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Utilization of Recycled Polypropylene for Production of Eco-composite Reinforced with Leaf Sheath Fiber of Betel-Nut Plant

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Abstract: The interface modification of leaf sheath fiber of betel nut was accomplished by NaOH & Silane coupling agent. The chemically treated (alkaline and silane) fibers are analyzed by FT-IR. The incorporation of these bio-fibers with Recycled Polypropylene (5-20%wt fiber) was carried out by twin screw extruder. The standard test specimens were prepared by injection molding. Mechanical and thermal characteristics of the blends systems were studied to evaluate the effect of chemically treated fibers content on recycled polypropylene. It has been found that as there is a decrease in tensile strength and elongation, the tensile modulus was found to be increased with the chemically treated fiber content. There is a good dispersion of both type treated fibers materials in recycled PP matrix result in an expansion in their use into more diverse, and technically demanding, application areas. Biodegradability (Soil Burial Test), properties of green composite is also tested. It suggest that these Eco-composite moderately biodegradable. This research paper seeks to provide use of recycled pp with non-food renewable material and consider the environmental and economic benefits of the materials.

Keywords: Biocomposites, Mechanical Strength, Recycled Polypropylene, Mercerization and Silanation

1. Introduction

Biocomposites offer a significant non-food market for crop-derived fibers and resins. Considerable growth has been seen in the use of Biocomposites in the automotive and decking markets over the past decade. Over the last decades, research has increasingly been conducted on renewable materials from Sustainable resources for a variety of applications. This has been influenced by the ever increasing demand for newer, stronger, stiffer, recyclable, fire repellent, less expensive and yet lighter-weight materials. Nowadays, polyethylene (PE) and polypropylene (PP) based plastic materials are being extensively used for commercial and household purposes. However, these polymer based plastics are substantially resistant to biodegradation. Thus their increasing accumulation in the environment is proving to be an ecological threat to the world. Consequently, over the last couple of decades exhaustive studies on the biodegradation of plastics have been carried out in order to overcome the environmental problems associated with synthetic plastic waste [1]. The improvement in the biodegradability of PP was observed by blending it with small additions (5%) of wood sawdust and wheat flour. Betel nut fiber (Bn)/polypropylene (PP) composites were prepared in the different ratio of 10:90, 20:80, 30:70, 40:60 (Bn wt%: PP wt%) using extruding and hot press molding technique. From the results, it can be inferred that Bn30: PP70 mixture composite (BnPP) showed better performance among the composites prepared [2].

The natural Activated Carbon (AC) composites prepared from carbon coconut shell reinforced with polypropylene (PP) [3]. The results showed that the tensile stress was increased when AC is increased specifically for sample 4 wt% and 8 wt%. Maximum tensile stresses lead by sample 4 wt% with

30 MPa. Two types of short natural fibers, namely jute and betel nut fibers, are employed as fibers for the formation of short fiber reinforced polypropylene composites in difference ratios (fiber content: 3, 5, 10 and 20 wt%) by hot-press molding technique [4]. For better compatibility, jute and betel nut fibers were subjected to alkalization and the hybrid composites of alkalized jute/betel nut and PP showed much improved performance with respect to other composites. However, this relies on having reasonable fiber/matrix interfacial strength, and strength can reduce with strongly hydrophobic matrices such as polypropylene with increasing fiber content unless coupling agents or some other interfacial engineering method is used [5].

The mechanical performance of polypropylene (PP) composites filled with betel nut (Areca catechu) short fiber (Bn) at different compositions using extruding and hot press moulding technique was examine by M. Masudul Hassan et al.[6] Results showed that Bn10:PP90 mixture composite (BnPP) had better performance among the composites prepared. Seaweed is subjected to hybridize with betel nut short fiber in PP composite to achieve superior mechanical performance. Bagasse filled recycled polyethylene bio-composites were produced by the compounding and compressive molding method. Two sets of composites were produced using uncarbonized (UBp) and carbonized (CBp) bagasse particles by varying the bagasse particles from 10 to 50 wt%. The bagasse particles added to the RLDPE polymer improved its rigidity and the hardness values of the composites. [7]

There are lots of waste materials available during fabrication process like Polyethylene, Polypropylene, polystyrene etc. These wastes constitute nuisance to the environment not only in India but the world at large. The ability to convert these wastes into useful engineering materials e.g. composites sharpens the focus of this present research work. From the available literature no investigation has been conducted on the application of the leaf sheath of betel-nut plant (Areca Catechu) in polymer composite materials. Fiber modification includes mercerization and Silanation for enhancing better surface interaction towards polymer matrix. Based on the above-mentioned situation, the study described in this work intends to study the mechanical properties of recycled PP composites reinforced with Leaf Sheath Areca Fiber.

2. Materials and Method

2.1. Materials and Equipment

The betel Nut tree which grow abundantly in the eastern & Northern region of India scientifically belongs to 'Areca' species named as "Areca Catechu". The leaf sheath is a basically waste material or used as fuel (fire resource) in rural areas. The Recycled PP used was collected literally from the industrial waste. The chemicals used are Sodium Hydroxide (NaOH), Methyl Alcohol (MeOH), and Acetic Acid (AcOH), Silane coupling agent (Vinyl Tri-methoxysilane) was from MERCK India Ltd. The equipment used in the research were: metal mold, sieves, digital weighing balance, hack saw, grinding machine, Vernier caliper, compounding machine, molding machine and bending and tensile testing machines, Rockwell hardness tester and Charpy impact tester.

2.2. Alkali Treatment (UT)

Generally, the first step is the mercerization process (pretreated process) for all of the fiber surface treatments. Mercerization causes the changes in the crystal structure of cellulose and then the different chemicals can be used on the fibers surface in order to improve the interfacial properties. Chopped fiber (leaf sheath of betel-nut plant) was pre-treated with 4% NaOH for about half an hour in order to activate the -OH groups of the cellulose and lignin in the fiber. Fabric was then washed many times in water and finally dried (Alkaline treated-UT). This treatment removes a certain amount of lignin, wax and oil covering the external surface of the fiber cell wall, depolymerizes cellulose and exposes the short length crystallites [8]. It is reported that alkaline treatment has two effects on the fiber: (1) it increases surface roughness resulting in better mechanical interlocking. (2) It increases the amount of cellulose exposed on the fiber surface, thus increasing the number of possible reaction sites.

2.3. Silane Treatment (TM)

1% of the vinyl- triethoxysilane was mixed with a Methanol/water mixture in the ratio 6:4 mixed well and was allowed to stand for an hour. The pH of the solution was carefully controlled to bring about the complete hydrolysis of the silane by the addition of acetic acid. Fiber was dipped in the above solution and was allowed to remain there for 1 and 1/2 hours. The Methanol/water mixture was drained out and fiber was washed in water. Silane treated fiber (TM) was then dried at room temperature for 2hour, followed by drying in the oven at 70°C for 24 hours [8]. Silane used in this work has two functional groups, a hydrolysable group which can condense with the hydroxyls of the Areca fiber and an organo functional group capable of interacting with the matrix. The hydrolyzed silane can undergo the condensation and bond formation stage when influenced by acid or base-catalyzed mechanisms. Besides these reactions, the silanols can condense to give polysiloxanes. The silanol groups are chemically attached to the fiber through an ether linkage [9].

2.4. Preparation of Bio-composites

At first the fiber from leaf sheath of betel-nut plant (UT& TM) was preconditioned in the hot air oven at the temperature 70°C for 24 hours and then the recycled PP and the fiber was compounded using Twin Screw Extruder. There are five temperature zones in extruder from hopper to die: melting begins from the second zone. The following temperatures were set at respective zones: 180°C, 190°C, 205°C, 210°C, 225°C with the die temperature 210°C The molten strand is then cooled by chilled water and chopped

into pellets using pelletizing machine and then left for cooling. The compounded is then again dried in the hot air oven at temperature 70°C and then for preparing the test specimens for analysis of the mechanical properties and the thermal properties of the composites were injection molded at 190°C, 200°C, 210°C and 220°C temperatures in the barrel zones of injection molding machine.

Table 1. *Different batches of Biocomposites.*

Batch	Recycled PP (%wt)	Fiber (%wt)	Note
0% Fiber	100	0	Non treated
5% UT	95	5	
10% UT	90	10	Only NaOH
15% UT	85	15	treated
20% UT	80	20	
5% TM	95	5	
10% TM	90	10	NaOH and
15% TM	85	15	Silane treated
20% TM	80	20	

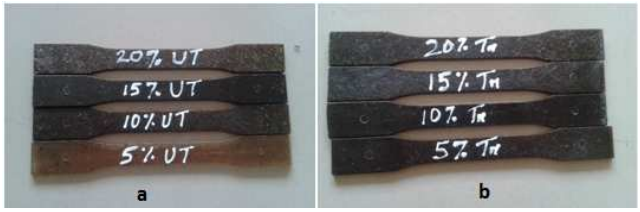

Figure 1. *Photograph of (a) Alkaline treated (UT) & (b) Silane Treated (TM) Biocomposites.*

2.5. FTIR Analysis

The FTIR imaging was performed in Agilent Resolution PRO, Cary 600 series. The generated bio-composite fibers were characterized using Fourier transform infrared spectroscopy (FTIR) to understand the nature of chemical interaction between fiber reinforcement and PP matrix. Changes in absorption peaks were observed in FTIR spectra of Biocomposites fibers as compared to the pure fiber which indicated possible chemical linkages between the fiber and polymer matrix.

2.6. Differential Scanning Calorimetry (DSC)

Differential Scanning Calorimetry (DSC) is a thermo-analytical technique in which the difference in the amount of heat required to increase the temperature of a sample and reference are measured as a function of temperature. Typical applications include determination of melting point temperature, heat of melting, measurement of the glass transition temperature, curing and crystallization studies and identification of phase transformations.

2.7. Mechanical Properties

The tensile test and flexural test were conducted according to ASTM standard testing methods D638 and D790, respectively. The tests were conducted on an INSTRON - Universal Testing Machine (Model-3382).

2.8. Biodegradable Test (Soil Burial Test)

Soil burial test methods have been established and standardized for testing resistance of plastics to micro-organisms. The aim was to access their resistance in soil, rather than the degradability. The test material is buried under laboratory or field conditions. Visual assessment of exhumed materials is carried out and mass loss measurements are also performed. Soil burial tests are used to give an indication of the duration of the test material in a given soil under given conditions. The choice of location can affect the test results.

3. Results and Discussion

3.1. FTIR Analysis

Untreated, alkali (NaOH) and silane treated leaf sheath fiber were investigated by FTIR analysis. Presence of alcohol in untreated fibre results peak at 1030 cm^{-1} which will not be shown due to removal of alcohols –OH group in both alkali & silane treated fiber. The C=O stretching vibration peak which shows presence of pectin is observed in both treated & untreated fibre, but their peaks slightly shifts from 1735 cm^{-1} to 2348 cm^{-1} due to intermolecular H-bonding, which dominates. Peak near to the 1654 cm^{-1} corresponds to–C=O stretching due to the presence of aliphatic carboxylic acid in cellulose chain, & the peak is eliminited due to removal of lignin & pectin which contain ester –C=O group. Surface modification of leaf sheath fiber exhibits the absence of characteristic peaks at 1643 and 1156 cm^{-1}. This is because of the decomposition of hemicellulose and partial leaching out of lignin by sodium hydroxide [2]. The disappearance of the peak at 1643 cm^{-1} is due to the dissolution of a portion of uranic acid, a constituent of hemicelluloses xylan. The shift of the peak from 2900 cm^{-1} to 2698 cm^{-1} indicates participation of some free hydroxyl groups in the chemical reaction. Band at 758 cm^{-1} correspond to silane bride.

The DSC analysis of all samples is carried out using differential scanning calorimetric by heating the sample at 10°C /min from 50°C to 250°C in argon in a thermal analyzer (Perkin Elmer). The plots are shown in the figure 2. The DSC curves in Figure 2 show endothermic melting peaks at 163.78°C for PP. Decreasing amounts of PP gave rise to decreasing melting enthalpies. This was to be expected, since the melting enthalpies are related to the amounts of polymer in the sample, and since the amounts of polymer decrease with increasing fiber content in the sample, the melting peak sizes and related enthalpies should correspondingly decrease. It is, however, possible that the presence of solid fiber may influence the crystallization behaviour of either or both polymers.

3.2. Differential Scanning Calorimetry (DSC)

Table 2. *DSC melting results of Biocomposites.*

Bio-composite	Peak (Tm) °C	Area under melting curve (mJ)	Melting Enthalpy (ΔHm) J/g
5% UT	163.45	301.761	73.600
10% UT	163.43	233.72	64.924
20% UT	162.81	128.578	47.621
5% TM	164.78	273.350	71.934
10% TM	164.44	257.560	59.898
20% TM	164.47	146.705	48.902
0 % PP	163.78	346.738	78.804

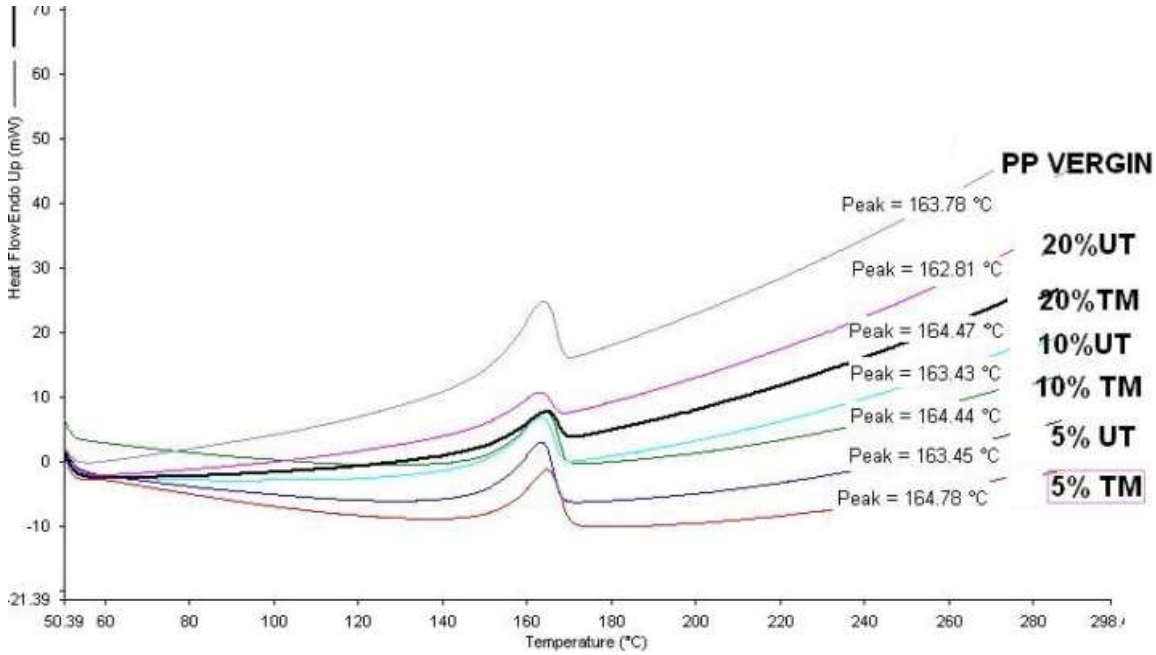

Figure 2. *DSC heating curves of Recycled Polymer (RPP) with fiber Biocomposites.*

3.3. Mechanical Properties

3.3.1. Tensile Test

Tensile properties such as tensile strength of the Leaf sheath fiber / Recycled PP composites containing 5% wt, 10% wt, 15% wt and 20% wt were measured and the results are presented in the Figure 3. It is observed that with an increase in fiber content from 5% wt to 20%, wt the tensile strength gradually decreased for alkaline treated biocomposite. But for 10% wt loading in silane treated biocomposite shows highest tensile strength and then decreases. This may be due to lack of stress transfer from the matrix PP to fiber.

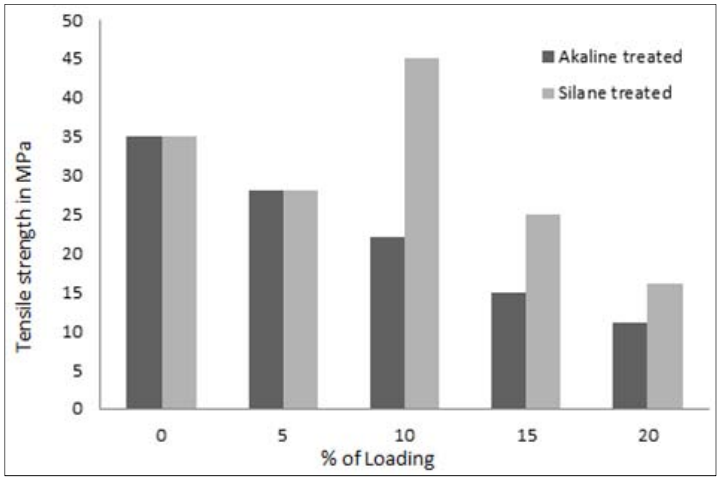

Figure 3. *Tensile strength for alkaline treated and silane treated biocomposites.*

3.3.2. Elongation Test

The elongation at break of the composite shows a similar trend as shown for the 10% wt Leaf Sheath fiber Composite (silane Treated) sample for tensile strength performance and maximum elongation at break. An increase of the elongation at break of the composites increases the toughness and ductility of the composite.[10]

The composite might be present as moderate physical-mechanics adhesion (better known as inter-diffusion that allows a kind of bonding between two polymers surfaces via diffusion of the macromolecules both polymers.

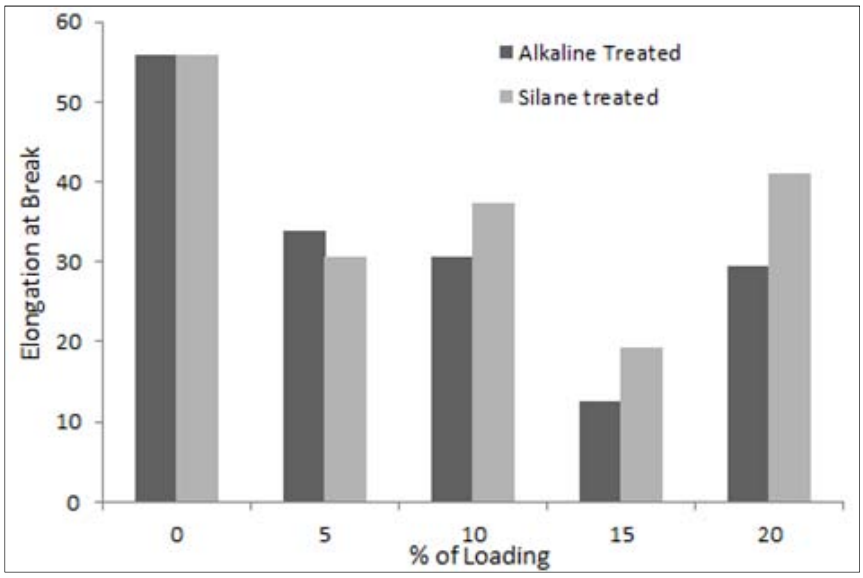

Figure 4. *Elongation at break of alkaline treated and silane treated biocomposite.*

3.3.3. Flexural Strength & Flexural Modulas

When compared between alkaline Treated & silane treated, flexural strength is highest in 10% wt alkaline treated composite. The increase in flexural strength is highest in 10% wt treated composite than other loading. So 15% wt loading for composite shows most efficient result on treatment with silane. Incase of Flexural modulas similar pattern is also observed. The highest flexural modulas increase is observed in 10% wt loading on treatment with silane.

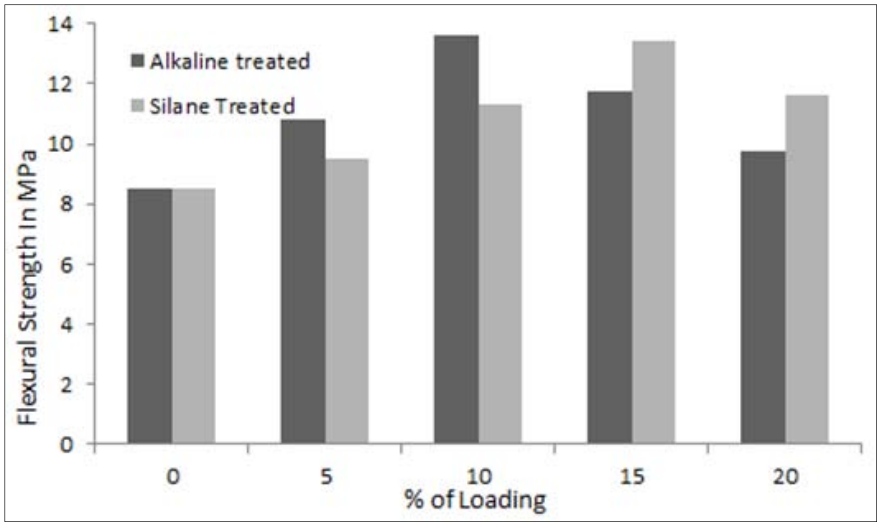

Figure 5. *Flexural Strength of Alkaline treated and Silane treated biocomposites.*

But the Flexural modulas shows a anomolous result which shows highest value in 20% wt loading of alkaline treated biocomposite. It shows a moderate value in both 10% wt & 15% wt treated composite. Recycled polymer shows lower value than composite with loading percentage. The flexural strength & modulas value is least in virgin polymer.

Flexural strength of fibre reinforced composites depend on the nature of matrix material and the distribution and the orientation of the reinforcing fibre, as well as the nature of the fibre- matrix interfaces and the interphase region.

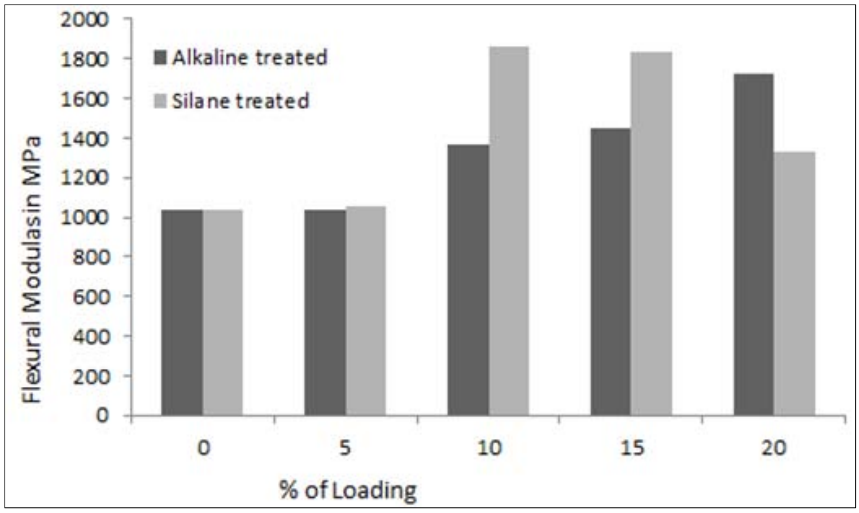

Figure 6. *Flexural Modulas of Alkaline treated and Silane treated biocomposites.*

Even a small change in the physical nature of the fibre for a given matrix result in prominent changes in the overall mechanical properties of composites. It is well known that different degrees of reinforcement effects are achieved by the addition of hydrophilic fibres to different polymers. This may be due to different adhesion strength between matrix and fibres.

3.3.4. Compression Strength

Compression Strength is an important factor to consider where structural stability is required. As one of the objective of this study is to utilize this composite in structural application, so that require measurement of Compression Strength for evaluation.

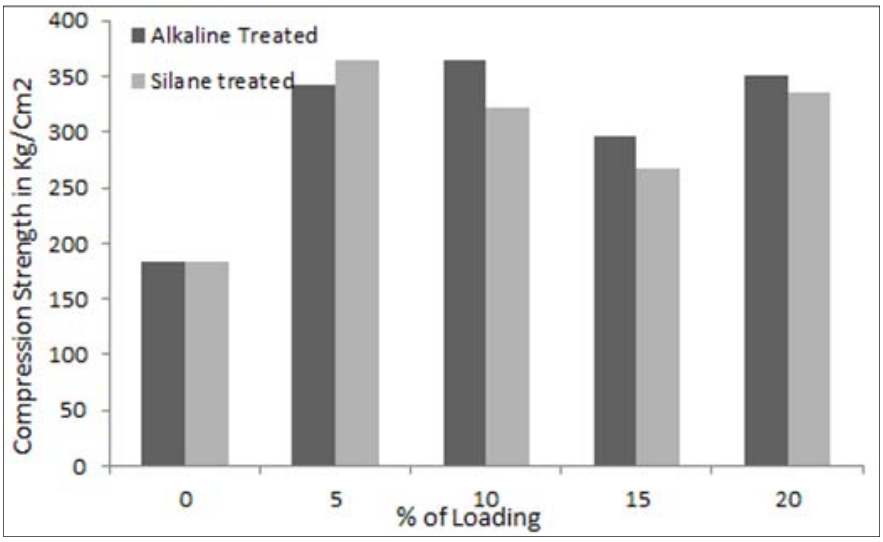

Figure 7. *Compression Strength of Alkaline treated and Silane treated biocomposites.*

As from graph plotted, in alkaline treatedbiocomposite it shows a little decreasing trend in compression strength with increase in loading, until 15% wt, but it suddenly increase in 20% wt loading. This may be due to more better packing of fibre results in better Compression stregth. Increase in fibre content increase the amorphous part & increse free space between polymeric chains, which imparts better chain movement & more dynamic dimension change which increase Compression Strength. The virgin polymer shows least Compression strength than composites.

In silane treated composite 10% wt & 20% wt loading shows better compression strength with moderate test value. This may be due to better interfacial bonding betweem fibres & matrix which imparts better stress transfer through composite.

3.3.5. Rockwell Hardness

Rockwell Hardness is a mesurement of resistance to penetration, indentation, scratching & surface deformation. It is basically measured for surface durability of composite & plastics. As shown in graph it initially marginally increase with fibre loading percent upto 10% wt in both silane treated & alkaline treated biocomposite the gradually decreases with increase in loading percent.

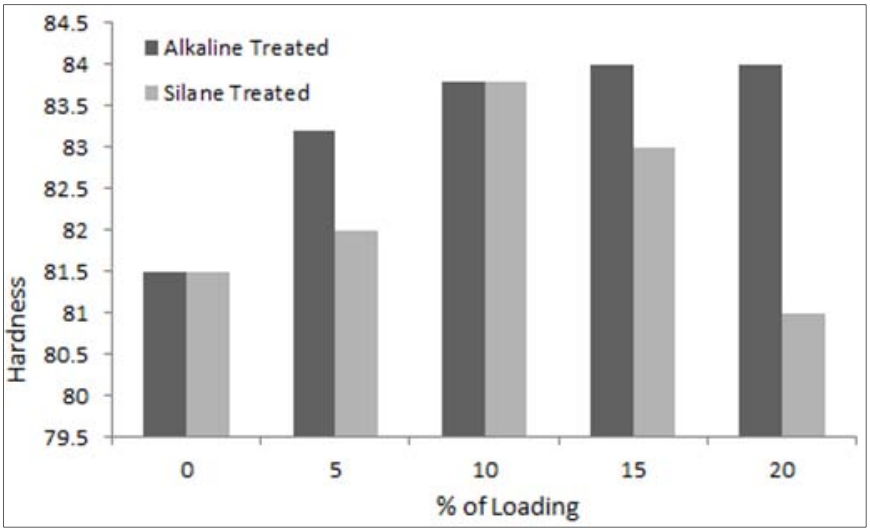

Figure 8. *Hardness of Alkaline treated and Silane treated biocompostes.*

As fiber percentage increase the surface stiffness increases due to decresing percentage of visco-elastic nature plastic, & as Hardness measures the non-recoverable visco-elastic property, so the general trend should be with incresing fiber percent Hardness also increses. Here also this trend is followed in both treated & untreated composite with an exception in 15% in treated composite. This may be due to lack of proper dispersion of fibre in composite matrix, which can impart better visco elastic property decrease in hardness.

3.3.6. Izod Impact Strength

Impact strength is defined as the ability of a material to resist fracture under stress applied at high speed. The impact properties of composite materials are directly related to its overall toughness.

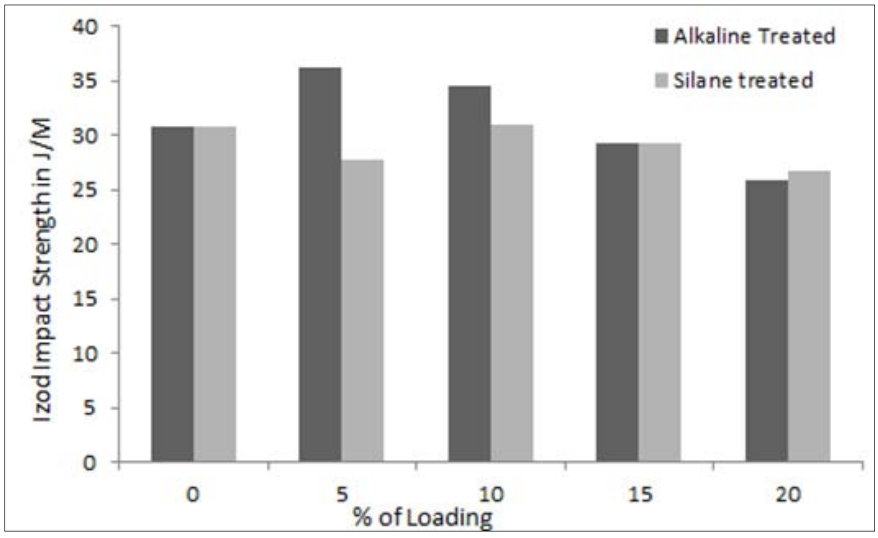

Figure 9. *Izod Impact Strength for alkaline Treated & silane treated biocomposite.*

Figure 9 shows the influence of the alkali treatment, different matrix materials and fiber loading on the Izod impact strength of the areca fiber composites. Figure 9 shows the variation of impact strength of areca composites under Izod impact test. It is seen that with the increase in fiber content in the composite, the energy absorption improves initially but then gradually decreases with increase in loading in case of treated composites.

It is imagined that as the size of the fiber becomes smaller, greater interaction between the fiber and matrix could result in better and more efficient stress transfer which intern could increase the impact strength of the composite [11]. The optimum fiber content varies with the nature of the fiber and matrix, fiber aspect ratio, fiber/matrix interfacial adhesion etc. the low value at high fiber content may be due to the presence of so many fiber ends in the composites, which could cause crack initiation and hence potential composite failure [12]. In view of the above results, it is noted that the composite of 10% wt leaf sheath content exhibits better mechanical behavior.

3.3.7. Soil Burial Test for Biodegradation

Cellulose possesses the tendency to be degraded when buried in soil. For this purpose, the composite samples were weighed individually and buried in soil for 1-4 weeks. Thereafter, samples were carefully withdrawn, washed with distilled water and dried at 105°C for 20 min and kept for 24h and then weight is recorded. Finally weight loss of various degraded samples is calculated. The weight loss indicating that these Biocomposites are partially biodegradable. Soil environment contains different kind of microorganism which leads to weight loss of Biocomposites.

4. Conclusions

Chemical treatment can reduce the hydrophilicity of the fiber to decrease the hydroxyl groups from the fibers and improved coupling between treated leaf sheath fiber and recycled PP which improved interfacial adhesion and so increases the mechanical properties. The tensile strength has reduced for the Biocomposites when compared to recycled polypropylene. In this study the benefits are regarded from an Economic, an Ecological and a Technical point of view for various parts of society. It will be an alternative way to develop the Biocomposites which can be particularly used for daily needs of common people whether it is house hold furniture, house, fencing, decking, flooring, and light weight car components or sports equipment. Waste disposal is becoming increasingly important with the recognition that landfill is not sustainable and as such costs are increasing, with more responsibility being placed on producers. These Biocomposites have low cost, easy availability and aesthetic designs will be the main driving force to transform the depended present to sustainable future. These kinds of Biocomposites are likely to see a period of sustained growth.

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