and dried appropriately. Using hand scissors, the fibers were cut into 2-3 mm size (Figure 7.1, c) and all dirt's were taken out from the fibers with the help of mesh. Before the composites fabrication the fibers were vacuum-dried for 5 hours at 100°C temperature.



(a) (b)

(c) (d)

Figure 7.1: Areca nut leaf sheath (a), ALS fiber (b), chopped ALS fiber (c) and PP powder (d).

7.3.2 γ- irradiated areca nut leaf-sheath (*areca catechu*) fiber

Areca nut leaf-sheath (*areca catechu*) fiber will be irradiated by γ- irradiation (Co-60) from 100 Krad to 500 Krad doses at a dose rate of 300Krad / h.

7.3.3 Chemical composition of ALS fiber

Areca nut leaf sheath fiber contains lignin, fatty and waxy matters, α – cellulose, hemicellulose, pectic matters and aqueous extract and the amount of these components are shown in the Table 7.1. 19.59% lignin, 66.08% α – cellulose and 7.40% hemicelluloses are present in the fiber. Small particles of cellulose of areca nut leaf sheath fibers are cemented and surrounded together by hemicellulose and lignin.

Table 7.1: Chemical composition of Areca nut leaf sheath fiber.

Sl. No.	Name	%
1	Fatty and waxy matters	5.06
2	Aqueous Extract	0.72
3	Pectic matters	1.15
4	α - cellulose	66.08
5	Lignin	19.59
6	Hemicellulose	7.40
	Total	100

7.3.4 Composite fabrication

Hot press machine was used to prepare composites. A family of five formulations of γ- irradiated of different radiation doses ((100–500 krad) ALS fiber (5%, 10%, 15%, 20% and 25% fiber) reinforced PP composites was prepared by varying the fiber content as shown in the Table 7.2. All formulations were made using moulds in the heat press under 5 bar consolidation pressure and 190°C temperature for 5 min. The model of heat press was 3856, Carver, INC, USA. After 5 minutes, in a separate press the moulds were cooled down to room temperature under 5 bar pressure. The prepared composite is shown in the Figure 7.2.

Table 7.2: Different formulations of composites.

Formulation no.	Weight percentage of ALS and PP in different composites
F1	5% ALS + 95% PP
F2	10% ALS + 90% PP
F3	15% ALS + 85% PP
F4	20% ALS + 80% PP
F5	25% ALS + 75% PP

Figure 7.2: Finished product (composite).

7.3.5 Mechanical properties of the composites

The tensile properties of the γ untreated and treated with different radiation doses optimized composites were performed using an electro-mechanical testing machine under a maximum load capacity of 5kN with a gauge length of 20 mm and crosshead speed of 10 mm/min. But, in case of bending properties, the crosshead speed was set at 10 mm/min with a span distance of 40 mm. Tensile and bending tests were performed following DIN 53455 and DIN 53452 standards methods, respectively. Five specimens for each sample were examined in the instrument.

7.3.6 Water uptake

Water uptake analysis was done by soaking the pre-weighed dried samples of γ untreated and treated optimized composite in the distilled water at 25°C temperature. After recording the initial dry weight, the samples were soaked into water and kept for a fixed period and then removed from water and wiped out the excess surface water using tissue paper, and the wet weight (final weight) was taken. The equation of calculation of water uptake is -

$$\text{Water uptake} = \frac{W_f - W_i}{W_i} \cdot 100 \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

Where, W_i is the initial weight of the samples, and W_f is the final weight of the samples after fixed time.

7.3.7 Degradation tests of the optimized composite

Cellulosic material undergoes deterioration when buried in moisturized soil (at least 25%). To investigate soil degradation profile of the composites, γ untreated and treated optimized composites were carried out for six months burying the samples in humid soil. After set time point, the samples were taken out, cleansed with fresh water and dried for 8 h at 80°C temperature. The variation of TS was intermittently observed in order to compute the degradable nature of the composites in this humid condition.

7.3.8 FTIR analysis

Pure polypropylene, fiber, γ untreated and treated optimized composites were examined by Fourier Transform Infrared Spectroscopy (FTIR) with an attachment of ATR accessory. The spectra were recorded in the wave number ranges of 4,000-600 cm^{-1} using a Perkin Elmer instrument.

7.3.9 Thermal analysis

Due to temperature effects, the physical changes are occurred in the material of the composites [26]. The thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) of the PP, γ untreated and treated optimized composites were performed to find out the physical changes. The increasing rate of temperature was $20^{o}C$/min under inert nitrogen atmosphere and maximum temperature was $900^{o}C$.

7.3.10 Surface morphology

For the morphological analysis of the composites, SEM is an effective technique. Surface morphology plays an important role in case of composite type materials. The strong interaction between fibers and matrix exhibits superior tensile properties. The compatibility and distribution between the fibers and the matrix materials could be found using SEM analysis. S-4000 model of Hitachi brand field emission scanning electron microscope was used to investigate the fracture surfaces of the tensile specimens of both γ untreated and treated optimized composite samples operating at 5 kV.

7.4 Results and Discussion

7.4.1 Mechanical properties of the composites

For the application of composite materials, the foremost elements of the composites are mechanical properties. Tensile and bending tests of the composites were done to measure the strength and stiffness of the composites. Tensile strength, bending strength and Eb% etc. mechanical properties were calculated. As shown in Figure 7.3, the maximum TS value found for 250 Krad doses of optimized composites and the value is 36.71 MPa. Beyond 250 Krad doses treated composites exhibited a decreased strength with irradiated up to 500 Krad. Similar situation was found for BS and figure 7.3 showed the maximum value (58.13MPa) for 250 Krad irradiated composites. From 100 krad to 250 Krad the values were enhanced and the values were gradually declined from 250 Krad to 500 Krad.

Figure 7.4 showed the highest Eb% value (9.55) for 250 Krad irradiated composites and reduces with the increase of doses. For 350 and 500 Krad dose the values % elongation at break were 8.34 and 7.65 respectively.

From the above outcomes, it is observed that 250 Krad γ-irradiation for of optimized composites has the outstanding effect on the mechanical properties. 250 Krad γ-irradiation composite appeared the highest TS and BS owing to the intercross-linking between the neighboring cellulose molecules as well as better fiber-matrix adhesion. The increasing of γ-irradiation doses (above 250 Krad), the mechanical properties of the composites appeared a reducing trend and it may be due to the two opposing phenomena, namely, photo cross-linking and photo degradation that take place simultaneously under γ-irradiation excess doses as well as weak interaction between fiber and matrix in the composite [4].

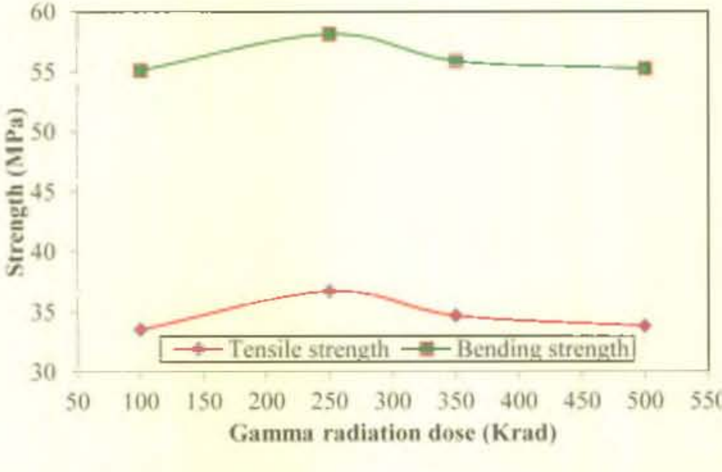


Figure 7.3: TS and BS of optimized (90% PP + 10% ALS fiber) composites against different radiation doses (100–500 krad).

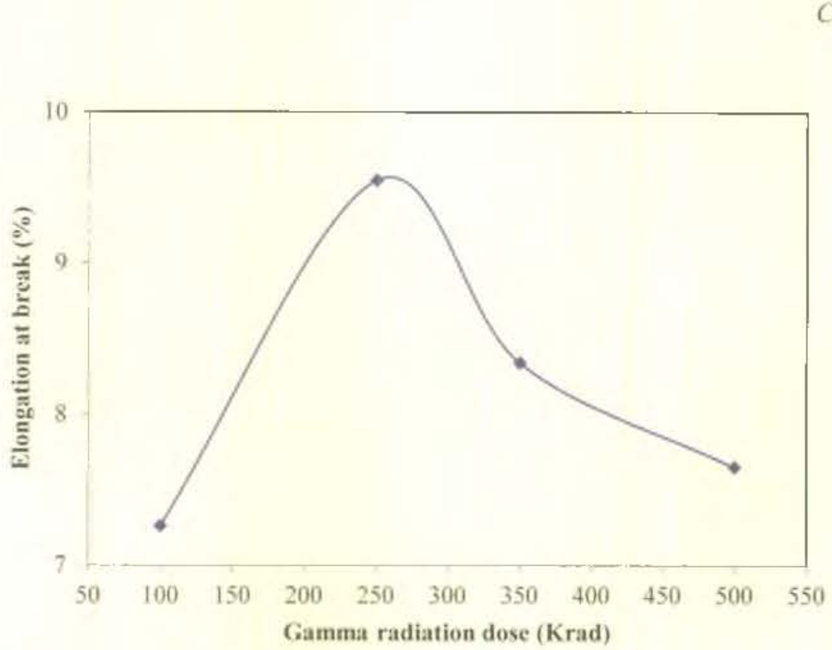


Figure 7.4: Elongation at break (Eb) of optimized (90% PP + 10% ALS fiber) composites against different radiation doses (100–500 krad).

7.4.2 Water uptake of the composites

Water uptake values of the 250 Krad γ treated and untreated composites were computed. The hydrophilic nature of the fiber influences the water absorption capacity. The subtraction value of initial weight from the final weight indicates the water uptake value. As shown in the Table 7.1, ALS fiber contains about 73.50% cellulose (α – cellulose and hemicelluloses) [27]. Figure 7.5 shows the water uptake profile of the 250 Krad γ treated and untreated composites. Untreated composites gained water up to 20 h but it was 30 h for γ irradiated composites. The minimal quantity of water absorption by the γ irradiated composites (0.11%) and the maximum quantity of water was computed by untreated composites (0.16%) for 60h examination.

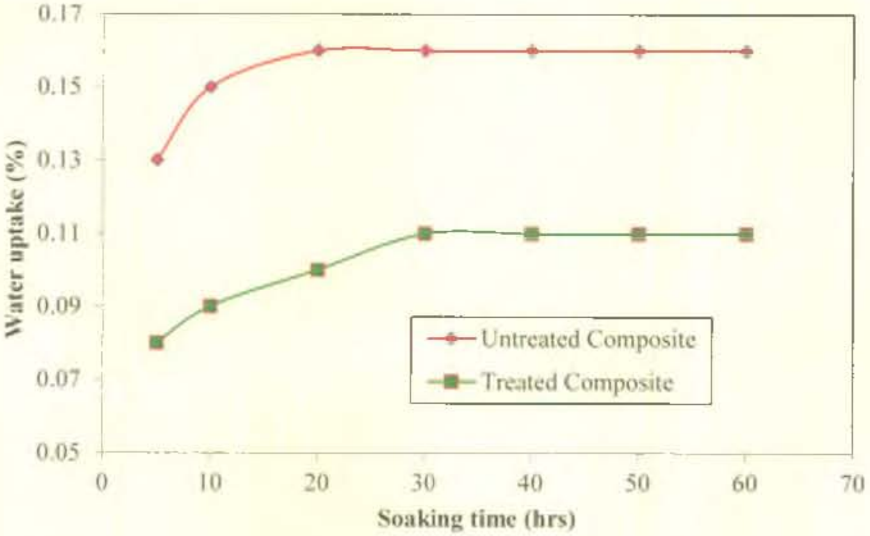


Figure 7.5: Water uptake (%) of optimized (90% PP + 10% ALS fiber) composites (untreated and 250 Krad γ treated) against soaking time.

7.4.3 Soil burial test

Soil burial test was performed to investigate the degradation trend of the treated and untreated composites samples in soil media for up to 24 weeks. Figure7.6 showed the Tensile Strength values against time (maximum 6 months). It was observed that in case of 250 Krad γ irradiated optimized (wt%) fiber reinforced composite, TS were declined slowly with degradation time compare with untreated composites. TS value of γ irradiated composites decreased gradually and it was 36.71, 34.66, 33.83, 33.12, 32.80, 32.13 and 31.76 MPa for 0, 4, 8, 12, 16, 20 & 24 weeks respectively. But it was 28.7, 26.7, 25.9, 25.1, 24.3, 23.2 and 22.80 MPa for untreated composite. It is clear that TS properties of the γ irradiated composites retained more compare to untreated composites and it may be due to the better interaction between fiber and matrix in the γ irradiated composites. During soil degradation test, water penetrates through the cutting edges of the composites which lead to the degradation of cellulose in the fiber, and such degradation is responsible for the diminished mechanical properties of the composites over time. In addition, bacterial attack could also responsible for the loss of the mechanical properties because biodegradable ALS fibers are normally attacked by microbial action [28].

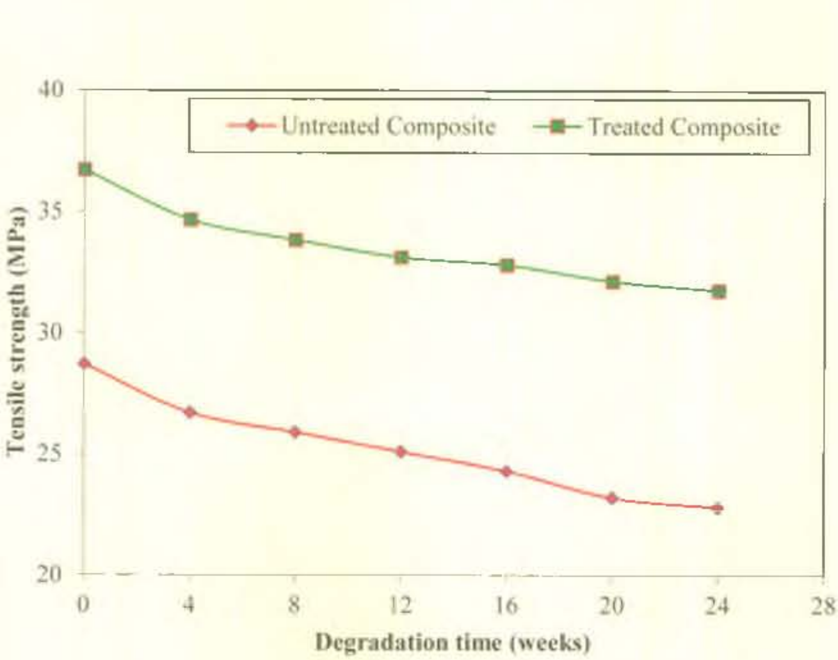


Figure 7.6: TS of optimized (90% PP + 10% ALS fiber) composites (untreated and 250 Krad γ treated) against soil degradation time (up to 24 weeks).

7.4.4 FTIR analysis

To analyze the structural characteristics of polypropylene, ALS fiber, optimized (10% ALS + 90%) PP composite and 250 Krad γ irradiated optimized (wt%) fiber reinforced PP composite, FTIR spectra is used. Figure 7.7, Figure 7.8, Figure 7.9 and Figure 7.10 showed the FTIR record of PP granules, ALS fiber, optimized (10% ALS + 90%) PP composite and 250 Krad γ irradiated optimized composite respectively.

As shown in Figure 7.7, the fingerprint region of pure PP was observed from 1455-600 cm^{-1}, particularly at around 1455 and 997 cm^{-1}ascribed to C=C stretching and C-H bending vibrations, respectively. The peak observed at 2916 cm^{-1} assigned to C-H stretching vibration. The spectrum of ALS fiber and composite appeared a peak at about 1740 cm^{-1} (Figure 7.8, 7.9 and 7.10) associated to carbonyl (C=O) stretching of acetyl groups of hemicellulose in the natural ALS fiber [29], along with characteristic peaks of polypropylene.

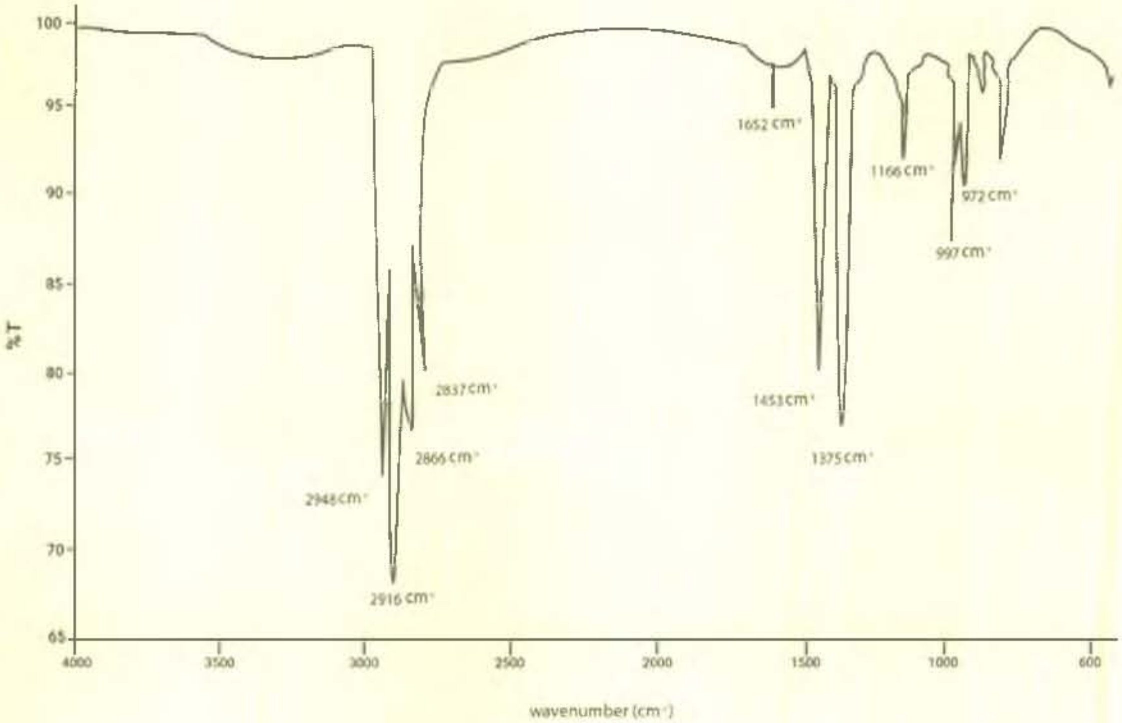


Figure 7.7: FTIR spectra of PP.

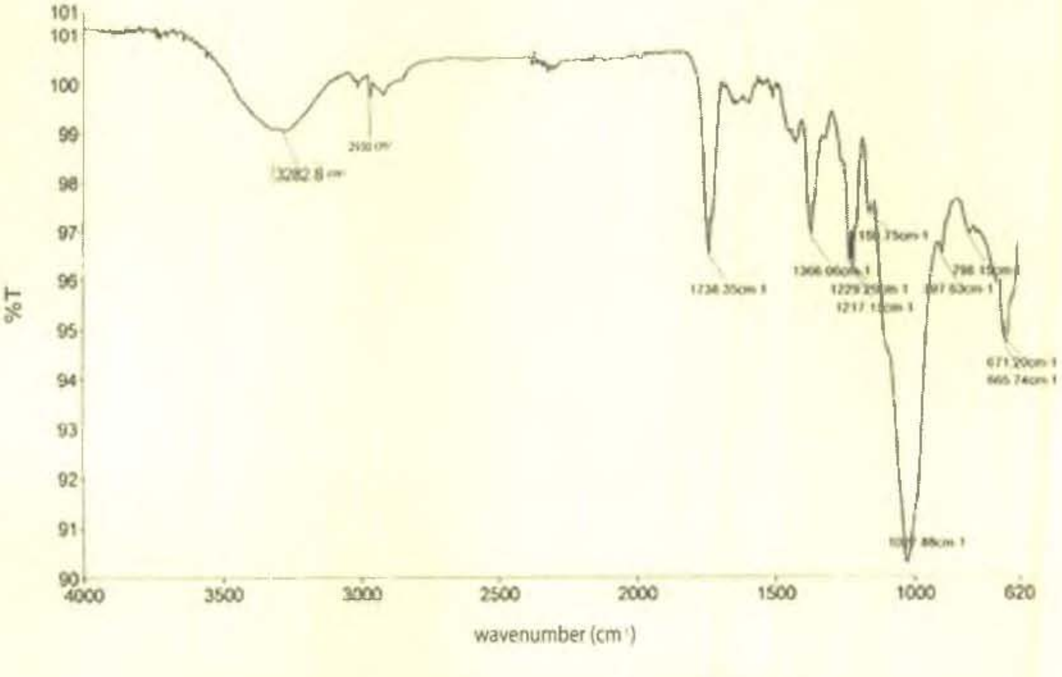


Figure 7.8: FTIR of ALS fiber.

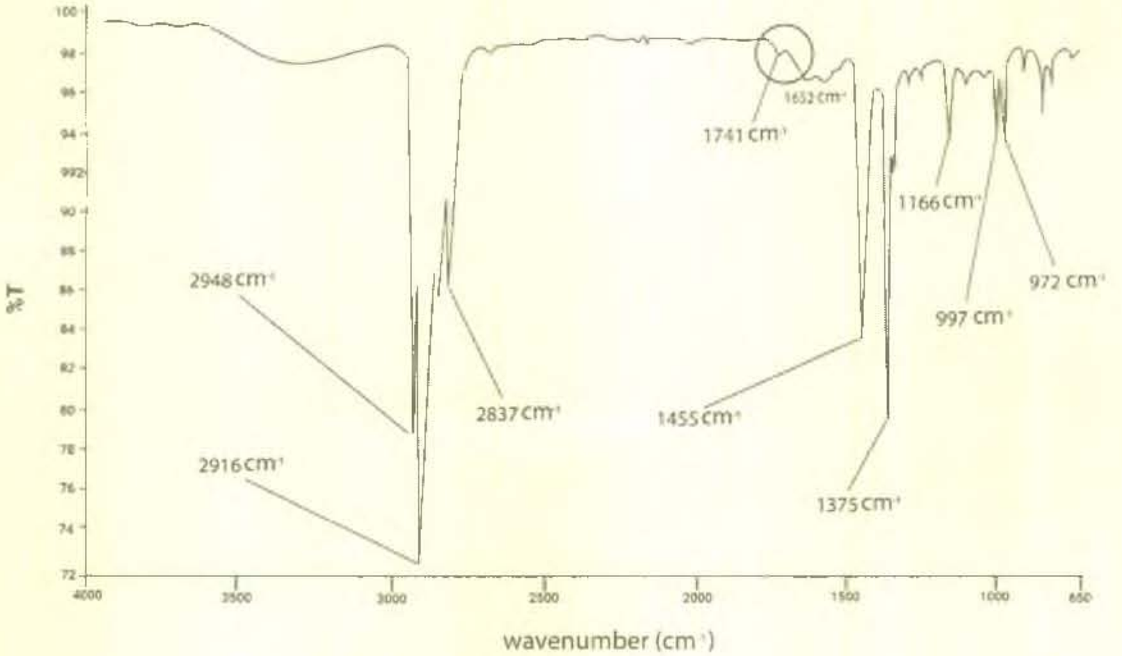


Figure 7.9: FTIR spectra of F2 composite.

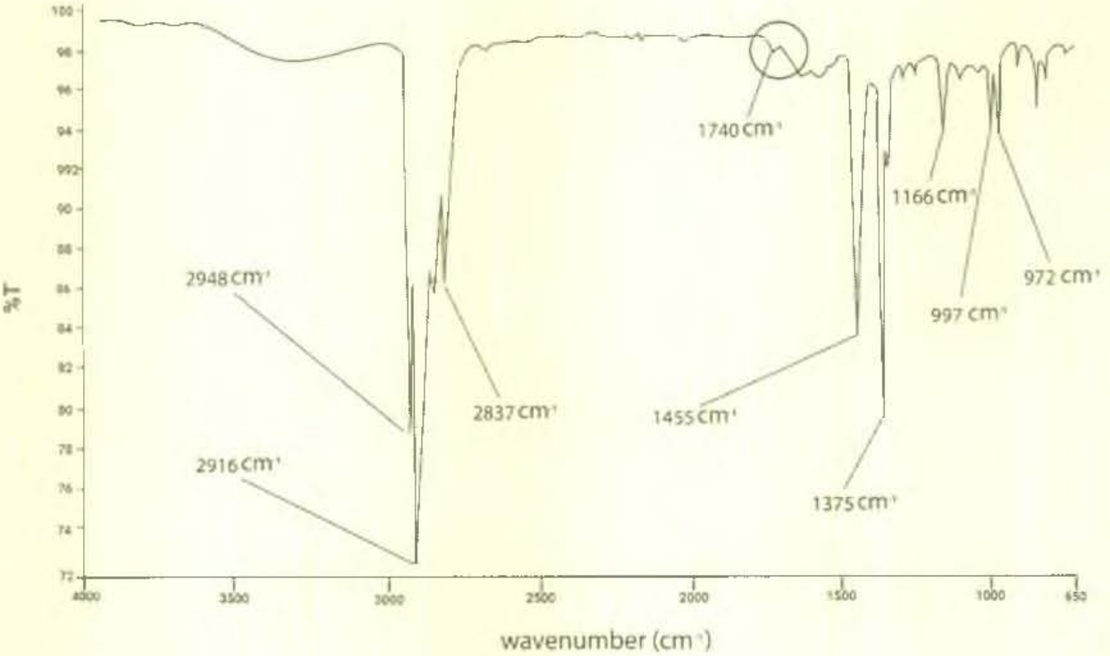


Figure 7.10: FTIR of 250 Krad gamma irradiated ALS fiber & PP blend composites.

7.4.5 TGA and DSC analysis

The thermal characteristics of PP granules, optimized composite and 250 Krad gamma irradiated composites were evaluated by TGA and DSC and presented in Figure 7.11, Figure 7.12 and Figure 7.13 respectively. As shown in Figure 7.11, the degradation process of PP started at about 380°C and at 500°C temperature, 98.73% mass change was completed. However, the decomposition of the composite started earlier (280°C) compared to the PP matrix and 10.38% degradation was completed at 420°C which might be due to the interaction between fiber and matrix material of the composites (Figure 7.12). But 4.73% mass change was completed at 250°C for 250 Krad gamma irradiated composites shown in Figure 7.13. Comparatively, the degradation of 250 Krad gamma irradiated composites started earlier may be due to theintercross-linking between the neighboring cellulose molecules of the composites [4]. DSC scans revealed the melting profile of matrix and composite. Figure 7.11, 7.12 and 7.13 revealed that the DSC scans of the PP matrix, optimized composite and 250 Krad gamma irradiated composites respectively and showed that in the all cases endothermic reactions were occurred and 477.2°C, 477.6°C and 463.0°C heat consumed respectively.

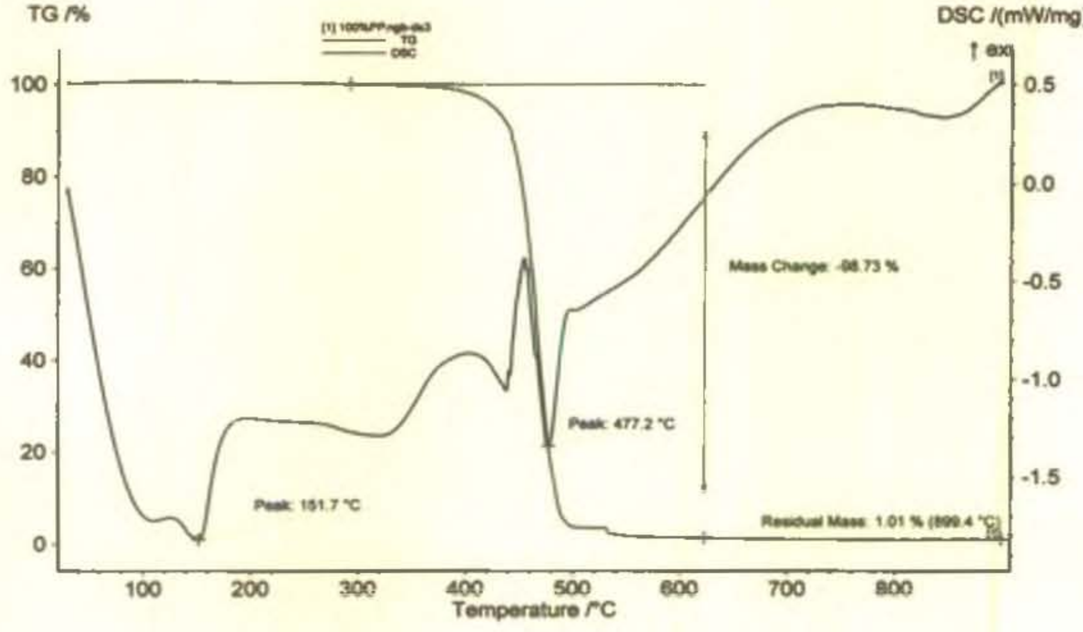


Figure 7.11: TGA and DSC of polypropylene.

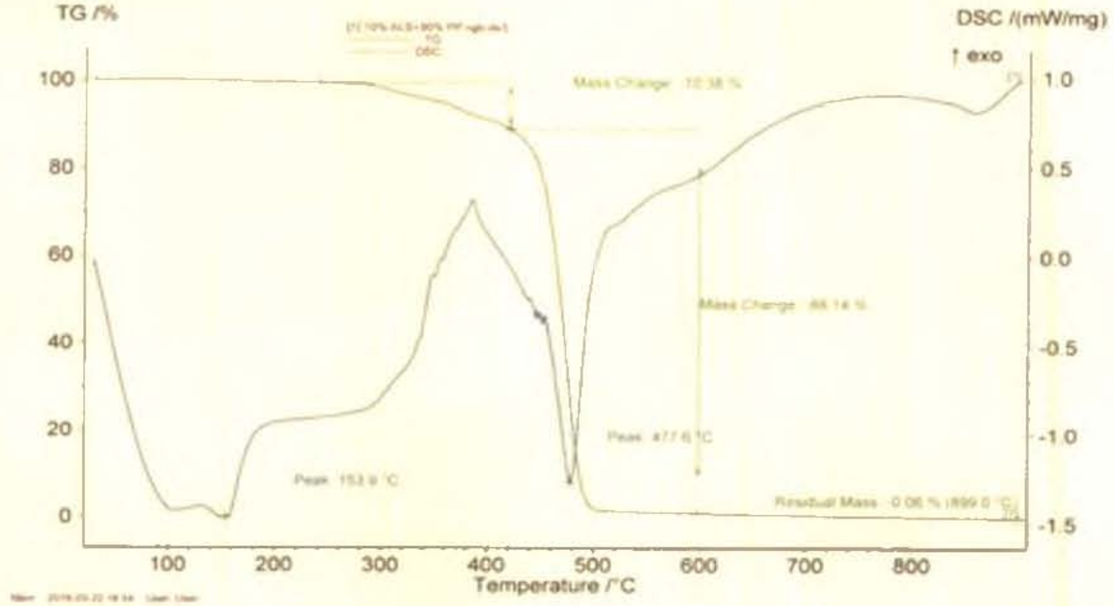


Figure 7.12: TGA and DSC of F2 composite.

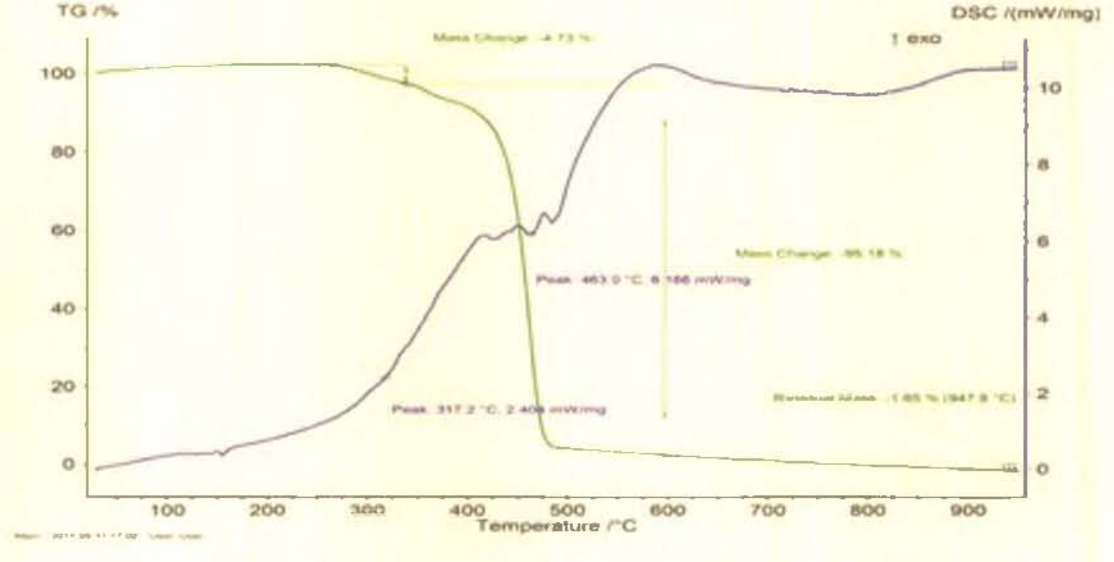


Figure 7.13: TGA & DSC of 250 Krad gamma irradiated 90% PP+10% ALS Fiber.

7.4.6 Surface morphology

Morphology of optimized polypropylene composite and 250 Krad gamma irradiated composite were analyzed by SEM (Figure 7.14 and Figure 7.15). SEM examinations represented that there was a significant dissimilarity in the fiber-matrix interaction between untreated (Figure 7.14) and γ-irradiated (Figure 7.15) composites. ALS is a strongly hydrophilic and natural biodegradable fiber. However, PP is strongly hydrophobic in nature. Thus it was expected poor fiber-matrix interaction for ALS-PP composites and SEM image reflected it (Figure 7.14). The hydrophilic nature of ALS fiber was reduced due to surface treatment with γ-irradiation which contributed to increase the adhesion property between fiber and matrix and ultimately improved the mechanical properties of the composites (Figure 7.15). Again, gaps between fiber and matrix were not observed for the irradiated SEM image [4].

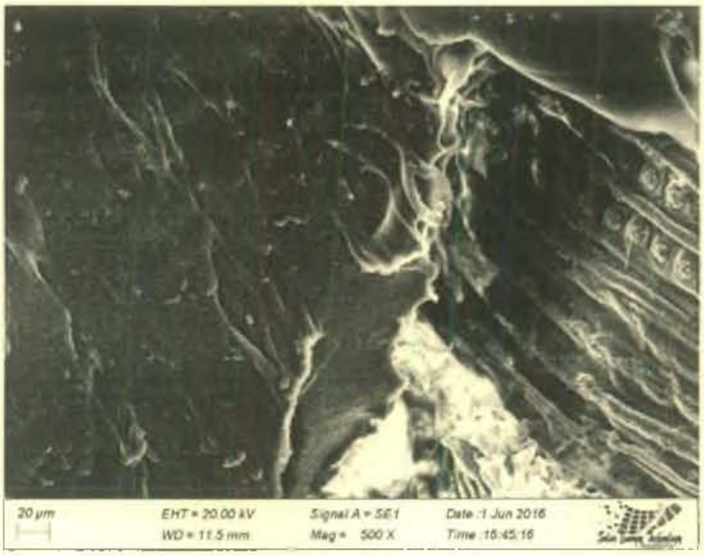


Figure 7.14: Image of (90% PP + 10% ALS) fibers reinforced polypropylene composites.

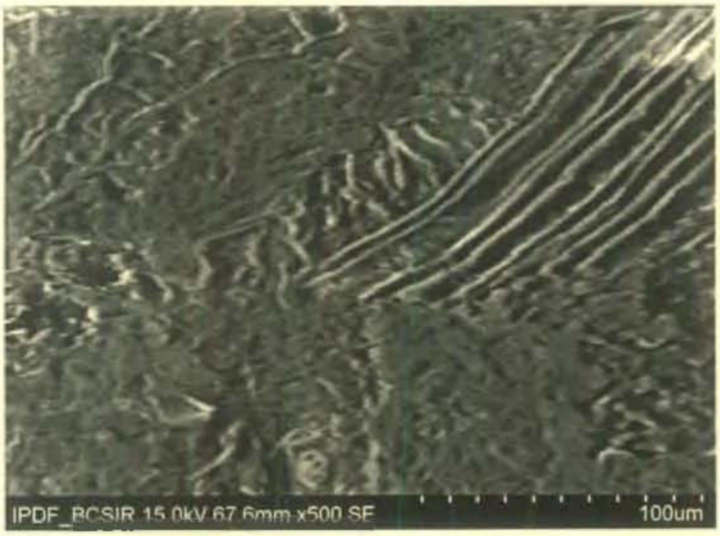


Figure 7.15: Image of 250Krad Gamma treated 90% PP + 10% ALS fiber prepared composites.

7.5 Conclusion

The aim of this study was to evaluate the advantages of γ-irradiated fiber reinforced PP composites. Gamma treated ALS-PP composites were produced by hot press machine. Composites prepared with 250Krad Gamma treated 10wt% fiber showed better mechanical properties and least amount (4.73%) of mass change occurred at the earlier stage due to the intercross-linking between the neighboring cellulose molecules. SEM supported the idea and mechanical properties were declined with enhancing of γ-irradiated doses. Elongation at break maximum for 250Krad Gamma treated composites and declined with the intensified γ-irradiated doses. Water uptake nature of treated optimized composite was lower compare with untreated composites. With respect to degradation time soil degradation studies revealed that tensile strength of Gamma treated composites could be lower than that of the untreated composites. Consequently, Gamma treated composites would be considered excellent building and construction materials which have superior mechanical and thermal effects.

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CHAPTER 8

Areca nut Leaf Sheath Fiber Reinforced Polyvinyl Chloride/Polypropylene Hybrid Composites: Effect of Fire Retardants

8.1 Abstract

The use of polyvinyl chloride and polypropylene as a hybrid matrix material with short areca nut leaf sheath fiber enhance the mechanical properties (TS, BS, TM and BM). 45% of polyvinyl chloride and 55% polypropylene mixed matrix material with 10% fiber reinforced composites appeared maximum mechanical properties. Scanning electron microscopy (SEM) also investigated. MgO was used as fire retardants with the composite material at different ratio and ignition time and firing time was identified. 30% and 20% MgO based composites represented highest ignition time and firing time respectively.

8.2 Introduction

Based on the source, natural fibers are classified of two types - such as plant fibers and animal fibers. About 3000 years ago, natural fibers were first time used to build walls with straw and clay in composite system in the ancient Egypt [1]. However, now-a-days composites have versatile uses. Depending on the physical and mechanical properties of the composites, it used in various fields as well as particular application [2]. It is noted that polymeric composites with natural fibers are with low cost and light in weight which also shows high strength and dielectric properties [3]. Scientist studied the characteristics of hybrid composites and appeared that hybrid composite adds advantages of weight saving, cost saving, increased strength etc [4]. Ignition time and total firing time of the hybrid composite was increased by adding fire retardant chemicals [5].

In this work, hybrid composite was prepared with polyvinyl chloride and polypropylene matrix material. Mechanical and morphological properties were analyzed [6-8]. MgO was added with the composite as a fire-retardant. Ignition time and firing time of the composite was studied.

8.3 Experiment

8.3.1 Materials

PP (Polyolefin Company, Private Ltd., Singapore), polyvinyl chloride (PVC) and MgO (Bayer, Germany) purchased from the local market of Bangladesh. ALS fibers were made ready from areca nut leaf sheath by water logged the fibers into water for two weeks. The water immersion made the areca nut leaf sheath soft and weakened the bond between the fiber and the resin and waxy materials, which eventually helped to peeling off the loosen fibers from the resinous materials, then the fibers cleaned with pure water and then air dried appropriately. Using hand scissors, the fibers were cut into 2-3 mm size and all dirt's are taken out from the chopped fiber. Then the chopped fibers were washed with distilled water and revealed rigorously to sunlight for about 24 hours. Ultimately, the fibers were dried in a vacuum oven at 100°C for 5 hours before the composites production.

8.3.2 Composite fabrication

Compression molding was used to make the polypropylene (PP) and polyvinyl chloride (PVC) matrix based unidirectional composites. At first, with the help of cleaning agent, the mould was cleansed. Then the dried ALS short 10wt% fibers with PP and polyvinyl chloride powder were used to prepare composites according to the ratio of PP:PVC = 25:75, 45:55, 55:45, and 75:25. All formulations were made using moulds in the heat press under 5 bar consolidation pressure and 190°C temperature for 5 min. The model of heat press was 3856, Carver, INC, USA. After heat press, the molds were cooled down to room temperature for few minutes in a separate press under 5 bar pressure.

MgO was added with optimized ALS fiber mixed PP-PVC hybrid composite in order to enhance the fire retardant property of the composite. 10g of moisture-free ALS fiber, 80g optimized PP-PVC matrix, and 10g of MgO powder were mixed with each other to produce 10% MgO mixed PP-PVC hybrid composite. Similarly, 20% and 30% of MgO mixed PP-PVC hybrid composites were produced and ignition time and total firing time were investigated.

Figure 8.1: Ignition time and total firing time experiment.

8.3.3 Mechanical properties of the composites

The tensile tests of the composites were performed using an electro-mechanical testing machine under a maximum load capacity of 5kN with a crosshead speed of 10 mm/min and gauge length of 20 mm. The crosshead speed was set at 10 mm/min with a span distance of 40 mm for measuring bending properties. Tensile and three point bending tests were performed following DIN 53455 and DIN 53452 standards methods, respectively. The average results of minimum five samples for each composite were revealed.

8.3.4 Surface morphology

Surface morphology plays an important role in case of composite type materials. For the morphological analysis of the composites, SEM is an effective technique. The compatibility and distribution between the fibers and the matrix materials could be found using SEM analysis. The strong interaction between fibers and matrix exhibits superior tensile properties. Hitachi S-4000 model field emission scanning electron microscope was used to investigate the fracture surfaces of the tensile specimens of hybrid composites operating at 5 kV.

8.4 Results and Discussion

8.4.1Tensile and bending properties of the composites

Mechanical property of the composites is an important factor for the application of interests. As such, tensile and bending tests of the composites were done to measure the strength and stiffness of the composites. The tensile and bending properties are presented in the Figures 8.2 and 8.3. As observed from the Figures 8.2 and 8.3, the maximum tensile strength, bending strength, tensile modulus and bending modulus value observed for 55% PP-45% PVC with ALS fiber formulation and the values are 41.32 MPa, 63.44 MPa, 785 MPa and 977 MPa respectively.

The significant enhancement in the tensile and bending properties of the composites was obtained for 10% fiber and 55% PP-45% PVC produced composite due to better fiber-matrix interaction. Interestingly, higher amount of matrix materials reduced the tensile and bending properties of the composites due to lower fiber-matrix interaction.

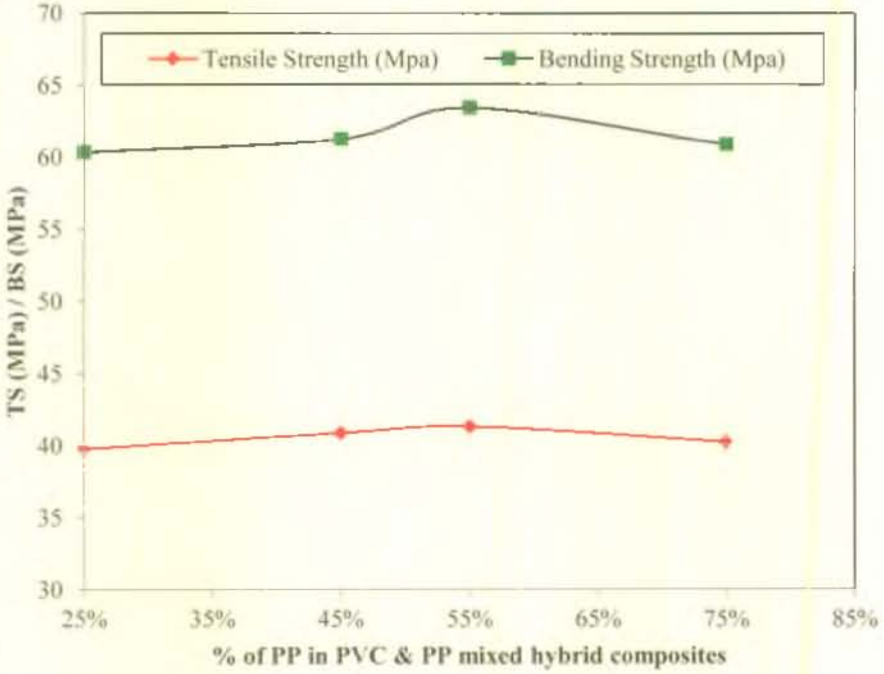

Figure 8.2: Tensile Strength & Bending Strength of different % of PP & PVC mixed hybrid composites.

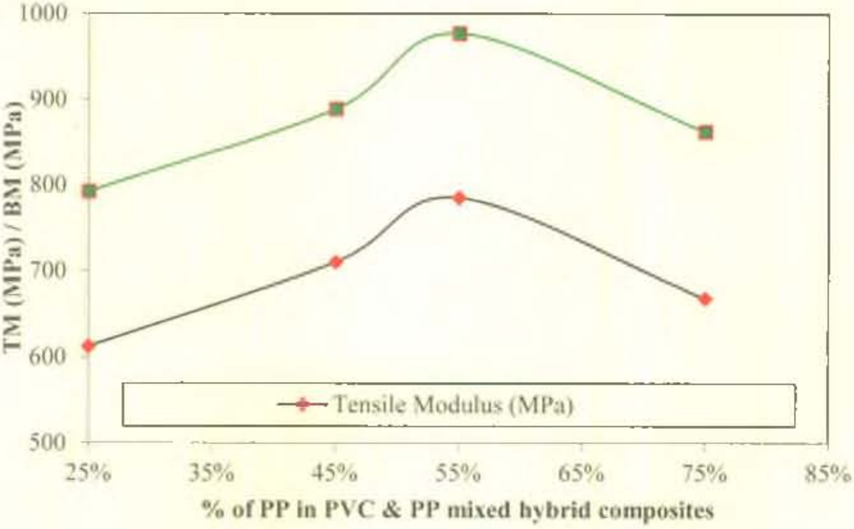

Figure 8.3: TM & BM of different % of PP & PVC mixed hybrid composites.

8.4.2 SEM analysis

The morphological properties of ALS short fibers reinforced polypropylene (PP) composites and PP/PVC (55% + 45%) mixed hybrid composites shown in Figure 8.4 and Figure 8.5 respectively were analyzed by SEM. It is worthwhile to mention that the properties of hybrid composites depends on the orientation and arrangements pattern of the fibers, fiber-matrix adhesion etc. Strong interaction between fibers and PP/PVC (55% + 45%) mixed hybrid composites, as evidenced by a scanning electron micrograph being taken on fracture surface composite concomitantly increases the mechanical properties.

Figure 8.4: 10% ALS fiber + 90% PP composites.

Figure 8.5:10% ALS fiber + PP/PVC (55% + 45%) mixed hybrid composites.

8.4.3 Ignition time (sec)

Figure 8.6 showed % of MgO vs ignition time (sec) of optimized hybrid composite. Ignition time of without MgO composite is 2.8 sec and 30% MgO mixed composite observed highest ignition time and it was 7.0 sec. Fire retardant compound, MgO hindrance burning which increases the ignition time.

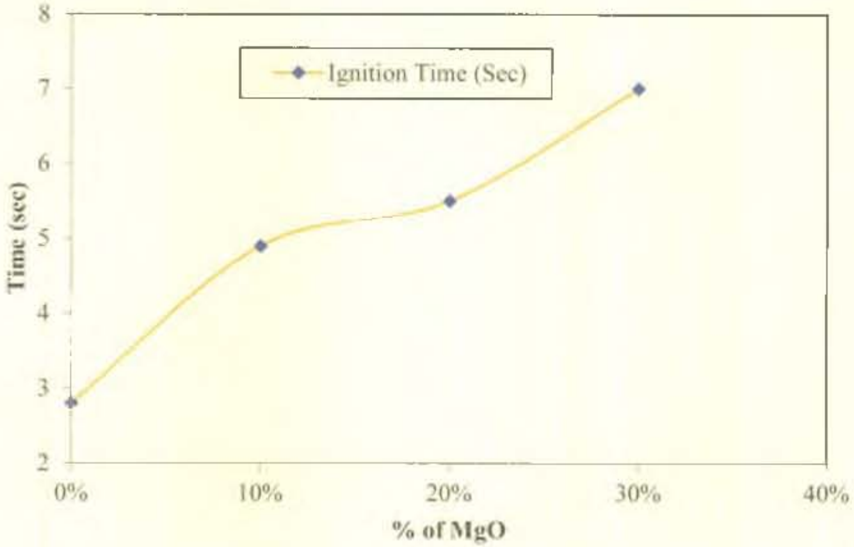

Figure 8.6: % of MgO vs Ignition Time (sec).

8.4.4 Total firing time (Sec)

Total firing time (sec) of optimized hybrid composite is investigated. Figure 8.7 showed % of MgO vs total firing time (sec) of optimized hybrid composite. Total firing time of without MgO composite is 180 sec and 20% MgO mixed composite observed highest firing time and it was 245 sec. MgO has fire retardant property which hindrance burning increases the firing time.

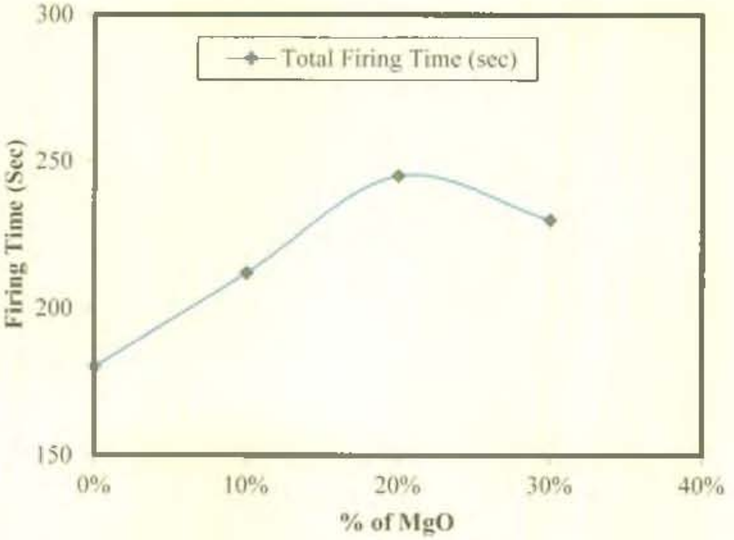

Figure 8.7: % of MgO vs Firing Time (sec).

8.5 Conclusion

This research indicates the highest mechanical properties of 10% fiber reinforced 90% PP/PVC (55% PP-45% PVC) mixed hybrid composite. The tensile strength, bending strength, tensile modulus and bending modulus of the above formulated composite are higher due to the better fiber-matrix adhesion. Ignition time for 30% MgO, and total firing time for 20% MgO content composites demonstrated good outcomes which were 7sec. and 245 sec. respectively.

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CHAPTER 9

Mechanical, Thermal and Morphological Analysis of Short ALS (Areca nut Leaf Sheath) Fiber Reinforced LDPE (Low Density Polyethylene) Composites: Effect of Chemical Treatment

9.1 Abstract

Short ALS (Areca nut Leaf Sheath) fiber (2-3 mm) reinforced low density polyethylene matrix based composites were prepared by compression mold machine using hot and cold press. ALS fiber is a cellulosic fiber also known as lignocellulosic fiber is renewable, environmentally friendly, cheap and completely biodegradable. Consequently, the demand of natural fibers is increased than synthetic fibers. Different fibers ratio (0, 3, 5, 7, 9 and 11wt %) were used to find out the maximum value of the ALS/LDPE composites. Among them 5% ALS fiber based LDPE composites appeared higher mechanical properties. TS, flexural strength, elongation at break (Eb%) were analyzed. SEM, TGA, DSC and biodegradation properties of ALS/LDPE composites were also analyzed. FTIR spectra of the LDPE and composites were also performed. Alkali treatment of the fibers led to increased mechanical, thermal and morphological properties was studied. Alkali treated fibers based composites appeared higher tensile strength and other properties compare to untreated fiber based composites due to chemical treatment enhanced fiber-matrix adhesion property.

9.2 Introduction

A good adhesion between fiber and matrix is essential to obtain full benefits of the composites [1]. Areca nut leaf sheath (ALS) fiber contains 66.08% α-cellulose (Table 9.1) which is more important to make a better attachment with matrix material. This fiber is a renewable, non abrasive and biodegradable [2]. The uses of this type of fibers are increasing gradually in construction and automotive sectors [3-7]. Thermoplastics [8] and thermosets [9-11] materials are prepared using short or long fiber reinforced polymer matrix based composites.

In the industrial applications low density polyethylene (LDPE) is extensively applied owing to low cost, moisture resistance, good chemical resistance and easily processed by all thermoplastic methods. Short ALS fibers are mixed with LDPE at different wt % ratio to get optimized wt% of fiber and matrix. Mechanical, thermal and morphological properties have been measured of the composites. Basically, natural fibers are hydrophilic in nature which is responsible for fiber matrix weak interfacial bonding as well as poor mechanical properties [12]. For better fiber matrix adhesion chemical treatment of fibers has been done.

9.3 Experimental

9.3.1 Materials

Low density polyethylene (LDPE) was procured from Polyolefin Company, Private Ltd., Singapore. To impart actual and homogeneous adhesion between fibers and matrix, LDPE was grinded to get 50-60 μm particles (Figure 9.1). Areca nut leaf sheath was collected from *Areca catechu* trees. Then the leaf sheath was soaked into water for two weeks to obtain areca nut leaf sheath fibers. The water immersion made the areca nut leaf sheath soft and weakened the bond between the fiber and the resin and waxy materials, which eventually helped to peeling off the loosen fibers from the resinous materials. The peeled off fibers were cleansed using huge amount of fresh water and dried appropriately. Using hand scissors, the fibers were cut into 2-3 mm size and all dirt's were taken out from the fibers with the help of mesh. Before the composites fabrication the fibers were vacuum-dried at 100°C for 5 hours.

Figure 9.1: Image of grind LDPE.

9.3.2 Chemical composition of ALS fiber

Areca nut leaf sheath fiber contains fatty and waxy matters, α–cellulose, lignin, hemicellulose, pectic matters and aqueous extract and the amount of these components are shown in the Table 9.1. 19.59% lignin, 66.08% α – cellulose and 7.40% hemicelluloses is present in the fiber. Small particles of cellulose of areca nut leaf sheath fibers are cemented and surrounded together by hemicellulose and lignin.

Table 9.1: Chemical composition of Areca nut leaf sheath (ALS) fiber.

Sl. No.	Name	%
1	Pectic matters	1.15
2	Fatty and waxy matters	5.06
3	α - cellulose	66.08
4	Hemicellulose	7.40
5	Lignin	19.59
6	Aqueous extract	0.72
	Total	100

9.3.3 Chemical treatment

The purpose of chemical treatment or chemical modifications of natural fibers is to develop the adhesion property between fiber and matrix. For chemical treatment, extra pure NaOH, Acetic acid glacial (99-100%) for neutralization and H_2O_2 (6% purified) are branded of Merck, Germany, purchased from the local market of Bangladesh.

9.3.3 (a) Alkali treatment

By alkaline treatment hydrogen bonding is disrupt in the structure, as a result increases the surface roughness [13]. Lignin, matters, fatty & waxy matters etc. of fibers are removed by this treatment. Again aqueous sodium hydroxide (optimized % of NaOH, i.e.10% NaOH =10 gm of NaOH in 100ml of water) [2] enhances the ionization of hydroxyl group of the natural fiber. 200g fibers are taken in 10% NaOH solution for 6 hrs [3, 14] at 70°C temperature [15] and shaking and stirring occasionally. Then 50% acetic acid glacial is used for neutralizing to eliminate absorbed alkali and cleansed with DI water perfectly.

$$\text{Fiber} - OH + NaOH \rightarrow \text{Fiber} - O - Na + H_2O + \text{impurity } [2]$$

9.3.3 (b) Bleaching with H_2O_2

Non-cellulosic parts of fiber such as lignin is removed after bleaching with H_2O_2 (6% purified) separates wax, fatty materials and lignin. 200g fibers are added to 10ml/L of 30% purified H_2O_2 at room temperature and stirring for 45 minutes. Then cleansed with DI water and dried in the oven at 50°C till reached at constant weight [2].

9.3.3 (c) Interfacial reaction between fiber and LDPE

<u>**Before chemical treatment**</u>

$$\text{Fiber} - OH + CH_2 = CH_2 \rightarrow \quad \text{Fiber} - CH = CH - \text{Fiber}$$

After chemical treatment

$$\text{Fiber} - \text{OH} + \text{NaOH} \rightarrow \text{Fiber} - \text{O} - \text{Na} + H_2O + \text{impurity [2]}$$

$$\text{Fiber} - \text{O} - \text{Na} + CH_2 = CH_2 \rightarrow \text{Fiber} - \text{CH} = \text{CH} - \text{Fiber}$$

9.3.3 (d) Chemical composition of untreated and treated ALS fiber

Table 9.2: Chemical composition of untreated and treated Areca nut leaf sheath fiber.

Sl. No.	Name of component (%)	ALS fiber	
		Untreated	**Treated**
1	Pectic matters	1.15	0.39
2	Fatty and waxy matters	5.06	2.96
3	α - cellulose	66.08	81.72
4	Hemi-cellulose	7.40	4.33
5	Lignin	19.59	10.11
6	Aqueous Extract	0.72	0.49
	Total	100	100

9.3.4 Composite preparation

The LDPE matrix based composites were formulated by compression molding. A family of five formulations of ALS fiber (0%, 3%, 5%, 7%, 9% and 11% fiber) reinforced LDPE composites was prepared by varying the fiber content as shown in the Table 9.3. All formulations were made using moulds in the heat press under 5 bar consolidation pressure and 150°C temperature for 5 min. The model of heat press was 3856, Carver, INC, USA. After 5 minutes, in a separate press the moulds were cooled down to room temperature under 5 bar pressure. The prepared composite is shown in the Figure 9.2.

Table 9.3: Different formulations of fiber and LDPE.

Formulation no.	ALS fiber (wt %)	LDPE Powder (wt %)
F1	0	100
F2	3	97
F3	5	95
F4	7	93
F5	9	91
F6	11	89

Figure 9.2: Image of Composite.

9.3.5 Determination of the tensile and flexural properties of the composites

The tensile tests of the composites were performed using an electro-mechanical testing machine under a maximum load capacity of 5kN with a crosshead speed of 10 mm/min and gauge length of 20 mm. For flexural properties, the crosshead speed was set at 10 mm/min with a span distance of 40 mm. Tensile and three point flexural tests were performed following DIN 53455 and DIN 53452 standards methods, respectively. Five specimens for each sample were examined in the instrument.

9.3.6 FTIR analysis

The structural analysis of the LDPE and composites was examined utilizing a Perkin Elmer Fourier transform infrared (FTIR) spectrometer with an attachment of attenuated total reflectance (ATR) accessory. The spectra were noted over the range of 4000-650 cm^{-1} at a resolution of 4 cm^{-1}.

9.3.7 TGA and DSC analysis

The thermogravimetric analysis (TGA) and DSC of the Low density polyethylene (LDPE) and optimized composite were performed to find out the physical changes occurring in the material of the composites due to temperature effects [16].

9.3.8 Surface morphology

Surface morphology plays an important role in case of composite type materials. The strong interaction between fibers and matrix exhibits superior tensile properties. Field emission scanning electron microscope (Hitachi S-4000) was used to investigate the fracture surfaces of the tensile specimens of both F2 and F6 composite samples operating at 5 kV.

9.4 Results and Discussion

9.4.1 Mechanical properties of the composites

Tensile strength, Young's Modulus, Elongation-at-break, Flexural strength, and Impact strength etc. mechanical properties are most important for composite type's materials. At untreated condition, the highest tensile strength value observed for 5% fiber based composite and it is 9.7 MPa and it is observed in Figure 9.3 and Table 9.4. From Table 9.5, it is clear that after alkali treatment tensile strength is increased approx. 20% and other property is also increased.

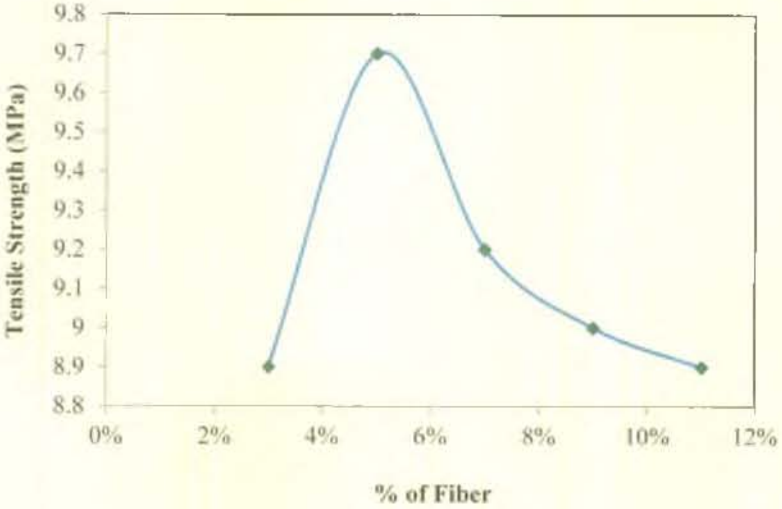

Figure 9.3: Tensile strength of different % of Areca nut leaf sheath fiber.

Table 9.4: Mechanical properties of LDPE/ALS fiber composites at different fiber loading [12]:

Fiber loading (wt%)	**Tensile Strength (MPa) Mean±SD**	**Young's modulus (MPa) Mean±SD**	**Elongation -at-break (%) Mean±SD**	**Flexural strength (MPa) Mean±SD**	**Impact strength (kJ/m^2) Mean±SD**
0	11.4±0.4	325±11	121±31	21.9±1.9	26.8±0.5
3	8.9±0.7	472±23	6±0.5	18.7±1.6	7.8±0.4
5	9.7±0.5	537±19	6±0.4	19.3±2.3	11.4±0.5
7	9.2±0.6	759±20	5±0.5	19.7±1.6	11.9±0.8
9	9.0±1.1	834±24	4±0.8	20.2±1.2	12.6±0.7
11	8.9±0.9	967±27	3±0.7	20.6±0.7	9.7±0.4

Table 9.5: Mechanical properties of LDPE/ALS fiber composites (at 5 wt% fiber loading) as a function of chemical treatment:

Fiber loading (wt%)	**Tensile Strength (MPa) Mean±SD**	**Young's modulus (MPa) Mean±SD**	**Elongation -at-break (%) Mean±SD**	**Flexural strength (MPa) Mean±SD**	**Impact strength (kJ/m^2) Mean±SD**
LDPE/UALS	9.7±0.5	537±19	6±0.4	19.3±2.3	11.4±0.5
LDPE/TALS	11.6	772±21	8±0.5	20±1.9	14±0.6

SD= standard Deviation, UALS = Untreated Areca nut Leaf Sheath, TALS = Treated Areca nut Leaf Sheath

9.4.2 Structural characterization

To study the structural characterization of LDPE and optimized LDPE-fiber composites, FTIR is a useful technique. FTIR spectra is utilized to determine the change of surface composition of the LDPE granules and optimized (wt%) fiber reinforced LDPE composites. Figure 9.4, Figure 9.5 and Figure 9.6 appeared the FTIR record of LDPE, optimized LDPE-fiber composite and after chemical treatment of the optimized (wt%) fiber based composite respectively.

From the Figure 9.4, it is observed that 719.7 cm^{-1} assigned to C-H bending vibrations, 1463.33 cm^{-1} ascribed to C=C stretching vibrations, 2847 and 2915 cm^{-1} assigned to C-H stretching vibrations.

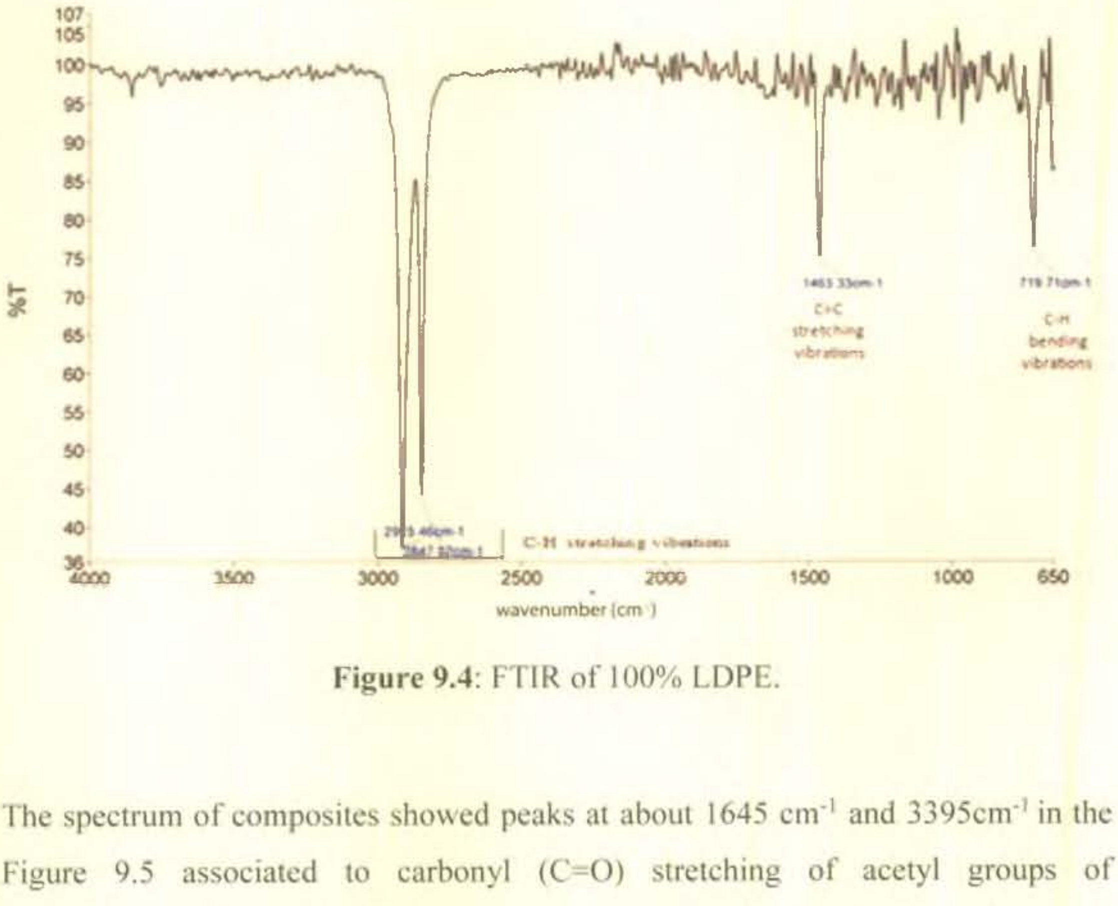

Figure 9.4: FTIR of 100% LDPE.

The spectrum of composites showed peaks at about 1645 cm^{-1} and 3395cm^{-1} in the Figure 9.5 associated to carbonyl (C=O) stretching of acetyl groups of hemicellulose and O-H stretching vibrations in the fiber respectively along with characteristics peaks of LDPE.

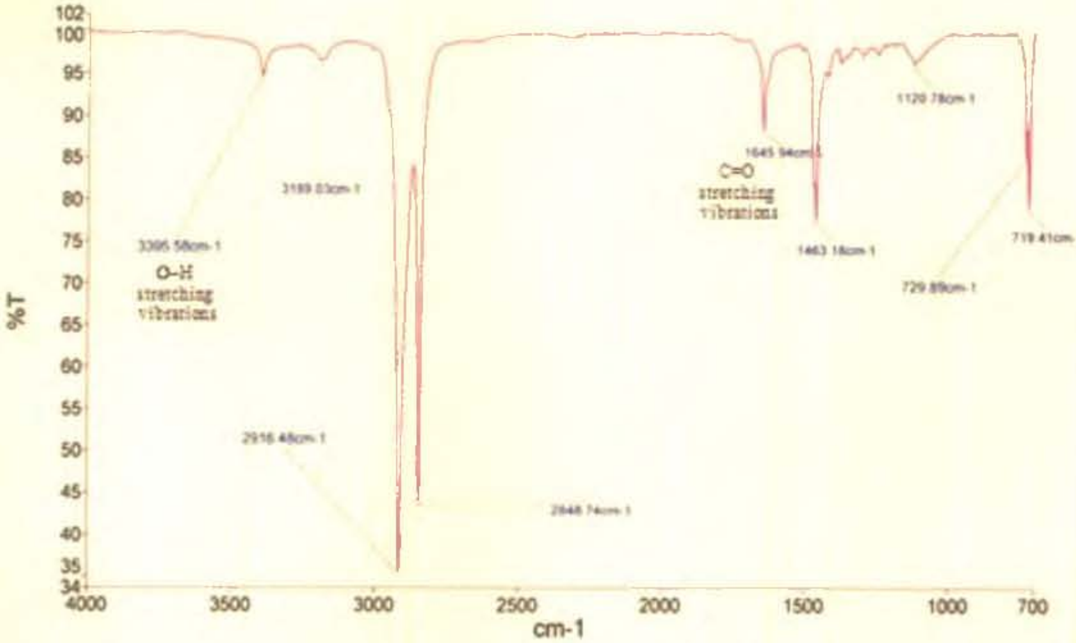

Figure 9.5: FTIR of 5% ALS + 95% LDPE.

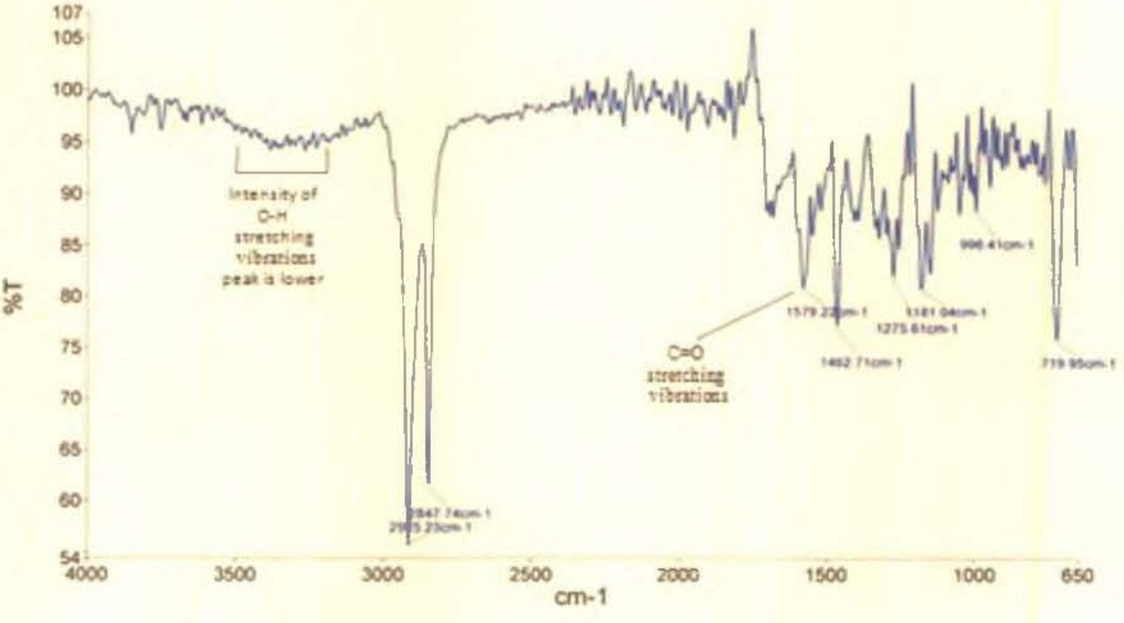

Figure 9.6: FTIR of 5% ALS (Treated) + 95% LDPE.

From the Figure 9.6, it is found that the spectrum of composites showed peaks at about 1645 cm^{-1} associated to carbonyl (C=O) stretching of acetyl groups of cellulose along with characteristics peaks of LDPE simultaneously intensity of O-H stretching vibrations peak is lower due to alkali treatment of fiber.

9.4.3 Thermal properties of the composites

TGA and DSC of LDPE granules, optimized composite (5 wt% fiber) and alkali treated composites were analyzed by following Figure 9.7, Figure 9.8, and Figure 9.9 respectively. As shown in Figure 9.7, Figure 9.8 and Figure 9.9 the degradation process of LDPE started at about 400°C and 99.15% mass change was completed at 500°C whereas it was about 380°C, 360°C and 92.23%, 98.55% mass change was completed at 500°C for the optimized and chemical treated composites respectively. The addition of fibers (Figure 9.8) and better fiber-matrix adhesion (Figure 9.9) decreased the thermal stability of the composites. The temperature peak of phase change of LDPE granules, optimized composite (5 wt% fiber) and chemical treated composites was 497.0°C, 492.3°C and 476.1°C respectively. The temperature peak of melting slightly decreased because of influence of added fibers, showing an interaction between fiber and matrix.

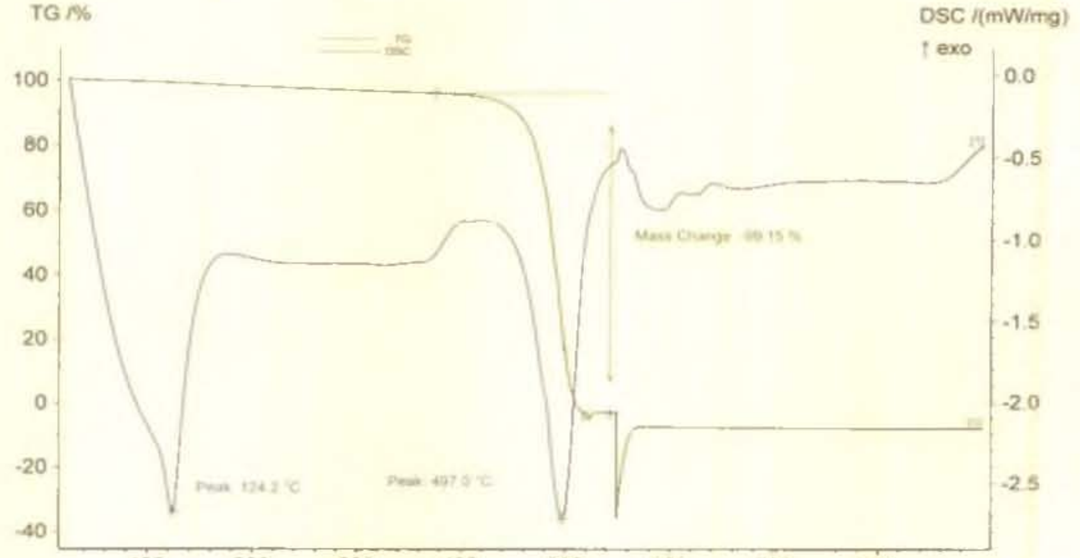

Figure 9.7: TGA & DSC of 100% LDPE.

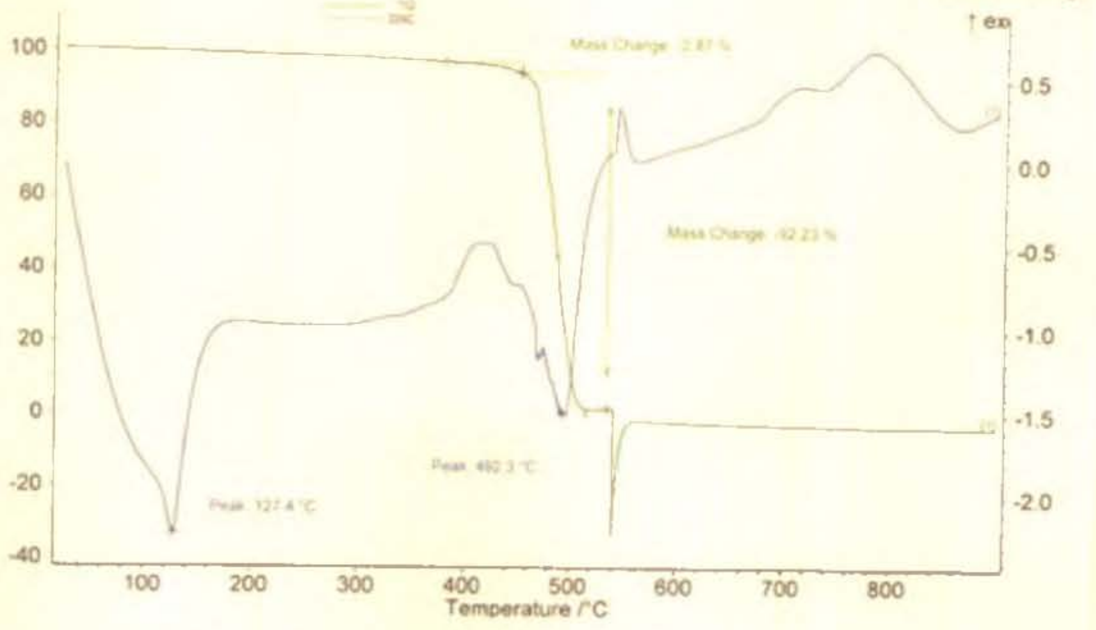

Figure 9.8: TGA & DSC of 5% ALS +95% LDPE

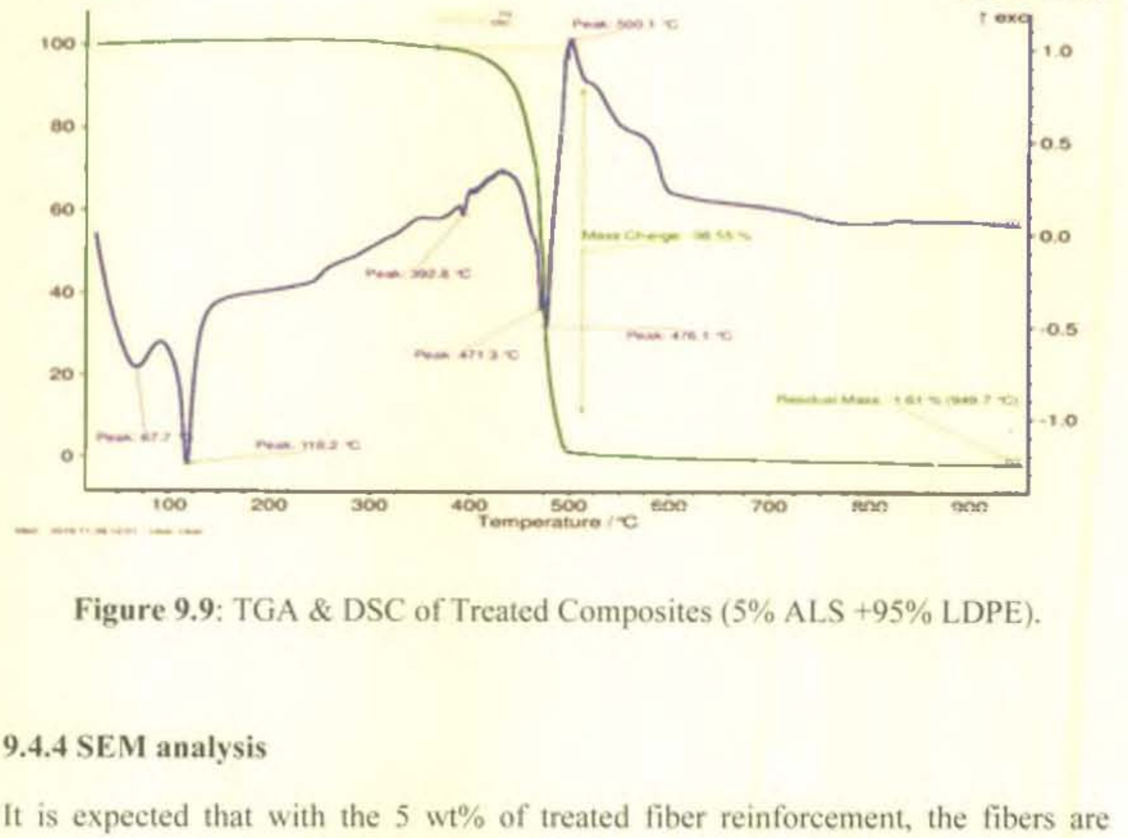

Figure 9.9: TGA & DSC of Treated Composites (5% ALS +95% LDPE).

9.4.4 SEM analysis

It is expected that with the 5 wt% of treated fiber reinforcement, the fibers are capable to provide to the effectual stress transfer between fibers and the LDPE matrix and concomitantly increases the tensile strength. The enhancement in the strength is attributable to the strong interactivity between treated fibers and the LDPE matrix, as evidenced by a scanning electron micrograph being taken on fracture surface of optimized composite containing 5 wt % fibers.

Figure 9.10: Image of 5% ALS + 95% LDPE composite.

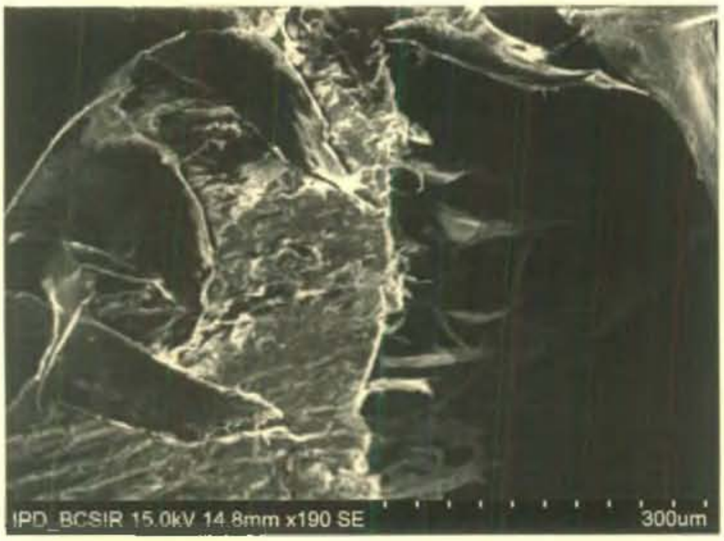

Figure 9.11: Image of treated fiber (5%) based LDPE composite.

9.5 Conclusion

Areca nut leaf sheath fiber based low density polyethylene (LDPE) composites were optimized with the help of mechanical properties and 5 wt% fiber content in the composites appeared higher mechanical properties. Mechanical properties were declined with the enhancing of fiber content owing to weak fiber-matrix adhesion. SEM and thermal properties supported the idea. Again, Eb declined with the intensified of fiber (wt%). Chemical treated fiber based composites appeared higher mechanical properties compare to untreated fiber based composites and morphological and thermal properties analysis supported the idea.

9.6 References

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CHAPTER 10

Nano Areca nut Leaf Sheath (ALS) Fiber-Gelatin Based PVA Film Preparation and Its Characterization

10.1 Abstract

Nowadays, a huge amount of agro waste materials are produced every day all over the world. Due to the many advantages of natural fibers from agro waste such as environmentally friendly, renewable and low cost etc., the uses of natural fibers are increased in the polymer industry. Natural fibers also known as lignocellulosic fibers contains lignin, α – cellulose, hemcellulose etc. Naturally, ALS fiber contains 66.08% of α – cellulose. After chemical treatment 81.72% α – cellulose extract from the fiber. In this present work, nano cellulose fiber (ALS) is used to prepare gelatin based PVA film. This nano cellulose has been blended with gelatin and PVA at different ratios. The mechanical, thermal and morphological status shows variation with different composition.

10.2 Introduction

The uses of polymeric materials at various purposes in life reach at a new level. Scientist and researchers are more concern and worried about non biodegradable polymeric products. Consider the environmental point of view, scientist and researchers are working to develop renewable or biodegradable polymeric materials using available materials in nature [1]. In this research work, nano cellulosic particles are generated from areca nut leaf sheath (ALS) fiber by acid hydrolysis with sulfuric acid [2-6]. Comparatively, highest amount of cellulose is obtained in ALS fiber and it is 66.08% [7]. Cellulosic part is separated from other parts of fiber using chemical treatment then nano particles [8-10] are produced by acid hydrolysis which is used with gelatin and PVA blend at different ratios. The structural characterization of the nano particles used gelatin based PVA film is studied. The mechanical, thermal and morphological properties of the film are analyzed. Among different formulations, GPnc4 (1% nano cellulose with "gelatin/PVA = 95/5") formulated film showed maximum strength and other properties are reported in this research work.

10.3 Experimental

10.3.1 Materials

From the Opsonin Pharma Limited, Barishal, Bangladesh, Gelatin (Type A, pharmaceutical grade) was collected. The synthetic polymer PVA (Molecular Weight: 72000, Fluka Chemie AGCH-9470 Buchs) and concentrated H_2SO_4 (E. Merck, Germany) were collected from the local market.

10.3.2 Nanocrystal suspension preparation

100% cellulose was extracted from ALS fiber by chemical analysis which was used for nano cellulose. For 1 g cellulose, 2 ml 64% H_2SO_4 was taken in a beaker and stirring with glass rod for 1 hour at room temperature. After 1 hour, 100 ml DI water was poured into the beaker and mixing properly. Then centrifuge the solution at 9500 rpm for 21 minute. Upper layer of water was removed and repeated the centrifuging step with adding 100 ml DI water until the solution become neutralized. Then the nanocrystal suspension was prepared at room temperature.

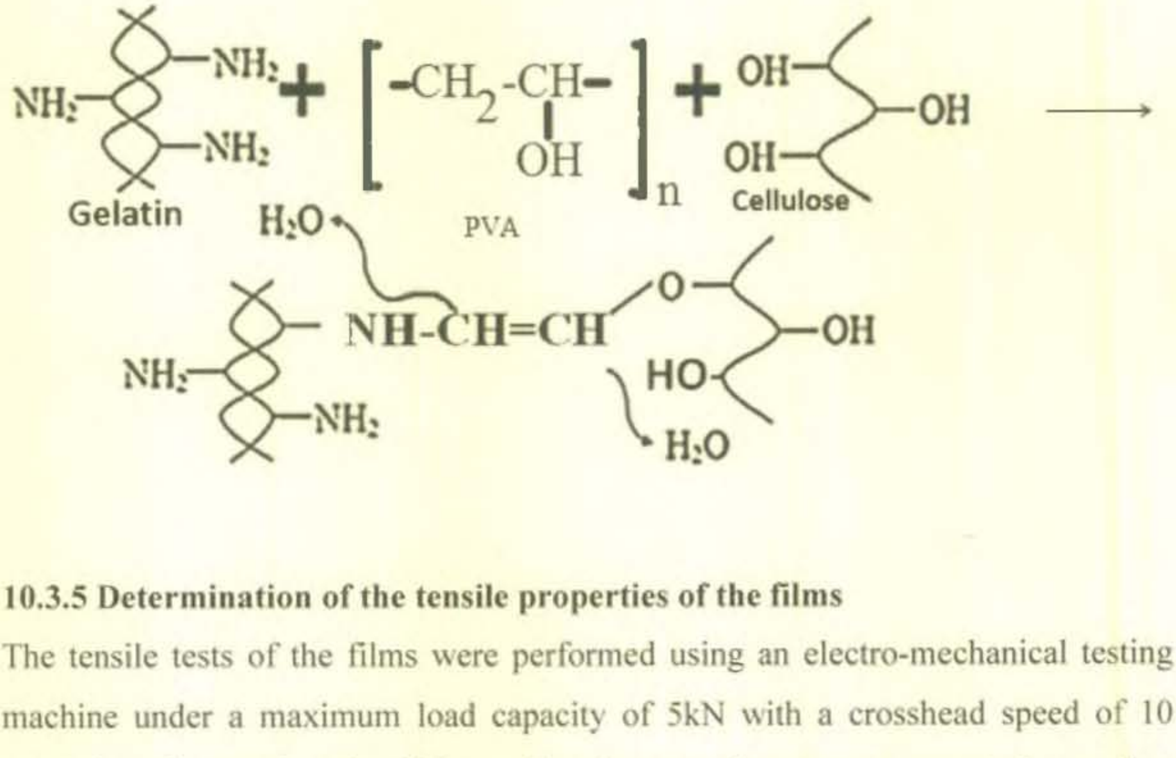
10.3.3 Gelatin-PVA/nano cellulose fiber blended film preparation

Gelatin and PVA were dissolved individually in hot water with continual stirring to produce the solutions. Then a measured quantity of PVA solution (5% w/w) and nano cellulose (1, 2 and 3% w/w used in Gelatin: PVA = 95:5) was mixed with gelatin solution according to Table 10.1. To prepare a homogeneous solution, the solutions were blended in hot water for about 90 minutes. The solutions were then poured onto the silicon paper-covered glass plate to form film. The solutions were maintained in a thickness of 4mm on the glass plate. Then the films were dried at room temperature for two days [11].

Table 10.1: Composition of different blending formulations (% w/w).

Formulations	Gelatin (%)	PVA (%)	Nano Cellulose (% used in Gelatin : PVA = 95:5)
GPnc1	100	0	0
GPcn2	0	100	0
GPnc3	95	5	0
GPnc4	95	5	1
GPnc5	95	5	2
GPnc6	95	5	3

10.3.4 Reaction among Gelatin, PVA &Cellulose [12]



10.3.5 Determination of the tensile properties of the films

The tensile tests of the films were performed using an electro-mechanical testing machine under a maximum load capacity of 5kN with a crosshead speed of 10 mm/min and gauge length of 20 mm. Tensile strength tests were executed according to DIN 53455 standards methods. Five specimens for each sample were examined in the instrument.

10.3.6 FTIR analysis

The structural study of the films was carried on applying a Perkin Elmer Fourier Transform Infrared (FTIR) spectrometer with an attachment of ATR accessory. The spectra were noted over the range of 4000-650 cm^{-1} at a resolution of 4 cm^{-1}.

10.3.7 Thermal analysis

Temperature is a primitive state mutable that affects most structural transformation, physical properties and chemical reactions. The thermogravimetric analysis (TGA) and DSC of the films used to find out the physical changes occurring in the material of the films due to temperature effects [13].

10.3.8 Surface morphology

In case of film type materials, surface morphology plays an important role. The strong adhesion between nano fibers-matrix such as gelatin and PVA exhibits superior tensile properties. Field emission scanning electron microscope (Hitachi S-4000) was used to investigate the breakage surfaces of the tensile samples of the films operating at 5 kV.

10.4 Results and Discussion

10.4.1 Mechanical properties of the films

Tensile strength and Elongation-at-break etc. mechanical properties of the films are most important features for the application of interests. The highest tensile strength value observed for 1% nano cellulose fiber added gelatin-PVA blended film and it is 39.2 MPa and it is observed in the Table 10.2.

Table 10.2: Mechanical properties of Gelatin-PVA/nano cellulose fiber blended film.

Formulations	**Tensile Strength (MPa) Mean±SD**	**Elongation -at-break (%), Mean±SD**
GPnc1 (Gelatin=100%)	41.34±1.7	3.32±0.4
GPnc2 (PVA-100%)	30.06±1.5	164.2±3.8
GPnc3(Gelain/PVA=95/5)	34.94±1.6	5.1±0.5
GPnc4(1% nano cellulose in GPnc3)	39.2±1.6	5.8±0.6
GPnc5(2% nano cellulose in GPnc3)	35.12±1.8	6.2±0.8
GPcn6(3% nano cellulose in GPnc3)	33.19±1.9	6.3±0.7

10.4.2 Structural characterization

To determine the structural characterization of gelatin, PVA, PVA with nano ALS & gelatin, PVA and nano ALS fiber film and optimized (wt%) nano cellulose blended gelatin-PVA film, FTIR is a useful technique. Figure10.1 showed the FTIR record of the films. From the Figure 10.1, it is observed that 1630-1657 cm^{-1} assigned to carbonyl (C=O) stretching of acetyl groups of nano cellulose, 1451 cm^{-1} ascribed to C=C stretching vibrations, 3152-3153cm^{-1}and 1542-1543 cm^{-1} assigned to N-H and C-N stretching vibrations respectively.

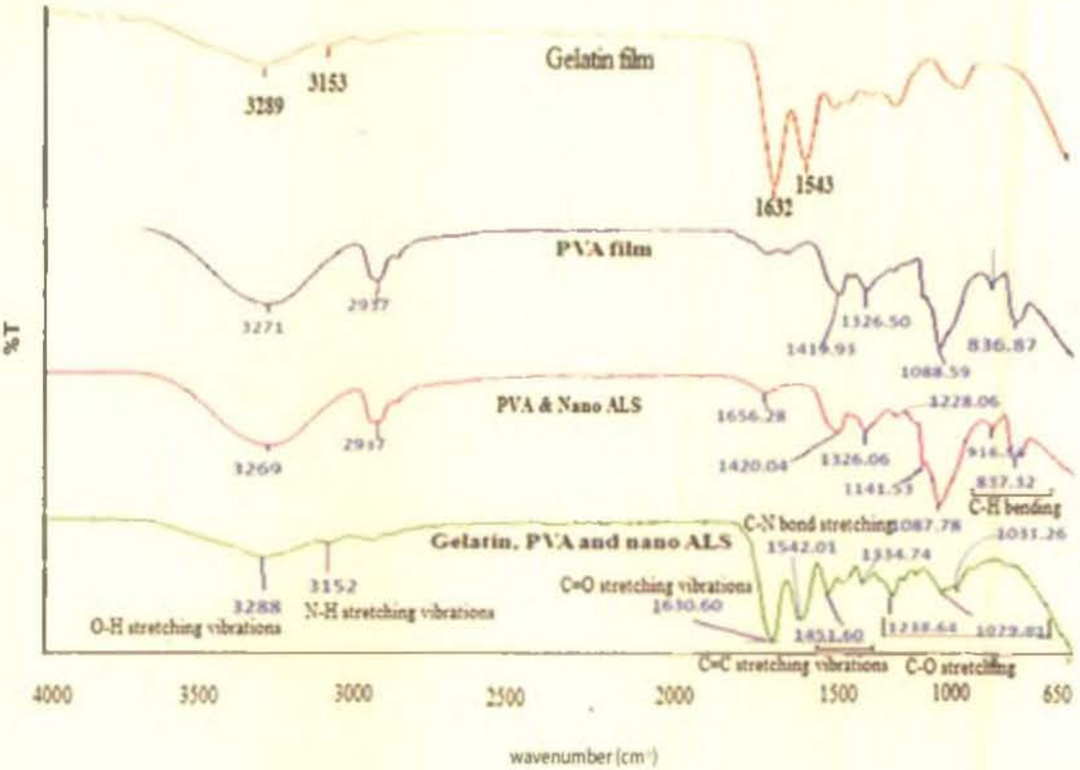


Figure 10.1: FTIR of gelatin, PVA, PVA with nano ALS & gelatin, PVA and nano ALS fiber film.

10.4.3 Thermal properties of the films

TGA and DSC of PVA & Nano ALS film and gelatin, PVA & Nano ALS film, were analyzed by following Figure 10.2 and Figure 10.3 respectively. As shown in Figure 10.2 and Figure 10.3 the degradation process of PVA & Nano ALS film and gelatin, PVA & Nano ALS film respectively and 6.95% mass change was completed at about 200°C temperature of the non gelatin film whereas 9.11% mass change was completed at 154.1°C for the gelatin, PVA & Nano ALS film. The better nano cellulose-matrix adhesion (Figure 10.3) declined the thermal stability of the film. The temperature peak of phase change of PVA & Nano ALS film and gelatin, PVA & Nano ALS film was 283.3°C, and 282.6°C respectively. The temperature peak of melting slightly decreased because of influence of added gelatin, showing an interaction between nano cellulose and matrix.

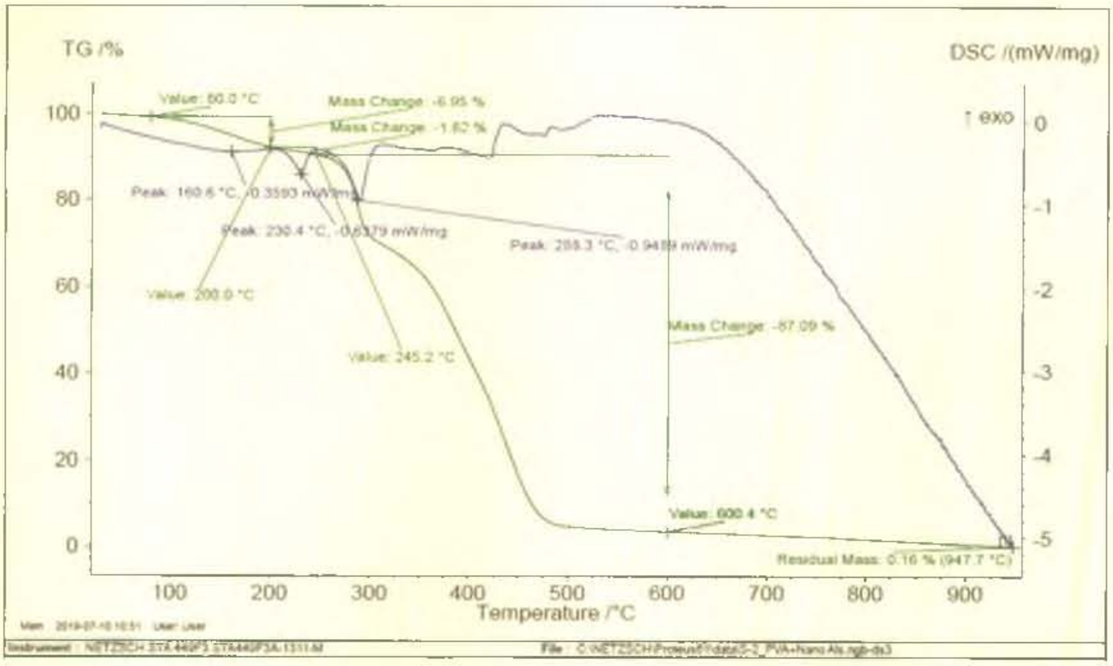


Figure 10.2: TGA & DSC of PVA + Nano ALS film.

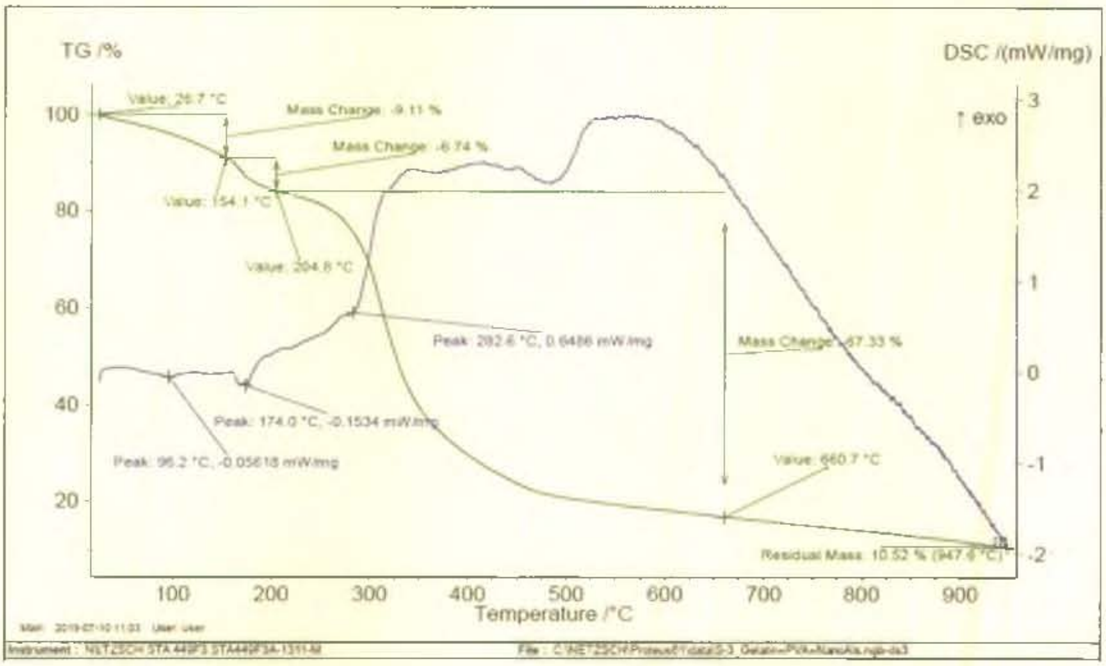


Figure 10.3: TGA & DSC of Gelatin, PVA & Nano ALS film.

10.4.4 SEM analysis

The nano particles are capable to provide to the effective stress transfer between gelatin and PVA and concomitantly increase the tensile strength shown in Figure 10.5. The improvement in the strength is attributable to the strong interaction among nano fibers with gelatin and PVA, as evidenced by a scanning electron micrograph being taken on fracture surface of optimized film containing 1 wt % nano fibers.

Figure 10.4: PVA + 1% ALS nano fiber film.

Figure 10.5: Gelatin, PVA & 1% ALS nano fiber film.

10.5 Conclusion

In this study reveal that 1% nano cellulose added PVA-95% gelatin blend film shows the highest TS (39.2 MPa), which is 12.19% higher than that of nano cellulose free PVA-95% gelatin blend film. It is also found that due to excess adding of nano cellulose (2% or 3%) in the PVA-gelatin blend film the TS values of the films are declined considerably while Eb values are enhanced. On addition, nano cellulose acts as filler between 5% PVA-95% gelatin blend film which enhance mechanical properties and morphological and thermal properties investigation supported the objective.

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CONCLUSIONS

CONCLUSIONS

The ambition of this analysis was to evaluate the benefits of reinforcing polypropylene (PP) with naturally areca nut leaf sheath, woven coconut leaf sheath and coconut stem short fibers. ALS fiber contained highest % of α-cellulose than that of others. Fibers filled composites were formulated by instrumental technique varying the fibers loading. Tensile and bending strength were intensified with the enhancing of fiber content up to 10 wt% and thus it was considered as optimum fiber loading. Again, reinforcement decreased the thermal stability of the composites. 250Krad Gamma treated 90% PP & 10% ALS fiber prepared composites showed highest quality than that of other doses. The highest TS, TM, TM and BM of the optimized wt% of ALS fiber used 55% PP in PP & PVC hybrid matrix based composite were found 41.32 MPa, 63.44 MPa, 785 MPa and 977 MPa respectively. It is noted that better fire retardant property was also attained for 20% MgO based ALS composites but in case of gelatin & PVA based 1% nano ALS fiber film, 30% MgO performed better result. In addition, 5 wt% ALS fiber-based LDPE composite represented almost highest mechanical properties. Alternatively, alkali treated ALS fiber prepared composites showed highest quality than that of untreated. The new agro-fibers based composites would make feasible to investigate latest utilizations in addition to new markets in fields of decorative and constructive purposes such as packaging, furniture, housing, automotive, aviation and shipping sectors etc.

LIST OF PUBLICATIONS

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2. **Pinku Poddar**, Shahin Sultana, Md. Ali Akbar, Husna Parvin Nur and Chowdhury, A. M. S., "Environmentally Sustainability of Short Areca nut Leaf Sheath Fiber Reinforced Polypropylene Composites", *MOJ Poly Sci.,* **2**(3):48-50, **2018**. DOI: 10.15406/mojps.2018.02.00046

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