SYNTHESIS, CHARACTERIZATION AND EVALUATION OF PHYSICAL PROPERTIES OF BIODEGRADABLE COMPOSITES FROM LENS CULINARIS

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ABSTRACT
This research paper deals with the synthesis and characterization of Lens culinaris based composites using resorcinol-formaldehyde as crosslinker. Acid, base and moisture resistance studies of the composites were also done. Moreover, biodegradation studies of the composites were also done using composting method and the different stages of the biodegradation were evaluated using scanning electron microscopy.

KEYWORDS: Lens culinaris, composites, biodegradation, acid-base resistant studies, SEM

INTRODUCTION
Ecological concerns have resulted in a renewed interest in natural and compostable materials, and therefore issues such as biodegradability and environmental safety are becoming important. Biodegradable polymers are designed to degrade upon disposal by the action of microorganisms. Since last decade, many attempts have been focused on grafting or blending of plastic materials with cheap and biodegradable natural biopolymers, such as starch, cellulose, chitin and psyllium to create new materials with desired properties. Developments of commercially viable biodegradable composites based on natural sources for a wide range of applications are being developed day by day. Composite is a material having two or more chemically distinct phases, which at the microscopic scale are separated by a distinct interface. From the structural point of view, composites are anisotropic in nature. Mechanical properties of composites are different in different directions and composite is a commodity having superior properties than the individual constituent. Some of the basic properties of composite such as light weight, high strength to weight ratio and stiffness make them a suitable product for the replacement of conventional materials like wood.

Composite materials are playing an important role in various industrial sectors. These materials generally lack in mechanical strength, which can be enhanced using synthetic or natural fibers as a reinforcing material. On the other hand, agro based materials can also be used as filler in composite materials to reduce the final cost and improve their environmental degradability. Most of the living tissues like bone, collagen, dentine and cartilage are essentially composites. Bio-composites (biodegradable composites) consist of bio-degradable polymers as the matrix material and biodegradable fillers, usually bio-fibers (e.g. lignocelluloses fibers). Since both these components are biodegradable, hence the composite as the integral part is also expected to be biodegradable. Composite materials have gone through significant developments in terms of use of different raw materials, processes and even applications. Petricca et al synthesized the hydroxyapatite-biodegradable polymer composites by a colloidal non-aqueous chemical precipitation technique at room temperature. Singha et al synthesized and characterized short Saccharum cilliare fiber reinforced polymer composites. Huang et al reported fully environment-friendly, sustainable and biodegradable ‘green’ composites fabricated using modified soy protein concentrate (SPC) resins, flax yarns and fabrics. Bledzki et al prepared the composites reinforced with cellulose based fibers. Gaspar et al reported the mechanical strength, water absorption and enzymatic degradation of
composite by the addition of natural polymer cellulose, hemi-cellulose, zein (protein) and polycaprolactone. The literature review has revealed that a very little work has been done on finding the application of this fiber as reinforcing material in the polymer composites.

In this research paper, biodegradable composites of *Lens culinaris* using resorcinol-formaldehyde as crosslinker were synthesized. Acid, base and moisture resistance studies of the composites were also done. Moreover, biodegradation studies of the composites were also done using composting method and different stages of the biodegradation were evaluated using scanning electron microscopy.

**MATERIALS AND METHODS**

**Materials:** Petroleum ether (60-80°C) (Merck), methanol (Merck), resorcinol (Merck), formaldehyde (Merck), sodium hydroxide (S. D. Fine), HCl (S. D. Fine) were used as received. *Lens culinaris* were procured from the local market. Weighing of the sample was done with electronic weighing machine (Afcoset). Drying was carried-out in Hot Air Oven (Jain Scientific Works, Ambala). SEMs of the samples were taken on LEO435VF (Electron microscopy). Hot pressing of the samples was carried-out with Carver Hydraulic Hot Press under 178 KN force.

**Biodegradation studies:** Biodegradation studies were carried-out by using composting method for 65 days. Weights of the test samples were taken at regular time interval (after every 7 days) and the confirmation of biodegradation was carried-out by SEM studies.

**Acid resistant studies:** Acid resistant properties of samples were studied by using 5N HCl for 72 hours. Weights of the samples were taken at regular time interval of 6 hours.

**Base resistant studies:** Base resistant properties of samples were studied by using 5N NaOH for 72 hours. Weights of the samples were taken at regular time interval of 6 hours.

**Water uptake resistant studies:** Water uptake resistance study of the samples was carried out by putting a definite amount of each sample in a definite volume of distilled water. The weight of each sample was taken after every 6 hours.

**Synthesis**

**Preparation of biodegradable matrix:** *Lens culinaris* were converted into a fine powder (400 gm) and the powdered material was kept for cold percolation in petroleum ether (60-70°C) for about 72 hours. After removal of the solvent, marc left was dried in oven at 40°C and again cold percolation was followed with 70% methanol for about 72 hours. Solvent was removed with filtration and the marc so left was washed repeatedly by distilled water till the impurities left were completely removed. The material was dried in oven at 40°C and the final weight was taken.

The percentage of the purified powder obtained was calculated as:

\[ \text{% Matrix powder} = \frac{F_w}{I_w} \times 100 \]

Where \( I_w \) = initial weight of the material taken; \( F_w \) = final weight of the material obtained

**Synthesis of crosslinker:** Resorcinol-Formaldehyde was prepared as per the method described earlier. Thick slurry of purified powdered material was prepared with distilled water in a beaker and a definite amount of resorcinol-formaldehyde was added to it. The mixture was stirred thoroughly so as to obtain a homogenous mixture. The reaction mixture was heated on water bath at 70°C for about 30 minutes. The pre-cured mixture was transferred into an iron die and was kept at ambient temperature for about 24 hours. Finally, the mixture was cured with hot pressing in a Carver Hydraulic Hot Press at 90°C for 60 minutes under 178 KN force.

**RESULTS AND DISCUSSION**

Amount of the purified matrix powder obtained after extraction with different solvents was found to be 55.6%. Amino groups and hydroxyl groups present in the natural matrix are the active sites where crosslinking with the resorcinol-formaldehyde take place during precuring and curing process which can be presented through the following mechanism Scheme I.

**Characterization**

**Scanning Electron Microscopy (SEM)**

In order to have the conducting impact, the samples were gold plated and the scanning was synchronized with microscopic beam so as to maintain the small size over a large distance relative to the specimen. The resulting images had a great depth of the field. A remarkable three dimensional appearance with high resolution was obtained in case of crosslinked matrix as well as different stages of biodegradation. Intricacies brought about by biodegradation were clearly illustrated by the SEM results of the different samples. The morphological changes in the features of crosslinking matrix after biodegradation at different stages were quite evident from the SEM images. The three dimensional network of the crosslinked natural matrix and its breaking down due to biodegradation at Stage-I, Stage-II, Stage-III and Stage IV could be clearly visualized from SEM studies. Moreover, SEM studies clearly exhibited marked differences between the SEM of crosslinked matrix having smooth homogenous surface with that of
biodegradation matrices of different stages possessing rough heterogeneous surfaces (Figures 1-5).

**Acid resistant studies**

From Figure 6, it is evident that natural matrix after crosslinking with resorcinol-formaldehyde got a lot of resistance towards 5N HCl. The uncrosslinked matrix got disintegrated within 6 hours whereas the crosslinked one was found to be stable towards 5N HCl up to 72 hours beyond which the samples got disintegrated. This could be explained on the basis that with the addition of hydroxyl groups containing resorcinol-formaldehyde to the natural matrix undergoes condensation reaction with the removal of water molecules during precuring and curing processes. This resulted in the formation of three dimensional networks containing covalent bonds thereby providing resistance against the acid attack.

**Base resistant studies**

From Figure 7, it is evident that natural matrix after crosslinking with resorcinol-formaldehyde got a lot of resistance towards 5N NaOH. The uncrosslinked matrix got disintegrated within 6 hours whereas the crosslinked one was found to be stable towards 5N NaOH up to 72 hours beyond which the samples got disintegrated. This could be explained on the basis that with the addition of hydroxyl groups containing resorcinol-formaldehyde to the natural matrix undergo condensation reaction with the removal of water molecules during precuring and curing processes. This resulted in the formation of three dimensional networks containing covalent bonds thereby providing resistance against the attack of water.

**Biodegradation studies**

As evident from Table 1 that biodegradation of the crosslinked matrix takes place under anaerobic conditions. It has been observed that there was a continuous decrease in weight of the sample. Total biodegradation of the sample was found to be achieved after the time interval of 60 days. The mechanism of the biodegradation can be explained through the following anaerobic oxidation:

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Anaerobic Oxidation
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Protein → CH₄ + CO₂ + Other lower hydrocarbons + Oxides of nitrogen (NOₓ)
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**CONCLUSION**

Natural matrices have been found to be eco-friendly because they are biodegradable, easily available, cheap and renewable source of raw materials. But these natural matrices face a lot of problems like water vulnerability, least resistance towards acids and bases and are less stable mechanically as well as thermally. In order to provide chemical resistance, water resistance and mechanical stability to the natural matrices, crosslinker like resorcinol-formaldehyde play an important role. The prepared crosslinked matrix was found to be highly biodegradable in nature along with resistance towards acid and base attack. Moreover, this crosslinked matrix was found to be stable towards attack of water. Thus, it could be concluded that preparation of biodegradable matrices and their crosslinking with resorcinol-formaldehyde resin as well as reinforcement with CNTs is of great importance from technology point of view.

**REFERENCES**

Table 1: Biodegradation studies of R-F Cross linked *Lens culinaris* matrix

<table>
<thead>
<tr>
<th>S. No.</th>
<th>Initial weight (gm)</th>
<th>7</th>
<th>14</th>
<th>21</th>
<th>28</th>
<th>35</th>
<th>42</th>
<th>49</th>
<th>56</th>
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<td>Natural matrix</td>
<td>1.0</td>
<td>0.55</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Cross-linked matrix</td>
<td>2.93</td>
<td>2.55</td>
<td>1.60</td>
<td>1.10</td>
<td>0.75</td>
<td>0.53</td>
<td>0.40</td>
<td>0.23</td>
<td>0.18</td>
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Figure 1: SEM of natural matrix

Figure 2: SEM of biodegradation stage I
Figure 3: SEM of biodegradation stage II

Figure 4: SEM of biodegradation stage III

Figure 5: SEM of biodegradation stage IV

Figure 6: Acid resistant studies
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