Investigation of Novel Biodegradable Almond Gum - Poly Vinyl Alcohol Bi-polymer Blend for Packaging Applications

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ABSTRACT

Biodegradable plastics are designed in a way that they will break when exposed to microorganisms. Bioplastics are commonly made from natural byproducts with rigorously controlled conditions of temperature and humidity in industrial environments. Bioplastics can be degraded naturally in the environment, thereby minimizing the hardship on the natural environment by the conventional plastics. The objective of this work is to synthesize biodegradable plastics from naturally available Almond Gum (Prunus dulcis). A biodegradable plastic from Almond Gum (AG) was prepared by solution casting technique. Further, to strengthen the characteristics of AG bioplastic, poly vinyl alcohol was blended with it and the composite bioplastic was prepared. The synthesized bioplastic membranes were subjected to physicochemical properties such as thickness, moisture, mechanical strength and water absorption analysis. Further, the work is carried out to analyze the antibacterial activity and biodegradable property of the synthesized bioplastic membranes. The prepared bioplastic showed water affinity, displaying its hydrophobic nature and a biodegradability of 43% was achieved. It also displayed quick degradation, thus recommended for commercial application.

Keywords: Bioplastic; Antibacterial; Almond gum; PVA; Biodegradable polymer.

1. INTRODUCTION

Biodegradable materials have been the subject of research in recent years, primarily due to concern about the potential harm caused by synthetic materials. In the present investigation, biodegradable plastics alternative to synthetic plastics were explored. With this perspective, Almond Gum (AG) and blend of AG with PVA were experimented. The biodegradable nature of these two polymers paves way to investigate these bioplastics as potential materials for use in packaging applications. Both materials offer the functionality of a home composting system. Further, AG and PVA may control waste management systems, which is a major issue in the contemporary society (Song et al. 2009).

Almond gum is a water-soluble polysaccharide gum extracted from the prunus tree, which sprouts from the cuts of the almond tree. The structure of almond gum incorporates aldobionic acid, L-arabinose, L-galactose, D-mannose, etc. Almond gum has 92.36% polysaccharide composition, made up of 46.83% arabinose, 35.49% galactose, and 5.97% uronic acid. Its biocompatible properties are due to its hydrophilic and hydroxide-hydrogen bond properties. It has variety of beneficial applications, such as food reinforcing, preservatives, emulsifiers, stabilizers, thickener, suspending agent, adhesive, glazing agent and sources of dietary fiber. Further, due to its low toxicity and high tensile strength, it is suitable for use in the pharmaceutical, cosmetic, and food packaging industries(Farooq et al. 2014; Mahfoudhi and Hamdi, 2015; Rezaei et al. 2016; Srivastava and Kapoor, 2005).

Poly Vinyl Alcohol (PVA) is frequently used to prepare blends. Hydroxyl groups present in PVA form hydrogen bonds which likely have a positive effect on the compatibility of the two polymers. Added advantage of PVA is its water solubility and biodegradability. Its molecular weight is 1.36 g/cm³ with a melting point of 230°C. It has excellent charge storage properties, dielectric properties and exceptional optical properties. It contains a carbon chain with a hydroxyl-group that acts as hydrogen bonding source and forms polymer composites easily (Abdullah et al. 2017; Fernandes et al. 2013). Chemical structure of AG and PVA is depicted in Fig.1.

The production of petrochemical based plastics pose environmental threats. The search for green and sustainable alternatives is intensified in recent years. Therefore, this study suggests the need for investigating
AG as bioplastics and the potential blend of composite of AG with PVA. Thus, this research aims to overcome the abundant plastic wastes left over by developing eco-friendly and decomposable plastics from agricultural wastes.

2. MATERIALS AND METHODS

Almond gum (dry) was purchased from Amazon and PVA was purchased from Sigma Aldrich. Glycerol and gelatin were used as plasticizers and were purchased from Sigma Aldrich. Distilled water was used as a solvent for the preparation of bioplastic membranes. Fig. 2 shows the photograph of (a) AG and (b) PVA used in the investigation.

2.1 Synthesis of Bioplastic Membrane

Solution casting method was adopted to prepare AG membrane and AG: PVA membranes. Initially, dry gum was powdered with a mortar and pestle followed by sieving process. Two grams of AG powder was dissolved in 40 mL of water at 50 ºC using a magnetic stirrer. The mixture was homogenized under constant stirring for 1 hour. Then, glycerol (5 mL) and gelatin (2 mL) were added to the AG solution. Manual stirring was followed for 1 hour in continuation with magnetic stirring to obtain a homogeneous solution. It was then poured into a glass plate coated with Teflon. After a period of 4 days, biomembrane of AG was obtained.

A similar procedure was adopted to prepare AG with PVA membrane. AG solution (1g of AG + 40 mL of water) and PVA solution (1 g of PVA + 40 mL of water) were mixed to obtain AG:PVA composite membrane. Fig. 3 shows the photograph of prepared AG:PVA membrane.

2.2 Tensile Analysis

The mechanical strength of the prepared bioplastic film was checked by CMT-10 Universal testing machine. All tensile testing measurements were conditioned according to the model ASTM D638. Each sample was maintained with a width of 15 mm and a length of 160 mm.

2.3 Film Thickness Analysis

The thickness of bioplastic films was measured by a digital screw gauge. The thickness was measured by holding the sample in between the stylus and anvil. The thickness value was noted directly.

2.4 Moisture Content Analysis

The weighed (initial weight) fresh sample (2x2 cm) was taken in a previously weighed crucible and dried.
in a hot air oven at 100°C for half an hour. The China crucible with dried sample (final weight) was weighed. In each case, the weight of China crucible was deducted and finally the moisture content was calculated.

\[
\text{Moisture content (\%)} = \frac{\text{Loss in Weight (g)}}{\text{Initial Weight of the sample (g)}} \times 100
\]

2.5 Water Solubility Analysis

Water solubility test was carried out for the sample dimension of 2×2 cm at room temperature. Initial dry weight of the film was noted by drying the film at 100°C. Water soluble capacity of the prepared bioplastics was analyzed by soaking the samples for 5 hours in 100 mL of distilled water. Then, the final dry weight of the sample was estimated again by subjecting the membrane to a temperature of 100°C.

\[
\text{Solubility (\%)} = \frac{\text{Initial dry weight (g)} - \text{Final dry weight (g)}}{\text{Initial dry weight of the sample (g)}} \times 100
\]

2.6 Biodegradability Analysis

The AG:PVA bioplastic sample was cut into square of 4×4 cm and buried in the soil for a period of 15 days. The weight of bioplastic was measured before and after the testing. The biodegradability of the sample was calculated by the formula.

\[
\text{Degradation (\%)} = \frac{\text{Initial weight} - \text{Final weight}}{\text{Initial weight}} \times 100
\]

2.7 Antibacterial Activity by Well Diffusion Method

The antibacterial activity of AG:PVA bioplastic against *Pseudomonas* and *Streptococcus* spp. was determined by Agar well diffusion method. The microorganisms (*Pseudomonas* sp. and *Streptococcus* sp.) were streaked in different Petri plates and activated at 37°C for 24 hours. Samples were made in the shape of a Petri dish and coated with 50 mL of *Pseudomonas* and *Streptococcus* spp. After 24 hours of incubation, the diameter of the clear zone showed inhibition of bacterial growth, and it was measured in millimeter (mm).

3. RESULTS AND DISCUSSION

3.1 FTIR Analysis

The FTIR spectra of AG and AG:PVA is shown in Fig. (4 and 5) and the corresponding assignments are shown in Table 1. The AG:PVA composite shows O-H stretching band at 3415 cm⁻¹. Asymmetric –CH stretching of methyl group of AG and –CH stretching of alkyl group of PVA overlaps and it occurs at 2924 cm⁻¹. The band at 2855 cm⁻¹ also corresponds to –CH stretching of PVA. The band at 1727 cm⁻¹ attributes to C=O stretching of both AG and PVA. The band at 1623 cm⁻¹ is due to the presence of carboxylic group in AG. The vibration band at 1422 cm⁻¹ corresponds to symmetric stretching of carboxylic group (uronic acid) of AG. The stretching of CH₂ of PVA overlaps with this.

The band at 1056 cm⁻¹ is the stretching vibration of alcoholic group of AG and C-O-C of PVA. The bands at 821 cm⁻¹ and 712 cm⁻¹ correspond to C-H band stretching of AG (Mansur et al. 2008; Singh et al. 2022).

![FTIR Spectra of AG membrane](image)

Fig. 4: FTIR Spectra of AG membrane

<table>
<thead>
<tr>
<th>Wavenumber (cm⁻¹)</th>
<th>Assignments</th>
</tr>
</thead>
<tbody>
<tr>
<td>3415</td>
<td>O-H stretching of AG and PVA (Mansur et al. 2008; Singh et al. 2022)</td>
</tr>
<tr>
<td>2924</td>
<td>Asymmetric –CH stretching of methyl group of AG and –CH stretching of PVA (Mansur et al. 2008; Singh et al. 2022)</td>
</tr>
<tr>
<td>2855</td>
<td>C-H stretching of PVA (Mansur et al. 2008)</td>
</tr>
<tr>
<td>1727</td>
<td>C=O stretching of AG and PVA (Mansur et al. 2008; Singh et al. 2022)</td>
</tr>
<tr>
<td>1623</td>
<td>Presence of carboxyl group in AG (Singh et al. 2022)</td>
</tr>
<tr>
<td>1422</td>
<td>Symmetric stretching of carboxylic groups of uronic acid of AG and CH₂ stretching of PVA (Mansur et al. 2008; Singh et al. 2022)</td>
</tr>
<tr>
<td>1056</td>
<td>Stretching vibration of alcoholic group of AG and C-O-C stretching of PVA (Mansur et al. 2008; Singh et al. 2022)</td>
</tr>
<tr>
<td>821, 712</td>
<td>C-H bonding vibration of AG (Singh et al. 2022)</td>
</tr>
</tbody>
</table>

Table 1. FTIR vibrational assignments of composite AG: PVA membrane

<table>
<thead>
<tr>
<th>Wavenumber (cm⁻¹)</th>
<th>Assignments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample</td>
<td>Tensile Strength (MPa)</td>
</tr>
<tr>
<td>AGbioplastic</td>
<td>2.75 ± 0.034</td>
</tr>
<tr>
<td>AG:PVAbioplastic</td>
<td>3.79 ± 0.037</td>
</tr>
</tbody>
</table>
Fig. 5: FTIR Spectra of AG: PVA membrane

Fig. 6: Stress vs Strain plot for AG and AG: PVA bioplastics

3.2 Tensile Analysis

Fig. 6 shows the stress versus strain curves for AG and AG:PVA membranes and Table 2 illustrates the tensile strength and elongation percentage of the aforesaid two membranes. Tensile strength of the prepared samples was analyzed to understand their mechanical capability.

The AG and AG:PVA membranes show high mechanical strengths of 2.75 MPa and 3.79 MPa, respectively. The elongation of AG:PVA is comparatively lower than that of the AG membrane. This might be due to the anti-plasticization effect of PVA. Similar results for starch bioplastics and composite bioplastics have been reported earlier(Oleyaei et al. 2016).

3.3 Film Thickness Analysis

The thickness of AG and AG:PVA bioplastic is 0.35 mm and 0.52 mm, respectively. Standardization of thickness of the prepared bioplastics is a vital factor to achieve adequate mechanical property. Marichelvam et al.(2019)reported the thickness for corn starch films as 0.15 mm.Venkatesan et al.have reported thickness of chitosan with AG bioplastics in the range 0.35 - 0.83 mm (Venkatesan et al. 2022). The average thickness of the as prepared bioplastics is found to be in the permissible limit suggested by the Government of India.Hence, this study recommends these prepared bioplastics to be used in packaging.

3.4 Moisture Content Analysis

The moisture content of the prepared biofilm was analyzed using oven dry method. The PVA:AG composite shows the least amount of water absorption. Table 3 depicts the moisture content (in %) of the prepared membranes. This result concludes that PVA has played a role in bringing down the hydrophilic nature of AG-based bioplastics further.

Table 3. Moisture content (%) of the prepared membranes

<table>
<thead>
<tr>
<th>Sample</th>
<th>Initial Weight (g)</th>
<th>Final Weight (g)</th>
<th>Moisture content (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>AG bioplastic</td>
<td>0.287</td>
<td>0.243</td>
<td>15.30</td>
</tr>
<tr>
<td>AG: PVA bioplastic</td>
<td>0.315</td>
<td>0.282</td>
<td>10.47</td>
</tr>
</tbody>
</table>

3.5 Water Solubility Analysis

Water solubility is monitored by means of immersing the samples in water followed by drying (Final dry weight). The solubility of AG membrane is considerably low, since AG contains monosaccharides. However, with the combination of AG and PVA, further decrease in solubility was noticed. AG membrane shows 42% of water affinity, whereas AG: PVA membrane shows 34% of water affinity. Hence, the result suggests the prepared bioplastics may be preferred due to their hydrophobic nature.Venkatesan et al. have reported water solubility of chitosan with AG bioplastics as 35.16% (Mansur et al. 2008). The present result is in line with previous reports.

Table 4. Antibacterial activity of the AG: PVA bioplastic membrane

<table>
<thead>
<tr>
<th>Micro Organism used</th>
<th>Zone of Inhibition diameter (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pseudomonas sp.</td>
<td>18.3 ± 0.1</td>
</tr>
<tr>
<td>Streptococcus sp.</td>
<td>16.4 ± 0.1</td>
</tr>
</tbody>
</table>

3.6 Antibacterial Activity

The AG:PVA composite bioplastic was prepared with the anticipation of utilizing it for packaging applications. The essential attribute for packaging application (especially for food stuff) is its antibacterial property. Pseudomonas and Streptococcus were used as a test group for this activity, using the zone
of inhibition. An interesting antibacterial profile has been observed and Fig.7 shows the zone of inhibition for AG:PVA bioplastic against the species *Pseudomonas* and *Streptococcus*. Table4 depicts the zone of inhibition for the microorganisms.

![Fig. 7: Antibacterial activity exhibited by the AG:PVA bioplastic membrane](image)

The prepared composite exhibits effective antimicrobial activity suitable for packaging applications.

### 3.7 Biodegradability Analysis

The biodegradability of the composite bioplastic sample (AG:PVA) was conducted. Initial weight of the sample was 0.432 g. After 15 days, the weight of the buried sample was noted as 0.245 g. A biodegradability of 43% was achieved in 15 days which concludes the biodegradable nature of the AG:PVA membrane.

### 4. CONCLUSION

The biodegradable plastics were prepared successfully using AG and PVA by solution casting technique. FTIR spectra established the functional compounds present in AG as well as in composite membranes. The characteristics of the bioplastics were evaluated using tensile strength, moisture content, water solubility, antibacterial activity and biodegradable property. Tensile analysis of the prepared samples shows appreciable tensile strength and it is high for the composite bioplastic. The synthesized bioplastic of AG is observed to have the moisture content of 15%. Water solubility analysis concludes the hydrophobic nature of prepared samples. The synthesized composite bioplastic exhibits antibacterial characteristics which is additional requirement for packaging products in addition to tensile strength. The prepared composite bioplastic was found to degrade in a fast phase which recommends the present work for commercial application.

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### CONFLICT OF INTEREST

Authors have no conflict of interest.

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### REFERENCES


