Mechanical Properties of Natural Rubber Composites Reinforced With Lignin From Caryota Fibre

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Abstract— Carbon, silica etc are added to natural rubber as filler to improve the mechanical properties. In the present study, a natural filler lignin was incorporated into natural rubber which was coagulated by microorganism rather than conventional method of using acid and was tested for their mechanical and wear behavior. The advantages of natural fillers are their low density, renewable character and biodegradability. Lignin was extracted from a cheap source, the Caryota fibre. Lignin reinforced natural rubber composite was made by co-precipitating rubber latex with various loadings of lignin. The tensile modulus, tear strength, abrasion resistance and hardness of the composite were found to be improved.

Keywords — Mechanical properties; natural rubber; lignin; Caryota fibre; composite.

I. INTRODUCTION

This Lignin is the most abundant renewable biomass resource next to cellulose. In polymer industry lignin has got great applications. Lignin based rigid polyurethane is excellent in flame retardance. Sulfur free lignin has been used for automotive brakes and epoxy resins for printed circuit boards. Polyphenylene oxide- based polymers and lignin esters blends exhibit good modulus of elasticity, tensile strength, and elongation at break values that are comparable or greater than the polyphenylene oxide- based polymer alone. Lignin can act as a water absorption inhibitor and as fluidization agents when used with polyamide when mixed in solid or melt form and processed by injection molding, blow molding, extrusion, or blow extrusion to fabricate articles. The use of alkali lignin polypropylene carbonate improves thermal stability and mechanical properties. The pulp and paper industries produce very large quantities of lignin , but the cost of extraction of lignin is very high.

So far, only limited studies are conducted on the application of lignin as a filler in rubber vulcanizates. Kumaran et al. [1] used lignin as a replacement of carbon black in styrene - butadiene and natural rubbers. Nichols [2] employed kraft lignin as a reinforcing agent in elastomeric composition based on polyethers, polyester amide and poly alkaline glycols. Physico-mechanical be improved for the final product properties were elastomeric reported to by using oxidised kraft lignin instead of lignin in the unmodified form. Haxs and Mills [3] have reported the possibility of development of lignin-reinforced rubber vulcanizates by co-precipitating lignin with rubber latex and subsequent treatment with an organic poly-isocyanate. Various lignin preparations as fillers in rubber vulcanizates have also been compiled by ACS Rubber Division library [4].

Lignin is incorporated in rubber compounding by the addition of lignin in the form of rubber latex co-precipitate [5], or by the direct incorporation of lignin into dry rubber. It was reported by Sagajilo[6], where the incorporation of lignin into rubber by means of conventional mill mixing techniques did not produce reinforced rubber. On the other hand, reinforcement is observed when the lignin-rubber mixture was obtained via latex co-precipitation or coagulating the lignin-latex mixture so as to form rubber master-batch. The main reason for using lignin-latex co-precipitate is because of the occurrence of lignin binding or aggregation during dry rubber mixing. The binding of lignin particles to one another by hydrogen bonds leads to agglomeration preventing the dispersion of lignin particles in rubber during dry rubber mixing [7].

In the application of lignin as filler, one of the main problems is the cost of lignin extraction. In the study presented here lignin is extracted from a cheap source, fibres in the sheathing leaf base of aryota urens which was dumped in the elephant kraals. The black coloured fibres are rich in lignin. Chemical analysis of the fibre has revealed that 26% lignin is present in the fibre. Lignin is extracted from Caryota leaf fibre by simple treatment with alkali.
II. MATERIALS AND METHODS

Extraction of Lignin from Caryota leaf sheath fibre

Caryota leaf sheath fibre was collected from Elephant Kraal at Kodanadu, Kerala, India. Caryota leaves are given as feed to Elephants. The sheathing leaf bases are the main wastes in the Kraals. The fibres were removed from the sheathing leaf bases, then it was thoroughly washed with Teepol and cut into small pieces and used for lignin extraction. The fibres were treated with 1% NaOH and heated for 10 minutes at 121° C for 15 minutes in an autoclave. The dark liquid portion was removed and then neutralised by adding formic acid and dried into a powder by vacuum evaporation in rotary evaporator.

Analysis of lignin fibre

Chemical composition of Caryota fibre was estimated according to ASTM procedures. Lignin-ASTM D 1106, Cellulose- ASTM D 1104, Ash content- ASTM D 1102.

Scanning Electron Microscopy

The SEM photograph of fibre was taken using JEOL 35C model scanning electron microscope.

FTIR analysis

Infrared spectra of Caryota fibre was obtained with Schimadzu model IR 470 infrared spectrophotometer, using solid KBr pellet technique. Fibre samples were cut into small pieces and grounded well before mixing it with KBr.

Preparation of lignin reinforced natural rubber composite

In order to prepare lignin filled rubber sheet, lignin was added directly into commercial grade natural rubber latex to produce varying loadings of lignin 0,5,10,15,20,25,30,35 parts per hundred rubber (phr). It was diluted with distilled water to produce dry rubber content of 30%. Then it was coagulated by the innovative method of coagulation by addition of yeast (published elsewhere) and dewatered by pressing in a roller and dried.

Preparation of test specimen was done on a laboratory two roll mixing mill with size 150X 300mm as per ASTM D3184-80 at a friction ratio of 1:1.25. The base formulation of the rubber compound is given in table.1. The rubber was first masticated by careful control of temperature, nip gap, time and uniform cutting operation. The nip gap, mill roll speed ratio and number of passes were kept the same in all mixes. The mixes were sheeted out as thin sheets and were kept for maturation for 24 hrs. The optimum cure time at 150°C was ascertained using a Rubber Process Analyzer.

In an electrically heated hydraulic press having 30x30cm platens at a pressure of 200kg/cm² the compounds were compression moulded. The rubber compounds were vulcanized to their respective cure times

<table>
<thead>
<tr>
<th>Table-1 Basic formulation of rubber compound ascertained</th>
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<tbody>
<tr>
<td>Ingredients</td>
</tr>
<tr>
<td>--------------------------------------------------------</td>
</tr>
<tr>
<td>Natural rubber</td>
</tr>
<tr>
<td>ZnO</td>
</tr>
<tr>
<td>Stearic acid</td>
</tr>
<tr>
<td>TDQ</td>
</tr>
<tr>
<td>CBS</td>
</tr>
<tr>
<td>TMD</td>
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<td>Sulphur</td>
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III. MECHANICAL TESTING

Stress strain measurements were carried out on a Schimadzu Model AGI universal testing machine having cross head speed of 500mm/min. Tensile modulus and elongation at break were measured according to ASTM D 412-87(method A). Dum bell shaped specimens were punched off from moulded sheet and tests were conducted.

Tear resistance was calculated as per ASTM D 624-86 using crescent shaped specimens. Cross head speed was maintained at 500 mm/min. It is represented in kN/m.

Abrasion resistance test was conducted on DIN 53516 abrader. Cylindrical samples having diameter 15mm and length 20mm was kept on rotating sample holder and was rubbed against rotating abrasive coated cylinder. The abrasion loss is expressed as loss in volume.

Hardness of composite was measured using Shore A type Durometer as per ASTM 2240-81. The instrument uses a calibrated spring for to provide indenting force.
IV. RESULTS AND DISCUSSION

Table -2 shows the chemical components present in the fibre. Presence of 26.3 % lignin in the fibre makes it a suitable source for extraction.

<table>
<thead>
<tr>
<th>Chemical constituents</th>
<th>%</th>
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<tbody>
<tr>
<td>Cellulose</td>
<td>34.92</td>
</tr>
<tr>
<td>Hemicellulose</td>
<td>28.73</td>
</tr>
<tr>
<td>Lignin</td>
<td>26.3</td>
</tr>
<tr>
<td>Ash</td>
<td>2.83</td>
</tr>
<tr>
<td>Others</td>
<td>7.22</td>
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The IR spectrum of Caryota fibre shows an absorption peak at 1730 cm\(^{-1}\) which is the characteristic band for carbonyl stretching associated with the carbonyl groups present in lignin. A band at 1600 cm\(^{-1}\) is due to the C-C stretching of the aromatic ring in the lignin components. The bands at 1374, 1314 cm\(^{-1}\) are due to the -CH deformation, -OH in plane bending and -CH\(_2\) wagging respectively.

Scanning electron microscopy is a very useful tool to gather information about topography, morphology, composition and micro structural information of materials. Scanning electron microscopic images of the fibre is shown the Figure 2 & 3.

From Fig.4 it can be observed that as filler loading increases, tensile modulus increases and reaches a maximum value at 25 phr and then decreases. Fig. 5 indicates that as filler loading increases, hardness of rubber composite also increases. Elongation at break decreases as the filler loading increases, which is shown in Fig.6.
V. CONCLUSION

The process of extraction of lignin from Caryota fibre is quite simple and as a composite with natural rubber it shows improvement of main mechanical properties. A filler loading of 20 to 25 phr (parts per hundred rubbers) was found to give optimum results. Since natural fillers are eco friendly it can be a good substitute for fillers like Carbon black.

REFERENCES