EFFECT OF BORAX ON MECHANICAL PROPERTIES AND 
BIODEGRADABILITY OF SAGO STARCH – POLY(VINYL 
ALCOHOL) BLEND FILMS

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Diterima 22 Juli 2005. disetujui untuk diterbitkan 15 November 2005

ABSTRACT

Biodegradable films of sago starch and poly(vinyl alcohol) (PVA) blends have been prepared by solution casting method. Glycerol was used as a plasticizer. Sodium tetraborate (borax) was used to increase the compatibility of sago starch and PVA. The optimum amount of glycerol is 20% (w/w). The optimum values of tensile strength and elongation at break of the films are obtained with an addition of 8% (w/w) borax. Both pure PVA and sago starch films are apparently transparent. However, their blend films are slightly opaque probably due to phase separation. Increase the amount of PVA in the film leads to an increase of the tensile strength and reduces the elongation of films. Borax increases the elongation at break of the sago starch – PVA blend films and also improves the tensile strength of the films. Biodegradation of original and modified sago starch – PVA blend films have been studied by soil burial method. Percentage of weight loss of SPB films increase with an increase of sago starch content.

Keywords: biodegradable films, sago starch, poly(vinyl alcohol), sodium tetraborate

1. INTRODUCTION

Starch has been incorporated to varying extents into plastics in an effort to develop a biodegradable alternative to petroleum-based commodity plastics. Starch is inexpensive, totally biodegradable and available in large quantities from certain crops produced in abundance beyond available markets. Replacement of petroleum based plastic with starch is also attractive from the standpoint of conserving the petrochemical resources.

Starch blended with the synthetic polymer poly(vinyl alcohol) (PVA) has been studied as a potential biodegradable polymer. Depending on the degree of biodegradability, it seems that PVA might provide a stable support medium for starch films. The mechanical properties and biodegradability of starch – PVA blended films have been reported by several researchers. However, starch and PVA are not compatible when each is added in high concentration. Lawton and Fanta showed that cast films containing starch and PVA could be produced if a small amount of poly (ethylene-co-acrylic acid) (EAA) was added to the formulation. Starch – PVA films cast without EAA will have phase separate during drying. However, the disadvantage of these blends is the lower biodegradation rate of EAA.

Sodium tetraborate decahydrate (known as borax) has a potential to react with both of starch and PVA. In this work, the potential of borax as a compatibilizer for starch – PVA mixture were examined. Therefore, in preparing the starch – PVA blend films, the poor mechanical properties of blend films can be overcome by simultaneous crosslinking reaction of borax with the hydroxyl group of starch and PVA.

Borax has been used for a long time to increase the viscosity of starch-based adhesive. If borax is added to a starch paste, the mass becomes like rubber and cannot be spread because it ‘ball up’ on the surface to which it is applied. Yu et al. found that, small amount of boric acid in starch – glycerol blend films can increase the elongation percentage notably, because the added boric acid reacts with both starch and glycerol, thus a very excellent interconnecting network form. At the same time, adding boric acid can make the starch material more transparent. Salmoral et al. reported that addition of boric acid in a concentration of 3% to the blends containing 11.7% starch of total weight gelatinized with water and glycerol (21.7%) has improved the tensile strength and percent of elongation of plastic compounds. The addition borax to starch – PVA – glycerol blends is suggested to improve the properties of the blends through the forming of interpenetrating network.
2. MATERIALS AND METHODS

2.1. Materials

Sago starch was provided by Nitsei Sago Industries Sdn. Bhd., Mukah, Sarawak, Malaysia. Sago starch was used as received without further purification. Poly(vinyl alcohol) was obtained from Merck with molecular weight of 72000 and 98% hydrolyzed. Sodium tetraborate decahydrate (borax) Na₂B₄O₇·10H₂O was obtained from BDH Chemical, England. Glycerol (density = 1.25 g/mL) was obtained from Univar Australia.

2.2. Preparation of Sago Starch Films

Sago starch was first dispersed in distilled water and then heated under stirring at 80°C in a three-neck flask for 45 minutes. Immediately, after gelatinization process, a required amount of glycerol was added. Stirring was continued until a homogeneous mixture was obtained. The mixture was then poured into a petry dish and dried in an oven for 1 day at 60°C. A film with thickness of about 100µm was obtained.

The film thickness was measured using a micrometer (Mitutoyo, Japan). Measurement were taken at five different locations on the film and recorded to the nearest 1µm. Mean thickness values were used for calculating mechanical properties.

2.3. Preparation of Sago Starch – PVA Blends Film

Sago starch was first gelatinized as described above. A required amount of poly(vinyl alcohol) solution was then added into the flask and stirred for another 1 hour at 50°C. This was followed by the addition of glycerol and stirring until a homogeneous mixture was obtained. The starch mixture was then poured into a petry dish and dried in an oven for 1 day at 60°C.

Table 1. Composition of sago starch – PVA blends films used to study effect of borax

<table>
<thead>
<tr>
<th>Sample</th>
<th>Composition</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Sago starch (g)</td>
</tr>
<tr>
<td>B02</td>
<td>2.50</td>
</tr>
<tr>
<td>B03</td>
<td>2.50</td>
</tr>
<tr>
<td>B04</td>
<td>2.50</td>
</tr>
<tr>
<td>B05</td>
<td>2.50</td>
</tr>
<tr>
<td>B06</td>
<td>2.50</td>
</tr>
</tbody>
</table>

2.4. Preparation of Sago Starch – PVA – Borax Films

Preparation of sago starch – PVA solution was carried out as describe above. To prepare sago starch – PVA – borax blends, solution of sodium tetraborate decahydrate was then added into sago starch – PVA solution, and stirred for another 30 minutes. This was followed by the addition of glycerol and stirring the mixture until the homogeneous mixture was obtained. The solution was then poured into a petry dish and dried in an oven for 1 day at 60°C.

Effect of the addition of borax on the film properties was studied by varying the amount of borax in the formulation from 0.2 to 0.6 gram (Table 1). Table 2 shows compositions of the blends containing fixed amount of borax and glycerol but varied of sago starch – PVA ratio.

2.5. Mechanical Properties

Samples for tensile measurement were conditioned at ambient temperature (25± 3 °C). The samples were placed in a closed chamber containing saturated NaBr solution in distilled water for 7 days . The measurements were carried out using an Instron machine model 4403 according to ASTM D638 with a crosshead speed of 50 mm/min.

2.6. Biodegradation

Biodegradation of all samples was studied using the soil burial method according to procedure used by Thakore et al. . The sample in the form of thin films were placed in soil, in a well 60 cm x 60 cm wide and 35 cm deep in position depending on the sample composition and scheduled time of sampling. Samples were also tagged for easier identification. The films were scheduled to remain at the site for 2 months with a sample removed every two weeks, in order to follow their biodegradation process.
Table 2. Composition of sago starch – PVA – borax films used to study effect of varying sago starch – PVA Ratio

<table>
<thead>
<tr>
<th>Sample</th>
<th>Composition Ratio</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Sago starch (g)</td>
<td>Poly(vinyl alcohol) (g)</td>
</tr>
<tr>
<td>SPB10</td>
<td>4.50</td>
<td>0.50</td>
</tr>
<tr>
<td>SPB20</td>
<td>4.00</td>
<td>1.00</td>
</tr>
<tr>
<td>SPB30</td>
<td>3.50</td>
<td>1.50</td>
</tr>
<tr>
<td>SPB40</td>
<td>3.00</td>
<td>2.00</td>
</tr>
<tr>
<td>SPB50</td>
<td>2.50</td>
<td>2.50</td>
</tr>
</tbody>
</table>

3. RESULTS AND DISCUSSION

Effect of varying of glycerol amount on tensile properties of sago starch film are shown in Figure 1 and 2. Tensile strength and elongation at break of each film were evaluated after it was stored at room temperature in 50 ± 5% RH for 7 days. The results show that the increasing amount of the plasticizer (glycerol) in the film reduces the tensile strength but increases the elongation at break. This is probably due to a greater mobility of starch macromolecules as the amount of glycerol in the film is enhanced. Films containing less than 20% (based on the weight of starch) of glycerol are brittle. Although they have a high tensile strength, they are not suitable for an application that needs a high elasticity. Adding of glycerol into the starch films decreases the tensile strength, but improves their plasticity and flexibility especially when the glycerol content is 20% and higher. However, too high of glycerol content causes the film to be sticky and difficult to handle. Consequently, the tensile strength and elongation at break of the film containing 33% glycerol could not be examined. The elongation of the film containing 20% glycerol is higher than 100% which meets the Swanson’s target value for the elongation at break. This is in accordance with the results of other researchers.

Figure 1. Effect of glycerol on tensile strength of sago starch film

Figure 2. Effect of glycerol on elongation at break of sago starch film
2.1

2.2

2.7

3.0

3.1

Figure 3. Tensile strength of sago starch – PVA blends film as a function of PVA contents

Figure 4. Elongation at break of sago starch – PVA blends film as a function of PVA contents

Table 3. Effect of borax content on tensile strength and elongation at break of the sago starch – PVA blend film

<table>
<thead>
<tr>
<th>Sample</th>
<th>Composition</th>
<th>Tensile Strength (MPa)</th>
<th>Elongation at break (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Sago Starch G</td>
<td>PVA G</td>
<td>Glycerol mL</td>
</tr>
<tr>
<td>B02</td>
<td>2.50</td>
<td>2.50</td>
<td>1.0</td>
</tr>
<tr>
<td>B03</td>
<td>2.50</td>
<td>2.50</td>
<td>1.0</td>
</tr>
<tr>
<td>B04</td>
<td>2.50</td>
<td>2.50</td>
<td>1.0</td>
</tr>
<tr>
<td>B05</td>
<td>2.50</td>
<td>2.50</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Pure PVA and sago starch films were clear and apparently transparent. However, the blended film was slightly opaque. It can be observed visually that the sago starch – PVA blends film was phase separated.

The variation of the tensile strength and elongation at break of sago starch – PVA blends film with the PVA content is shown in Figure 3 and 4 respectively. An increase of the amount of PVA leads to an increase of tensile strength and reduction in elongation at break. This behavior is probably due to the decrease of the macromolecule chain mobility as a result of the presence of the strong intermolecular hydrogen bond as well as the association, which are established between the macromolecules of the blend component.

Table 3 shows the influence of borax content on the tensile strength and elongation at break of sago starch – PVA blend films containing the same amount (2.50 g) of sago starch and PVA. The results show that the maximum values for both tensile strength and elongation at break are obtained when the borax content in the film is 8% (B04 sample).
Table 4. Effect of borax on the tensile properties of sago starch – PVA blends film

<table>
<thead>
<tr>
<th>Composition of Sago Starch – PVA</th>
<th>Tensile Strength (MPa)</th>
<th>Elongation at Break (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Without Borax</td>
<td>With Borax</td>
</tr>
<tr>
<td>90 : 10</td>
<td>2.1</td>
<td>2.1</td>
</tr>
<tr>
<td>80 : 20</td>
<td>2.2</td>
<td>3.6</td>
</tr>
<tr>
<td>70 : 30</td>
<td>2.7</td>
<td>6.6</td>
</tr>
<tr>
<td>60 : 40</td>
<td>3.0</td>
<td>9.8</td>
</tr>
<tr>
<td>50 : 50</td>
<td>3.1</td>
<td>12.1</td>
</tr>
</tbody>
</table>

3.1. Mechanical Properties of Sago Starch – PVA – Borax Blend Film

Table 4 shows tensile strength and elongation at break of sago starch – PVA blend films with and without borax. Incorporation of borax in the films containing 20% PVA or more increases both tensile strength and elongation at break of the films. However, addition of borax into the film containing 10% PVA only improves its elongation at break for almost 100%; the tensile strength remains the same. Addition of borax also increases the viscosity of the blend solution. When the solvent of the solution is evaporated, the films formed are more compact than that of without borax.

The addition of borax to the starch – PVA – glycerol blend films improves properties of the blends through the formation of interpenetrating polymer network (IPN). IPN is one of the three general classes of heterogeneous material, along with composites and blends. IPNs differ from composites because there is no load-bearing reinforcing agent like fibers put into the material. Blends and IPNs are most closely related but there are distinct differences. Blends generally consist of two or more polymers that are mixed together but none of the components are crosslinked to any appreciable extent. On the other hand, IPNs are comprised two polymeric components in which one or both of the components are crosslinked.

3.2. Biodegradability of Starch-based Films

In this research, the biodegradation of original (SP samples) and modified (SPB samples) sago starch – PVA blends films were studied by soil burial. This method is the most often used to follow the biodegradation of starch-based films. In the soil, water diffuses into the polymer sample, causing swelling and enhancing biodegradation.

In general, the biodegradation rate of starch-based films depends on factors such as water absorption by the film, temperature, microbial population, oxygen content, and mainly, amount of starch incorporated in the film. Figure 5 and 6 show the % of weight losses of SP and SPB film samples, respectively, due to the degradation.

The main enzymes that consume starch are the α- and β-amylases. These can be found in most microorganisms such as bacteria and fungi, in animals, in human body and also in some plants. The mechanism proposed for the α-amylase catalysis is the formation of the complex in an active side close to the ether bond between two α-D-glucopyranose group and finally the breakage of the glycosidic ether group. The same mechanism is also proposed for β-amylase, which can break the α-D-(1-6) linkages of amylopectin.

4. CONCLUSION

The addition of borax to the starch – PVA – glycerol blend films improves properties of the blends through the formation of interpenetrating polymer network (IPN). Percentage of weight loss of both SP and SPB films increase with an increase of sago starch content. The biodegradation rate of starch-based films depends on factors such as water absorption by the film, temperature, microbial population, oxygen content, and mainly, amount of starch incorporated in the film.

ACKNOWLEDGEMENTS

The author gratefully acknowledges The Ministry of Science and Technology Malaysia for supporting the part of this work through the IRPA project. The author would also like to thank Assoc. Prof. Dr. Mohamad Zaki Ab. Rahman and Prof. Dr. Wan Md. Zin Wan Yunus of Chemistry Department, Universiti Putra Malaysia for helpful suggestions and discussion.
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