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## Mechanical, thermal and morphological analysis of EPDM/ polypropylene coconutpith composites

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### ABSTRACT

Increasing concern about global warming and depleting petroleum reserves have made scientists to focus more on the use of natural fibers such as bagasse, coir, sisal, jute etc. This has resulted in creation of more awareness about the use of natural fibers based materials mainly composites. Eco-friendly coconut pith fillers are used as cost effective filler in many of the rubber based composites. The advantages of coconut pith over traditional reinforcing materials such as glass fibers, talc, and mica are acceptable specific strength properties, low cost, low density, non abrasivity, good thermal properties, enhanced energy recovery and biodegradability, and recyclable in nature. Among various natural fibers, both coir and jute fibers are widely available cheap, relatively water – proof and is resistant to damage by salt water. Coconut piths have an outstanding potential as reinforcement in thermoplastics. Commonly used thermoplastics are Polyethylene, Polypropylene, Polystyrene, Nylon etc. These composites are used in automotive components, building materials. EPDM/Polypropylene blends and composites are the most commonly used ones in the thermoplastic elastomers (TPE) in industry. In this current study morphology, thermal behaviour mechanical properties of EPDM/ Polypropylene coconut pith composites have been investigated. Coconut pith in different ratios was added into EPDM / Polypropylene thermoplastics vulcanizates using Brabender Plastic order.

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### KEYWORDS

Coconut pith;  
EPDM;  
Polypropylene;  
Thermoplastic elastomer;  
Composites.

### INTRODUCTION

Many of our technologies require materials with unusual combination of properties that cannot be met by the conventional metal alloys. They are being extensively used in variety of engineering applications in many different fields such as aerospace, oil, and gas and process industries. The need for lighter materials has led to

the increase use of polymer composites. Lightweight and high performance polymeric materials, in many applications, have already replaced steel as major structural materials. Composite materials, are engineered or naturally occurring materials made from two or more constituent materials with significantly different physical or chemical properties which remain separate and distinct within the finished structure. A

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composite material is made by combining two or more materials to give a unique combination of properties, one of which is made up of stiff, long fibers and the other, a binder or 'matrix' which holds the fibers in place. The use of bio-fibers, derived from renewable resources, as reinforcing fibers in both thermoplastic and thermoset matrix composites provide positive environmental benefits with respect to ultimate disposability and raw material utilization. The degrading nature of natural fiber composites under exposure to environmental conditions such as humidity, sunlight, and microorganisms is one of the most important issues in finding suitable outdoor applications. Short fiber reinforced-composites are finding ever increasing applications in engineering and in consumer goods. They can offer a unique combination of properties, and they are more economical than are competing materials. Fiber reinforcement improves stiffness and the strength and, for many polymers it improves toughness, although the toughness may be decreased in polymers that are already tough in unreinforced form. Natural fibers, such as wood, sisal, jute, hemp, and coir, are now established as reinforcing fibers used to produce newly developed composite materials with polymers. Coconut piths are widely used along with Rubber and Thermoset/Thermoplastics blends to make composites. Most of the works were done to utilize the naturally occurring material in the polymer matrix for the cost reduction and property enhancement purposes.

R. Viswanathan and L. Gothandapani studied regarding the particle board made from UF and PF resin using coconut pith as the filler<sup>[1]</sup>. Coconut pith with various particle sizes were employed to make the composite. Better mechanical properties were obtained for PF resins composite than the UF resin ones. From another study from the same author's water absorption characteristics and swelling characteristics of coconut pith particle boards were studied for different particle board thickness and different coconut pith ratios. The influence of fiber treatment on the performance of coir polyester composites were studied by Rout et al. Bleaching and alkali treatment<sup>[2]</sup> were done to the coconut piths. The mechanical properties such as tensile, flexural and impact strength increases as a result of surface modification. The mechanical, thermal, morphological, and water absorption properties of recycled milk pouch

polymer/coconut pith composites with different treated and untreated fiber contents were evaluated and compared with those of virgin LDPE-LLDPE/coconut pith composites by Choudhury et al. Recycled milk pouch coconut pith composites have given inferior mechanical properties than virgin LDPE-LLDPE composites<sup>[3]</sup>. Addition of small amount of coupling agent such as Styrene Maleic Anhydride has improved the mechanical and thermo oxidative properties. C. Y. Lai, S. M. Sapuan studied the mechanical and electrical properties of coconut pith reinforced Polypropylene composites<sup>[4]</sup>. Mechanical and electrical properties increased by treating the coconut piths. The treatment agents used included alkali, stearic acid, acetone, and potassium permanganate. Monteiro et al. studies on coconut pith polyester composites specially the mechanical properties<sup>[5]</sup>. Composites prepared with two molding pressures and with amounts of coconut pith up to 80 wt% were fabricated. Up to 50 wt% of fiber, rigid composites were obtained. For amounts of fiber higher than 50%, the composites performed like more flexible agglomerates. Studies on the coir epoxy composites were carried out by Harish et al and found out that the properties are lesser than GFRP laminate specimens<sup>[6]</sup>. The major problem identified with natural fibers during incorporation in hydrophobic polymers is their poor compatibility. To alleviate this problem, various fiber-polymer interface modifications have been proposed which results in improvement of performance of the resulting composites, reports on the physico mechanical properties of chemically treated palm and coconut pith reinforced polypropylene composites justified that chemically treated palm/coir composites yielded improved mechanical properties<sup>[7]</sup> compared to the raw (non-treated) ones, while coconut pith composites yielded better mechanical properties compared to the palm ones. Modification of the coconut piths using bromine and stannous chloride solution to improve the fire resistant properties<sup>[8]</sup>. After improving flame resistance properties they made coconut pith epoxy composites and found out that the fire retardant properties has been significantly improved, also studies on chemical modification and its effect on the mechanical properties of HIPS/coconut fiber composites<sup>[9]</sup>. They had mixed alkali treated and bleached coconut piths with high impact polystyrene. Tensile test result shows that addition

of 30% alkali treated and bleached fibers reinforcing the HIPS matrix provided considerable changes in the mechanical properties in comparison with pure HIPS. Anuar et.al investigated that blend of polypropylene (PP) and ethylene propylene- diene monomer (EPDM) at a ratio of 70:30<sup>[10]</sup>. The optimum fiber loading were investigated from 0% to 20% by volume. The effect of coupling agent maleic anhydride polypropylene (MAPP) on the TPE composite has been investigated. The result show that, with increasing the kenaf fiber content gradually increased the tensile strength and flexural strength for both treated and untreated PP/EPDM-KF composite, from the scanning electron micrograph (SEM) it has shown that the composite, with compatibilizer promotes better interaction between TPE and kenaf fiber, he also studied the effect of both a thermoplastic elastomer (EPDM) and short aramid fibers on polypropylene (PP) crystallization kinetics and tensile behavior<sup>[11]</sup>. The results have shown that PP/EPDM blends are non-miscible in the melt and, at low EPDM percentages in the blend (25%), the particles of EPDM act as nucleating agents and co-crystallize with PP. Aramid fibers are effective nucleating agents for PP crystallization, giving rise to the phenomenon of PP transcrystallinity on their surface. In order to improve the compatibility between natural fibers and polypropylene (PP) and ethylene propylene diene terpolymer (PP-EPDM) blends, the functionalization of both matrices with maleic anhydride (MA)<sup>[12]</sup>. The morphological observations carried out by scanning electron microscopy show that the incorporation of small amounts of functionalized polymer considerably improves the adhesion at the fiber-matrix interface, the mechanical properties and morphology of ternary composites based on PP/EPDM blends reinforced with natural flax fibers showed improvement in tensile, flexural and impact behaviors of the composites<sup>[14]</sup> and hence we can conclude that flax fibers behave as an effective reinforcing agent in these systems, the grass fiber Polypropylene/ Natural rubber/ EPDM composites. Adding NR or EPDM to PP composites, a significant increase in the impact strength and elongation at break is observed in the PP composite with rubber content more than 20% by weight. However, the tensile strength and Young's modulus of the PP composites decrease with increasing rubber contents. Sabu Thomas and coworkers stud-

ied on the short coconut pith reinforced natural rubber composites. The surface morphology of coconut pith is modified by sodium hydroxide treatment<sup>[15]</sup>. From the mechanical properties, it was observed that coconut piths should be immersed in 5% sodium hydroxide solution for 48 h for maximum tensile properties. Anisotropic swelling studies indicated poor adhesion between untreated coconut pith and natural rubber. The swelling is smaller in composites containing alkali- treated coconut pith along with the resorcinol hexamethylenetetramine bonding agent. A number of small value added products can be made using coconut pith and coconut pith to the polymer matrix. Even though thermosetting plastics seems to be the best suited for composite preparation with coconut pith and fiber, rubber and other thermoplastic materials finds better in compatibility and properties. The coconut pith can also be used to replace the conventional fillers, so that cost reduction can be of prime concern. Many of the works has been carried out in thermoplastics and rubber/thermoplastics blends. In EPDM/PP blends most of the work has been carried out using fibers such as flax, jute, aramid, kenaf etc.

## EXPERIMENTAL

### Materials

The polypropylene used in this study was B120MA grade made by M/S Reliance Industries Limited, Mumbai. EPDM – KELTON 512 was obtained from DSM Elastomers, Netherlands Paraffinic oil was procured from Neelam Lubricants, Mumbai. Coconut pith was obtained from Rubber Park, Ernakulum. Dicumyl Peroxide 40 (DCP 40) for dynamic vulcanization was obtained from BP chemicals, Mumbai. The formulations of EPDM/Polypropylene/Coconut pith composites used in this study is shown in TABLE 1

TABLE 1 : Formulation of the ternary composites batches

INGREDIENTS (phr)	C0	C1	C2	C3
EPDM (KELTAN 512)	60	60	60	60
POLYPROPYLENE (B 120 MA)	40	40	40	40
COCONUT PITH	0	10	20	30
PARAFFINIC OIL	10	10	10	10
DCP - 40	2	2	2	2

(phr: Parts per hundred grams of polymer)

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### Preparation of ternary biocomposites

Polypropylene was incorporated with coconut pith in a laboratory mixer at a temperature just near to the softening temperature that is 150°C, maintaining the rotor speed and fill factor i.e. 50rpm. Polypropylene/coconut pith master batch was then melt blended with EPDM at 180°C, at a rotor speed of 50 rpm for a total time of 12 min., DCP 40 added after 8 min. for dynamic vulcanization to take place, After melt mixing, the molten mass was taken out from the laboratory mixer and while hot, passed through two-roll mill mixing mill to chill it and sheet it to about 2 mm thick. The sheet was then cut and press molded for 7 min. in a compression molding hydraulic press at 180°C, under specified pressure of kg. Wax film was placed between the sheet and the press plates to avoid adhesiveness. The sheet was then cooled down to room temperature still under pressure. The test specimens were die-cut as per ASTM D 638 from the compression molded sheet and used for measuring mechanical properties after 24 hours of conditioning at room temperature.

## RESULTS AND DISCUSSIONS

### Mechanical properties

The measurements of tensile properties, tensile strength, elongation at break, tensile modulus as ASTM D 638 and tear strength ASTM D 624, of samples were carried out by an Instron 1185 tester according to standards, at a crosshead speed of 50 mm/min. Hardness testing was carried out using Shore A durometer according to ASTM D 2240. The impact strength of the samples were carried out using Izod impact strength tester according to ASTM D 256 standards. The tensile strength, modulus, hardness, tear strength and elongation at break of ternary biocomposites with polymer blend ratio of EPDM/Polypropylene in the ratio of 60:40 with change in the concentration of coconut pith – 0, 10, 20, 30 phr are depicted in the below TABLE 2,

The tensile strength values decreases slightly up to 20 phr of coconut pith loading. This is because after the addition of coconut pith into the EPDM / Polypropylene matrix the stiffness of the compound increases. Tensile modulus increases as the coconut pith concentration increases. This is due to the increase in rigidity

TABLE 2 : Mechanical properties of ternary biocomposites

Sample Code	C0	C1	C2	C3
Tensile Strength (Mpa)	10.5	9	7.8	6.5
Elongation at Break (%)	480	350	300	275
100% modulus (Mpa)	200	240	260	245
Impact strength (kJ/mm <sup>2</sup> )	5.5	9	12	10.5
Tear Strength	10.5	11	12	14
Hardness (Shore A)	83	83.5	84.5	86

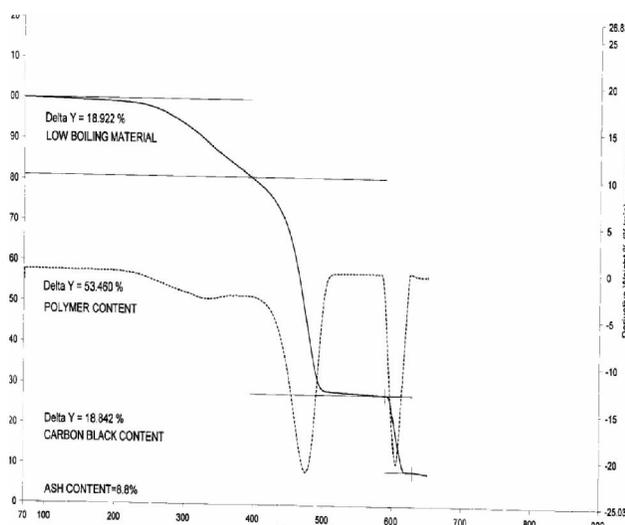
of the material after the addition of coconut pith. The increased concentration of coconut pith filler increases the impact strength up to 20 phr. The decrease in properties is due to the less interaction of the filler into the matrix. Tear strength increases as the coconut pith content increases it is mainly due to the large surface area of the coconut filler which improves the interaction with the EPDM / Polypropylene matrix. Elongation at break decreases as the coconut pith content increases in composites is due to the incompatibility between the polymer and filler. Moreover, the deformation of the filler is generally much lesser than that of the polymer matrix, thus, the filler forces the matrix to deform more than the overall deformation of the composite. Hardness increases as the coconut pith content increases.

### Thermal analysis

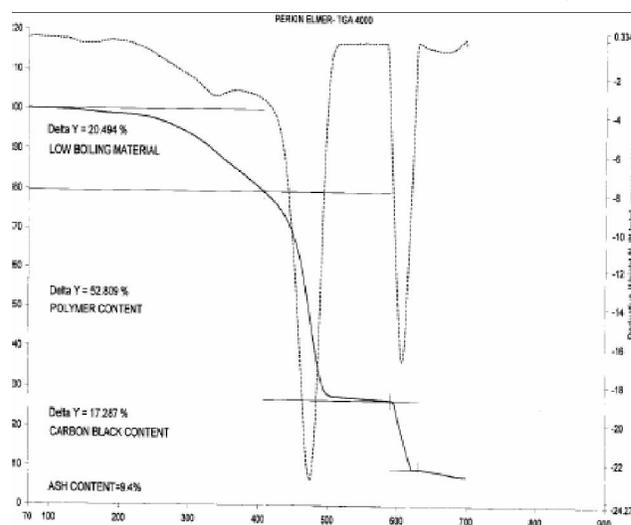
In order to investigate the influence of coconut pith on thermal stability of EPDM / Polypropylene composites, TGA study was carried out. TGA was carried out in PERKIN ELMER-TGA 400 with a heating rate of 20°C / minute Figure 1 a,b,c,d shows the TGA thermographs of EPDM / Polypropylene / coconut pith composites with variable dosage of coconut pith.

Thermographs reveal that the onset of degradation of the composites shifts towards a higher temperature on increasing coconut pith concentration in the blend indicating higher thermal stability. The increase in thermal stability is due to the addition of coconut pith because coconut pith itself is having higher thermal stability. So it imparts thermal stability to the EPDM / PP coconut pith composites. From figure 2 we can conclude that as the concentration of coir pith increases thermal degradation temperature increases. After the addition of 0 to 30 phr coir pith the degradation temperature has increased from 270°C to 314°C.

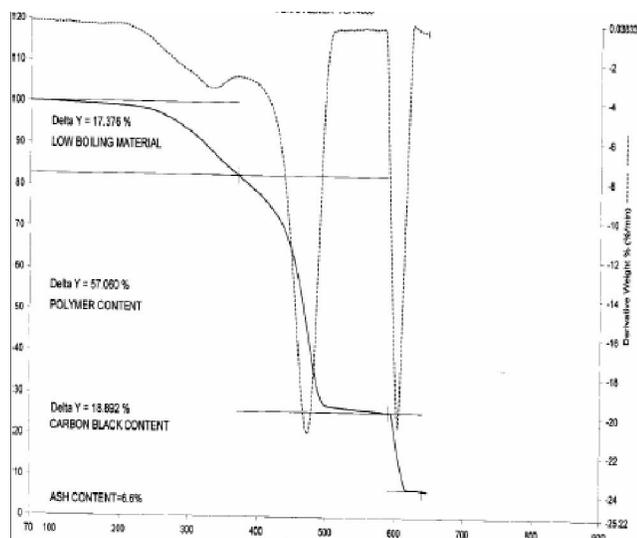
### Scanning electron microscope studies



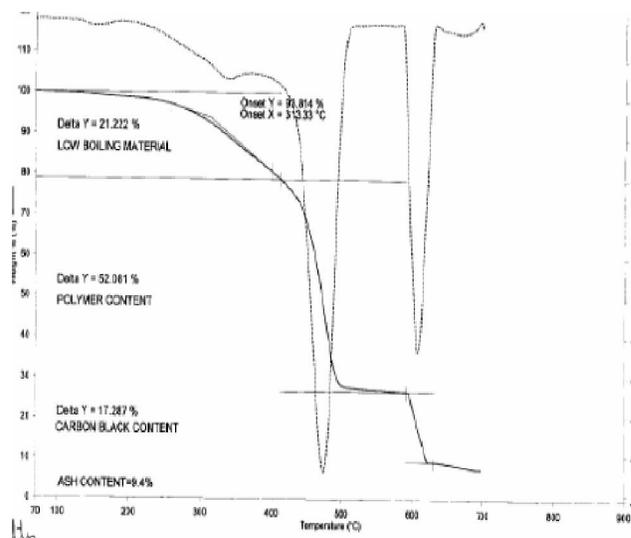
(a) 0 phr coconut pith (270.5°C)



(b) 10 phr coconut pith (282.31°C)



(c) 20 phr coconut pith (300.12°C)



(d) 30 phr coconut pith (314.36°C)

Figure 1 : Thermographs of 0, 10, 20, 30 phr of coconut pith in EPDM / PP composites

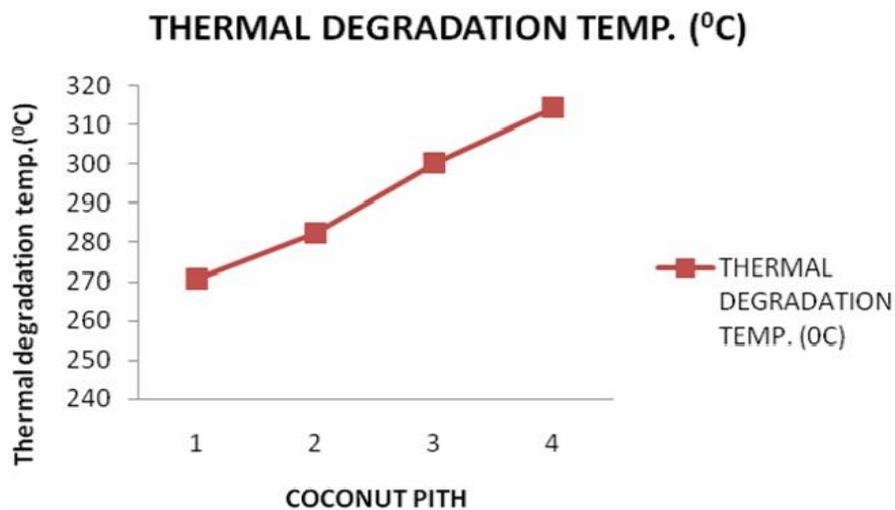
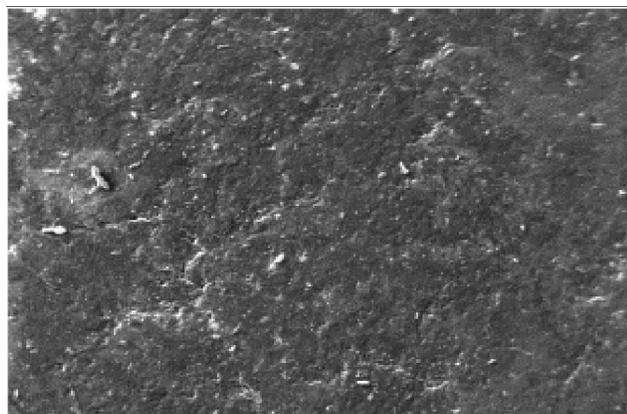
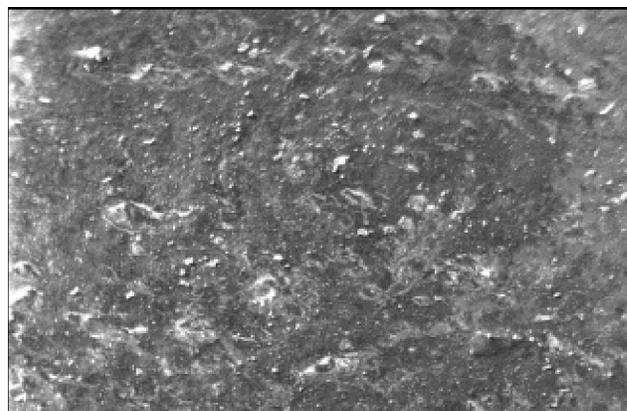


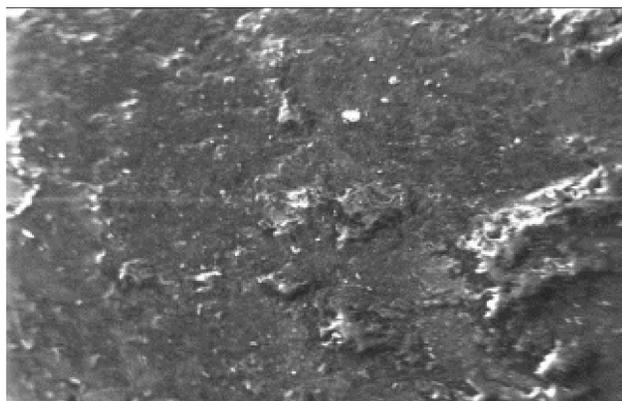
Figure 2: Effect of coconut pith degradation temperature



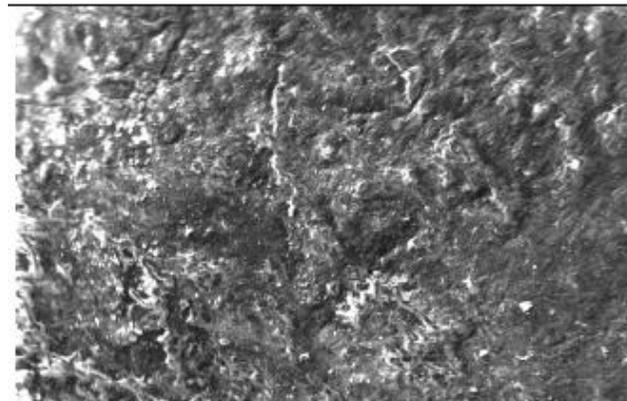
(a) 0 phr coconut pith



(b) 10 phr coconut pith



(c) 20 phr coconut pith



(d) 30 phr coconut pith

Figure 3 : SEM of 0, 10, 20, 30 phr of coconut pith in EPDM / PP composites

Morphological analysis was carried out in order to study the matrix after the addition of coconut pith and the dispersion of coconut pith into the matrix. Phase morphology of the various blends was investigated by a Digital Scanning Electron Microscope (SEM). The images were obtained at a tilt angle of  $0^\circ$  with an operating voltage of 20 kV.

We can see from the figure that the rubber particles are dispersed throughout the polypropylene matrix in the form of aggregates and the size of rubber particles is less than  $2 \mu\text{m}$  in the unfilled thermoplastic elastomer sample. It should be noted that after addition of coconut pith the size of rubber particles increases and the voids increases. Initially the coconut pith cannot penetrate into the rubber phase but after adding curing agent DCP- 40, the rubber phase become more polar. Therefore, it is possible that some coconut pith goes to EPDM phase before the curing cycle ends. At 30 phr concentration the dispersion of coconut pith is difficult, as 30 phr coconut pith acts as diluents hence mechanical prop-

erties decreased.

## CONCLUSIONS

The study was conducted to determine the properties of EPDM / Polypropylene coconut pith composites. The effect of coconut pith loading on 60:40 (EPDM/PP) was determined and observed that hardness, tensile modulus, impact strength values increases up to 20 phr dosage. At 30 phr loading of coconut pith the properties decreases due to the dilution effect. Tensile strength and elongation at break values decreases because the addition of coconut pith increases the material stiffness and it decreases the elongation. Maximum Thermal degradation temperature increases with increase in dosage of coconut pith because of its high thermal stability. Morphological analysis shows that rubber particles are dispersed throughout the polypropylene matrix and at 30phr coconut pith concentration the dispersion of coconut pith is difficult due to polarity dif-

ference.

## REFERENCES

- [1] R.Viswanathan, L.Gothandapani; Mechanical properties of coconut pith particle board, *Bioresource Technology*, **67**, 93-95 (1985).
- [2] J.Rout, M.Misra; The influence of fiber treatment on the performance of coir-polyester composites, *Composite Science and Technology*, **61**, 1303-1310 (2001).
- [3] Arup Choudhury, Sandeep Kumar; Recycled Milk Pouch and Virgin Low-Density Polyethylene/Linear Low-Density Polyethylene Based Coconut pith Composites, *Journal of Applied Polymer Science*, 775-785 (2006).
- [4] C.Y.Laia, S.M.Sapuan; Mechanical and Electrical Properties of Coconut pith-Reinforced Polypropylene Composites, *Polymer-Plastics Technology and Engineering*, **44(4)**, 619-632 (2005).
- [5] S.N.Monteiro, L.A.H.Terrones; Mechanical performance of coconut pith/polyester composites, *Polymer Testing*, **27**, 591-595 (2008).
- [6] S.Harish, D.Peter Michael; Mechanical property evaluation of natural fiber coir composite, *Material Characterization*, **60**, 44-49 (2009).
- [7] Md.Mominul Haque, Mahbub Hasan; Physico-mechanical properties of chemically treated palm and coconut pith reinforced polypropylene composites, *Bioresource Technology*, **100**, 4903-4906 (2009).
- [8] Akhil Kumar Sen, Sandeep Kumar; Coir-fiber-based fire retardant nano filler for epoxy composites, **101**, 265-271 (2010).
- [9] Kelly C.C.karvalho; Chemical modification effect on the mechanical properties of HIPS /coconut fiber composites. *Bioresources*, **5(2)**, 1143-1155 (2010).
- [10] H.Anuar, N.A.Hassan; Compatibilized PP/EPDM-Kenaf Fiber Composite using Melt Blending Method, *Advanced Materials Research Vols*, (2011), 743-747 (2011).
- [11] M.Arroyo, R.Zitzumbo; Composites based on PP/EPDM blends and aramid short fibers. Morphology/behaviour relationship, *Polymer*, **41**, 6351-6359 (2000).
- [12] M.A.Lo'pez Manchado, M.Arroyo; Enhancement of Mechanical Properties and Interfacial Adhesion of PP/EPDM/Flax Fiber Composites Using Maleic Anhydride as a Compatibilizer, *Journal of Applied Polymer Science*, **90**, 2170-2178 (2003).
- [13] J.Biagotti; Ternary Composites Based on PP-EPDM Blends Reinforced with flax fibers. Part 2 : Mechanical Properties /Morphology Relationship, *Polymer Engineering and Science*, **43**, 1031-1043 (2003).
- [14] Yupaporn Ruksakulpiwat; Improvement of impact property of natural fiber polypropylene composite by using natural rubber and EPDM rubber, *Composites: Part B*, **40**, 619-622 (2009).
- [15] Sabu Thomas; Short Coconut pith –Reinforced Natural Rubber Composites: Effects of Fiber Length, Orientation and Alkali Treatment, *Journal of Applied polymer Science*, **55**, 583-594 (1994).