

# Synergism Between Sago Starch and Chitosan in Enhancing Biodegradable Film Properties

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**Abstract** The research studied properties of biodegradable film from the mixture of sago starch and chitosan. The biodegradable film was obtained by solution casting method with addition of glycerol 1.5% as plasticizer. Mechanical properties of biodegradable film from sago starch and chitosan were determined including tensile strength, elongation, color and biodegradability. To study shape surface morphology of biodegradable film, the microfracture of biodegradable film was observed by scanning electron microscopy (SEM). Biodegradable film with the composition of sago starch and chitosan 1:1 significantly had the lowest number of tensile strength. The study revealed that addition of chitosan improved biopolymer elongation and brightness, but it weakened tensile strength of biopolymer and reduced its biodegradability.

**Keywords** Sago starch · Chitosan · Biodegradable film · SEM

## 1 Introduction

Biodegradable film from the mixture of polysaccharides and proteins has been studied intensively as a substitute for synthetic polymer. Researchers have their own definitions of biodegradable film/biodegradable plastic. Biodegradable film is a plastic material that its chemical structure is change under certin condition therefore affects its properties. The chemical structure changes can occur from the attack of microorganisms such as fungi, bacteria and algae. Another definition of the biodegradable plastic is a polymer in which the molecular weight is being lower because of the degradation wherein at least one step in the degradation process occurs through the natural metabolism of the organism.

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One of the important properties of biodegradable film is its biodegradability that is an ability of materials to be degraded. The term of biodegradation sometimes confused with the term of deterioration. Deterioration can be defined as a loss in physical integrity of a material, whereas biodegradation is a biochemical transformation of compounds by microorganisms and results in mineralization or incorporation into microbial biomass. Biodegradation process produces  $\text{CO}_2$  and  $\text{H}_2\text{O}$  under aerobic conditions or  $\text{CH}_4$  and  $\text{CO}_2$  under anaerobic conditions. From the definition, a distinction can be drawn that the deterioration will be fragmented plastic, but the plastic particles will still remain in the environment, while biodegradation will decompose the plastic particles through the process of mineralization.

Starch including from mung bean, potato, sweet potato and its derivatives has been widely studied in the manufacture of biodegradable films and capsules. Starch is used as a substitute for gelatin in the manufacture of these biodegradable films for its simplicity to be molded into films and its properties as high oxygen barrier, and it has a good mechanical strength [1]. Among other starches, sago starch has some advantages as the base material of biodegradable films because it has low temperature on gelatinization, low degree on sineresis, high degree on viscosity and it is easy to be attacked by mold. Another advantage is that sago starch has fairly high amylose content approx. 27% which is suitable for making a biodegradable film with solid gel [2].

Basically, polysaccharide-based biodegradable films have brittle properties and their mechanical properties are unfavorable. Therefore, in the manufacture of biodegradable film gelling agent, e.g., chitosan, and plasticizer, e.g., glycerol, sorbitol and polyethylene glycol are added to overcome fragility and to improve elasticity [3]. Chitosan is a natural cationic polysaccharide obtained from deacetylation of chitin which is widely available in nature. According to the chemical structure, chitosan consists of monomer 2-amino-2-deoxy-D-glucose (glucosamine) which shows the properties of biomedical polymers such as non-toxic, biocompatible and biodegradable. Chitosan structure is similar to cellulose, and its ability to form a gel in the acid is due to its properties to build a matrix in the polymer system. In the manufacture of biodegradable films, the addition of chitosan and the presence of polyacrylic acid (PAA) produce  $\text{CaCO}_3$  crystals that will affect the morphology of film. There was areport that the addition of chitosan to maize starch biodegradable film produces a film with more strength.

The aim of the study was to produce biodegradable film which has similar properties with synthetic plastic. The synergism between sago starch and chitosan was then studied including the character of biodegradable film produced, the effect of sago starch and glycerol concentration on the physical and mechanical properties, i.e., tensile strength and elongation, and the rate of biodegradable film degradation.

## **2 Research Method**

### **2.1 Sample Preparation**

Biodegradable film was prepared from sago starch and mixture between sago starch and chitosan as follows: (1) sago starch without chitosan (BS); (2) sago starch blended with chitosan and the ratio was 1:1 (BKS I); and (3) sago starch blended with chitosan and the ratio was 2:1 (BKS II). The film was prepared by mixing polysaccharides with glycerol and distilled water at 50 °C. The solution was stirred using an agitator at 100 rpm for 30 min. A commercial sago starch and chitosan were then added to the glycerol solution, while it was stirred and distilled water was added until the solution reached 200 mL. The stirring process was continued until it became a clear solution. The solution then poured onto five pieces of 20 cm × 30 cm acrylic glass plate and stood for 15 min. After the frame was released, biodegradable films on acrylic glass plate were then dried in oven at 50 °C. After drying, the film was released from acrylic glass plate and wrapped by aluminum foil.

### **2.2 Tensile Strength and Elongation Measurement**

Biodegradable film was cut and linked horizontally on Instron Universal Testing Machine tools (Zwick Z.05 texture analyzer) connected to a computer for data analysis.

### **2.3 Brightness Test**

Measurement of brightness used a color index Chroma Meter Minolta CR-600 with the attribute “*L*” indicating the level of brightness, “+*a*” degree of redness, “-*a*” degree of greenness, “+*b*” yellowish level and “-*b*” the bluish level.

### **2.4 Biodegradability Test**

Biodegradability test was conducted using soil burial test of Behjat et al. [4] with slight modifications. Testing was done by cutting the film with the size of 5 × 10 cm. It was then buried approx. 20 cm below the ground for one week. The film was observed and the film remained was measured.

## 2.5 Scanning Electron Microscopy (SEM)

Observations using electron microscopy were conducted to determine the microstructure of biodegradable films.

## 3 Results and Discussion

### 3.1 Film Properties

Physical characteristic of biodegradable film was shown in tensile strength and elongation (Table 1). Chitosan affected characteristics of biodegradable film. The cross-link between sago starch and chitosan (BKS I and BKS II) lowered tensile strength of film produced due to decrease in starch concentration. The decrease in starch concentration was in line with the decrease in amylose content in biodegradable film. The result was similar to the previous research that mung bean starch with the highest amylose content (30%) had the highest tensile strength [1]. The increase in amylose content also increased aggregation from the formation of hydrogen bonds between polymers that formed amylose microcrystal that resulted in film with high tensile strength [5].

The result showed that BKS I film had a higher tensile strength than BKS II film. Studies showed that increase in the ratio of starch and chitosan composite decreased tensile strength of the film. The decrease in tensile strength that was in line with the increase in starch ratio was caused by the formation of intramolecular hydrogen bonds' starch that was larger than the intermolecular bonding, resulted in separation phase between the two main components [6].

Starch contains macromolecules, such as amylose and amylopectin, which form a solution when heated with water and become a gel after cooling. During the process of forming a gel, inter- and intramolecular cross-linking is established to form microcrystalline regions [7]. Starch components without plasticizer resulted in increase in crystallinity that occurred during the formation of the film which increased tensile strength. This was more obvious in more rigid starch films [8]. It also occurred in elevated concentrations of chitosan that result in decreased crystallinity of the films [6].

**Table 1** Mechanical properties of sago starch–chitosan biodegradable film

Sago starch–chitosan concentration	Tensile strength (MPa)	Elongation (%)
BKS I	29,277 <sup>b</sup>	493,239 <sup>a</sup>
BKS II	24,502 <sup>b</sup>	447,872 <sup>a</sup>
BS	69,004 <sup>a</sup>	318,846 <sup>a</sup>

*Description* The value is the mean of three replicates of analysis  $\pm$  standard deviation; same superscript letters indicate no significant difference ( $P > 0.05$ )

For elongation, BSI film had the highest elongation values though not significantly different from other treatment. It showed that the addition of chitosan can increase the percentage elongation value of the film. Elongation value was inversely proportional to the film tensile strength values. Previous study showed that the combination of starch–chitosan film had significantly higher elongation value than the film made from starch or chitosan itself [9]. Higher amylose content in the film will result in a stronger starch intermolecular force, therefore the film with less flexibility and stronger in tensile strength has lower number in elongation [6].

### 3.2 Film Brightness

Table 2 shows that addition of chitosan affected starch color. *L* value stated brightness parameters with the range value 0–100 (black to white), while the *b* value stated chromatic color mixture of blue–yellow with +*b* values of 0–70 for yellow and 0 to –70 for blue.

Increasing the *L* value indicated the level of brightness of the starch. The results of the study showed that the reduced levels of sago improved the brightness of biodegradable films. In *b* value, decreasing the levels of sago in biodegradable films was in line with the decrease in *b* value. Previous study from Chillo et al. [10] indicated the same phenomenon where the *L* and *b* values increased with increasing chitosan concentrations.

### 3.3 Biodegradability Test

Biodegradation is the result of an enzymatic process and involves both microorganism and macroorganism. Microorganism accelerates degradation by enzymes and may occur in aerobic or anaerobic conditions. Linear polymer is generally more easily degraded than the branched polymers [11]. Table 3 shows the result of film biodegradation.

Sago starch–chitosan film (BKS I and BKS II) had lower degradation compared to sago–starch film (BS). It indicated that addition of chitosan inhibited the

**Table 2** Effect of chitosan on degree of white sago starch films

No	Film	Color		
		<i>L</i>	<i>A</i>	<i>B</i>
1	BKS I	31.49 <sup>b</sup>	–0.67 <sup>a</sup>	3.85 <sup>c</sup>
2	BKS II	25.47 <sup>a</sup>	–0.39 <sup>a</sup>	2.21 <sup>b</sup>
3	BS	25.98 <sup>a</sup>	–0.73 <sup>a</sup>	1.86 <sup>a</sup>

*Description* The value was the mean of three replicates of analysis  $\pm$  standard deviation; same superscript letters indicate no significant difference ( $P > 0.05$ )

**Table 3** Biodegradation of sago starch–chitosan film

Sago starch–chitosan concentration	Soil burial test (cm <sup>2</sup> /week)
BKS I	62
BKS II	24
BS	104

degradation by microbes because chitosan was antimicrobial and antifungal, the same phenomenon which was found in Fernandez-Saiz et al. [12] and Ziani et al. [13]. In other work, hydrogen bond interactions between tapioca starch and chitosan reduced the availability of hydrophilic groups, diminishing their interactions with water molecules. It was also observed that when chitosan coating was applied to salmon fillet pieces, the antimicrobial effectiveness was greater than when chitosan–tapioca starch blends were applied [14]. However, observations of the rate of degradation showed that degradation of BKS II film was smaller than BKS I. It might be due to the presence of other organisms in the treatment media that were accelerating the degradation process.

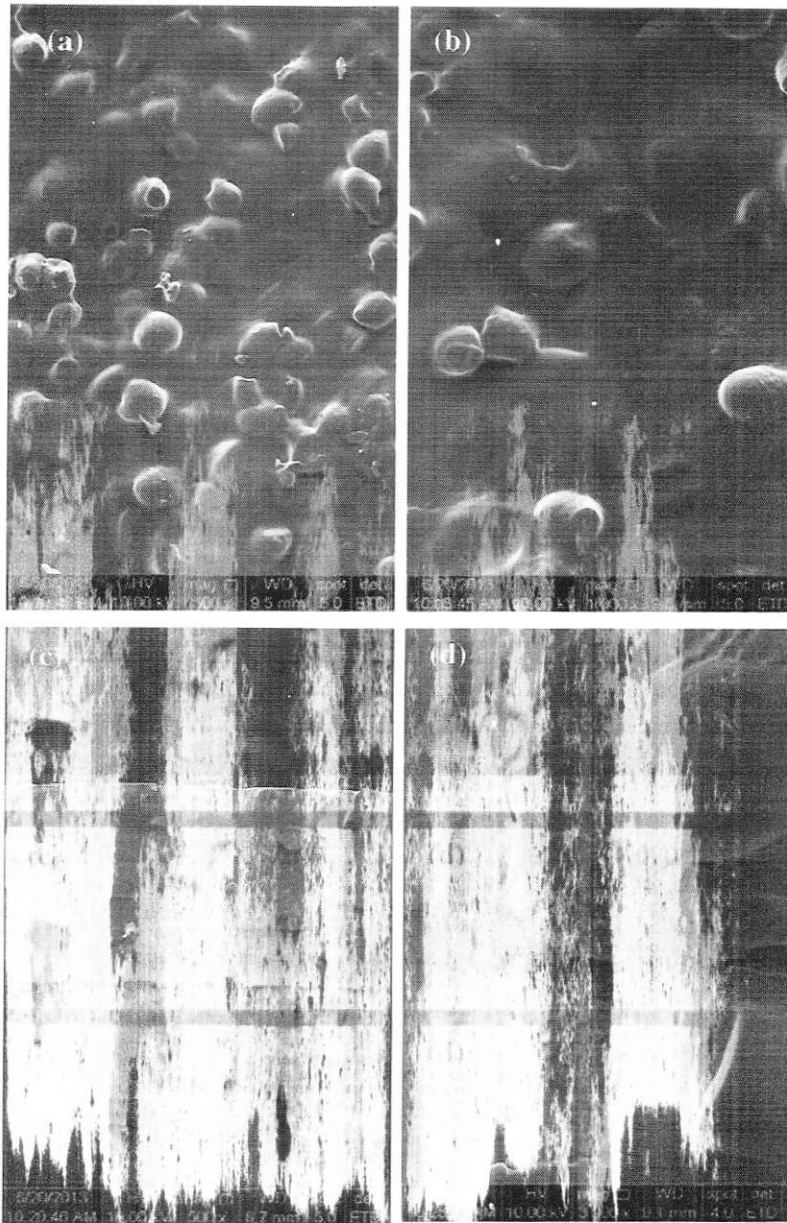
### 3.4 Microstructure Observation Using SEM

Observations using electron microscopy were conducted to determine microstructure of the film. The SEM analysis was only conducted for BKS I that had better mechanical properties compared to BKS II and BS. The results of SEM analysis of BKS I are presented in Fig. 1.

Figure 1 shows the outer structure of BKS I that was compact, continuous, prevalent and not porous. The result was similar to the chitosan film SEM from Valenzuela et al. [15] and sago starch film Afiq and Azura [16]. The structure was not homogeneous that might be caused by the imperfection of intramolecular bond between chitosan and sago starch. The imperfection was due to the starch melting point that did not reach maximum temperature. It prevented starch granule from fully shedding so that it became soluble and lost its intermolecular bond [14].

## 4 Conclusion

The study shows that chitosan improved mechanical properties of sago starch film in elongation value and also improved film brightness. However, addition of chitosan decreased tensile strength value and degradation rate. The investigation of permeability characteristics and mechanical properties with addition other resources was needed to obtain comprehensive knowledge of biodegradable film.



**Fig. 1** Scanning electron microscopy observations of BKS I with various magnifications after soil burial test for 1 week. Biodegradable film looks over 500 $\times$  magnification (a) and 1000 $\times$  (b), 500 $\times$  magnification side view (c) and 3000 $\times$  (d)

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