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Indian
Journal of Advances in
Chemical Science

Indian Journal of Advances in Chemical Science 4(4) (2016) 355-361

Effect of Gelatin Content on Potato Starch Green Composite Films

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Received 1st December 2015; Accepted 26th September 2016

ABSTRACT

Bio-based solution cast green blend films were prepared using potato starch (PS) and gelatin using Propylene Glycol as a plasticizer. The effect of the gelatin content on mechanical properties (tensile strength [TS] and % of elongation) were investigated. The moisture content and water solubility of the pure and blend films was also determined. The ternary blend containing 30% of gelatin content resulted in films with high TS when compared to other films. Increasing gelatin concentrations increased TS, reduced elongation at break and decreased water solubility and moisture of the blended films. Thermogravimetric analyses thermograms and scanning electron microscopy micrographs confirmed improved thermal stability and homogeneity of PS-gelatin films. These results indicated good compatibility between the molecules of PS and gelatin at the 30% of PS/gelatin blend films.

***Key words:** Potato starch, Gelatin, Green blend films, Packaging applications, Casting method, Thermal stability, Mechanical properties.*

1. INTRODUCTION

The growing environmental awareness imposes to the users and eco-friendly attributes to both materials and processes. As a consequence, biodegradability is not only a functional requirement but also an important environmental attribute. Until now, the use of non-renewable (nonsustainable), petroleum based synthetic polymers for making packaging materials has increased significantly. The main problems associated with synthetic polymers are that they are nondegradable and non-renewable, so they pose serious ecological problems. The development of new biodegradable packaging materials from environmentally friends and renewable raw materials is an inevitable and interest research area.

Currently, there has been an increasing interest in the development of green polymer films from biomass resources with a small ecological load and at the end of their life; most of them to end up in landfills [1,2]. Green polymer films are generally designed from biomass such as polysaccharides, proteins, lipids, and their derivatives [3-7]. Among these green polymers, starch is an ideal polysaccharide material and composed of amylose, a linear or sparsely branched polymer and amylopectin both of which are polymers of a-D-glucose units, a highly branched polymer being used in the production of biodegradable packaging materials due to its low cost, biocompatibility, biodegradability, and ease of chemical derivatization [8-11].

Besides, the thermoplastic starch (TPS) film additives in the form of plasticizers (glycerol, sorbitol, and propylene glycol) used to enhance the flexibility of biopolymer films is also stated to impair the mechanical and barrier properties [12]. However, wide application of starch-based films has been limited due to their moisture sensitivity, poor mechanical properties, and brittleness [13]. Modifications are often accomplished by blending TPS with either non-biodegradable polymers or biodegradable polymers [14-16]. Blending of polymers is an easier and more effective way to develop new materials with a variety of designed physical and chemical properties at the lowest cost. The use of natural blends of protein, polysaccharides, and lipids directly obtained from agricultural sources takes advantage of each component in the original system and appears to be a new opportunity for material in the area of films used for packaging edible food stuff. Proteins are extensively used to form polymer films [17]. The interest in blending polysaccharides, proteins and lipids is due to the complementary advantages and disadvantage of these components [18-20]. Polysaccharide-based films generally have good mechanical properties and are also

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effective barriers against low polarity compounds, but they do not offer a good barrier against moisture [21]. Nevertheless, protein-based films are usually excellent barriers to oxygen, carbon dioxide, and some aromatic compounds, but their mechanical properties are not satisfactory, which limits their applications [22]. Some green films based on the blends of polysaccharides and proteins such as methylcellulose-wheat gluten [23], hydroxypropyl starch-gelatin [24], soluble starch caseinate [25], and glucomannan-gelatin [26] showed varying results on the physicochemical properties.

Gelatin is a protein, translucent, colorless, brittle (when dry), flavorless foodstuff, and derived from collagen obtained from various animal by-products [27,28]. Gelatin contains 14% hydroxyproline, 16% proline, and 26% glycine [29]. A typical structure is -Ala-Gly-Pro-Arg-Gly-Glu-4Hyp-Gly-Pro- [30]. Gelatin is an abundant raw material, produced in the whole world at low cost and has excellent film forming properties [31]. Many natural polymers studied have been reported to be blended with gelatin [32]. The properties of blended films based on gelatin and different kind of starches such as corn starch [33], and cassava starch [34,35] have been investigated. However, there are if any limited studies about potato starch (PS) films with added gelatin.

Therefore, the aim of this work is to prepare the green blend films based on PS, gelatin and propylene glycol by casting, different contents of gelatin being added into the PS matrix. The effect of introducing gelatin into the PS matrix on the thermal stability, tensile properties, water solubility, and morphology was then investigated. It is envisaged that this type of films developed might find application exclusively in food packaging industry.

2. EXPERIMENTAL

2.1. Materials

All raw materials used in this study are available as commercial products. PS was obtained from S.D. Fine Chemicals Limited Mumbai, India. Gelatin was parched from Labcon Chemicals Limited South Africa. EP grade propylene glycol was purchased from Sigma-Aldrich Chemicals, South Africa.

2.2. Fabrication of PS/Gelatin Green Composite Films

The PS/gelatin blend green films were produced using solution casting method described below. Film-forming solutions were made by dispersing PS (control) and gelatin containing different contents based on the total mass of dry solid (5 g) and propyleenglycol (20% wt) in 200 ml of distilled water as shown in Table 1. To achieve starch gelatinization, the mixture was stirred constantly at 90°C for 40 min at 1500 rpm using a magnetic stirrer on a hot plate. Film thickness was controlled by measuring the volume of film-forming solution. The film forming solutions (200 mL) were cast on Teflon plates and cured at (60 ± 1)°C for 10 h with circulating air. The formed films were removed from the plates and kept at (25 ± 1)ºC in a desiccator containing Mg(NO3) 2.6H2O (50 ± 2% RH) for 72 h before characterization. The processing path of the film casting is shown in Figure 1.

Table 1: Composition of film solutions.

Films	**Composition of film solution (g/100 ml water)**		
	Potato starch	**Gelatin**	**Propylene glycol**
Control	5	0	1.5
Gelatin (10%)	4	1	1.5
Gelatin (20%)	3	2	1.5
Gelatin (30%)	2	3	1.5
Gelatin (40%)	1	4	1.5
Gelatin	0	5	1.5

2.3. Morphology

The blend films were dried in vacuum and stored in a desiccator before the scanning electron microscopy (SEM) micrographs recorded. The dry samples were gold-coated by an electrodeposition technique to impart electrical conduction. The morphology of the films was studied using JEOL JEM-7500F (Tokyo, Japan) operated at an accelerating voltage of 2 kV.

2.4. Thermal Properties

Thermogravimetric analyses (TGA) of blend films were carried out using SDT Q 600TA instruments. The specimens were scanned at a heating rate of 10°C/min in the range of 30-600°C in N_2 atmosphere.

2.5. Mechanical Properties

Tensile tests were performed as per ASTM D 882-91 standard. Specimens were cut into rectangular shape with dimensions 100 mm × 20 mm × 0.2 mm. Tensile properties such as the maximum stress and % elongation at break were determined using an MTS-universal testing machine at a crosshead speed of 50 mm/min maintaining a gauge length of 50 mm. In each case, five specimens were tested and the average values are reported.

2.6. Moisture Content and Water Solubility

The moisture content in the film was determined by measuring the weight, on drying films in an oven at 110°C until no further reduction of weight was observed. Three replicates of film specimens were used to measure the moisture content. The film solubility method was adapted from the literature [36]. The initial dry matter of each film was obtained after drying film specimens at 65°C for 24 h. These dried films (about 0.3 g) were weighed

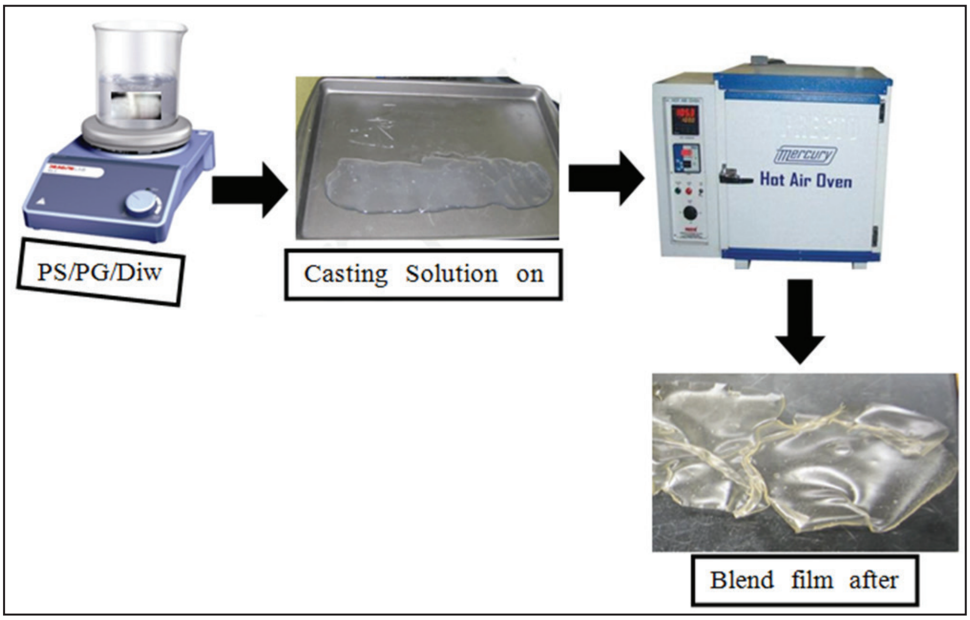

Figure 1: The processing path of the film casting.

(initial dry weight) and immersed in beakers containing 50 mL distilled water at 23°C that was then sealed and periodically agitated for 24 h. Tests were performed in triplicate, and the solubility was calculated using Equation:

$$\text{Solubility }(\%)=\left[\frac{\text{Initial weight}-\text{Final weight}}{\text{Initial weight}}\right]\times 100$$

3. RESULTS AND DISCUSSION

3.1. Morphological Analysis

Figure 2a-c shows SEM images of starch, gelatin, and blend films. All of the films showed visibly smooth and compact surface structures similar to the microstructure of cassava starch-gum bio-based films [37]. The SEM image of PS film (Figure 2a) showed a relatively smooth and continuous cross section without pores and cracks, which confirmed a dense and homogeneous structure. The micrographs of gelatin film exhibited a patchy and rough cross section (Figure 2b), which may occur because of the flexibility of gelatin films. Blending with PS was noted to improve the internal structure of gelatin films, in which PS-gelatin blended films showed less roughness and more compact and dense appearance (Figure 2c). The observed microstructures also confirmed the tensile strength (TS) improvements imparted by adding gelatin to PS films. Based on the mechanical property results (Section 3.3), a PS blended film with 30% content of gelatin was chosen for cross-section analysis. SEM micrographics of starch and gelatin films showed compact structures. Blend films presented a more heterogeneous surface which seems to have a good compatibility among the polymers. Possibly, the addition of gelatin to starch matrix changes the microstructural arrangement of gelatin chains that are regrouped in helicoidal way during films drying.

3.2. Moisture Content and Water Solubility

Solubility of starch in water is an important criterion for the applications of starch blend films. The solubility of PS as a function of gelatin content is shown in Figure 3. It was obvious from Figure 3 that the water solubility of control films was 80% and for the other films the solubility was decreased with increasing gelatin content with 40% of gelatin content in PS having lowest solubility than other the PS/gelatin blend films. This may be attributed to good compatibility between PS and gelatin when the content of gelatin in the matrix is 40%. Possibly the crystallinity changed with 40% gelatin which resulted in decreased water solubility. These results are consistent with the water solubility of cross-linked fish gelatin-chitosan films reported literature [38]. Figure 3 also shows the moisture content of the starch films as a function of gelatin content. It is evident from this figure that the moisture content decreased with increasing gelatin content. The observed results can be attributed to the formation of intermolecular hydrogen bonding between the hydrogen of the hydroxyl groups of starch and the hydrogen of the amino groups of gelatin, thus hiding the hydrophilic films are generated as the content of gelatin is increased in the film forming solution.

3.3. Mechanical Properties

Mechanical properties (TS and elongation) of the PS/gelatin blend films with variable gelatin content is presented in Figure 4. It is clearly evident that the gelatin content had increased TS but decreased the percentage elongation (% E) at break.

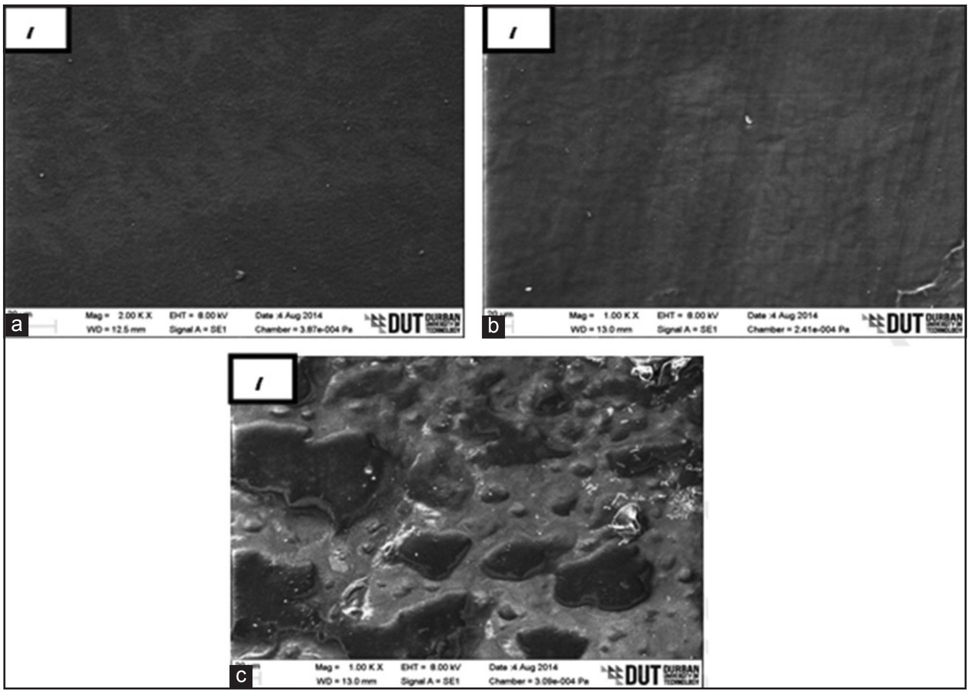

Figure 2: Scanning electron microscopy observations of potato starch films: (a) Potato starch film, (b) gelatin film and, (c) The film with 30% gelatin.

These results are in agreement with results obtained by other researchers for chitosan-cassava starch-gelatin film [39,40]. Films with 30% of gelatin had the highest TS. The increasing TS of the blended PS films with increasing gelatin content is attributed to the formation of the macromolecular polysaccharides of large chains, as starch, can interlace with gelatin, increasing their mechanical resistance. Both polymers can form structural networks by interactions between anionic groups of polysaccharides and cationic groups of gelatin, strengthening the final structure [41]. Blend films are systems with free hydroxyl groups to form hydrogen bonds between PS, gelatin and propyleenglycol. Therefore, at the 40% content, the TS of the films slightly decreased. This phenomenon may possibly occur because starch intramolecular hydrogen bonds are formed rather than intermolecular hydrogen bonds [42].

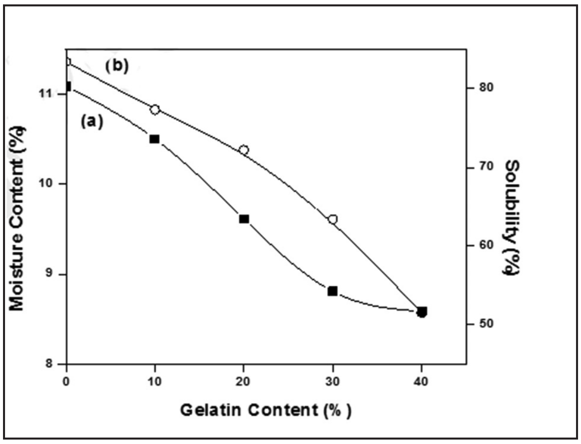

Figure 3: Effect of gelatin contents on (a) moisture content and (b) film solubility.

Table 2: Thermal transition parameters of films.

Film	$T_{initial}$ (°C)	T_{final} (°C)
Pure PS	150.18	390.94
PS/gelatin (30%)	112.08	339.56
Pure gelatin	141.22	419.14

PS: Potato starch

3.4. Thermal Properties

The properties of blend films with various levels of gelatin were also analyzed in terms of thermal performance using TGA. Thermal transition parameters of films are shown in Table 2. The TGA curves of pure PS, pure gelatin and 30% of PS/gelatin blend films are depicted in Figure 5. A three-step weight loss mechanism was observed in TG of pure PS. The initial weight loss at 50-180°C is due to the loss of volatile components including water and propylene glycol. The second step in the approximate range of 250-350°C is attributed to the pyrolysis of starch. There are two significant weight losses appeared TG curves of pure gelatin films. The first weight loss at 50-180°C is due to the vaporization of water absorbed in the sample, while the other one at 240-460°C was attributed to the thermal degradation of gelatin molecules. In the case of PS/30% gelatin blend films, three-step weight loss mechanism was observed, attributed to the loss of volatile compounds,

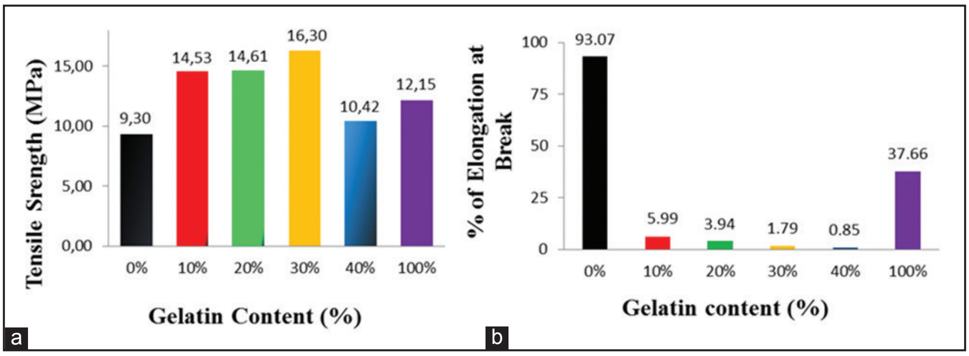

Figure 4: Effect of gelatin concentration on, (a) tensile strength, and (b) elongation at break of potato starch based films.

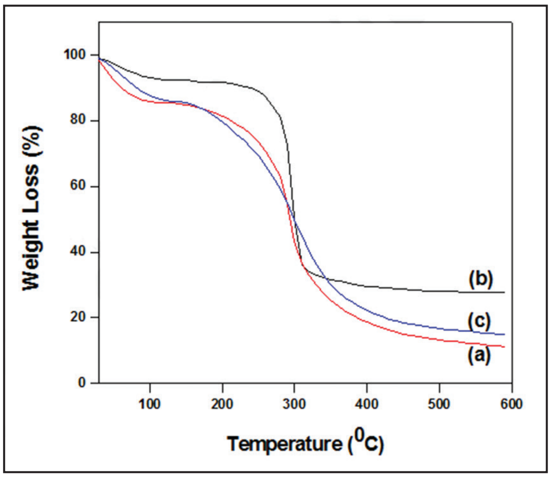

Figure 5: The thermo gravimetric analyses curves of potato starch (PS), gelatin and PS/gelatin blend films. (a) Potato starch film, (b) the film with 30% gelatin, and (c) gelatin film.

pyrolysis of PS and pyrolysis of gelatin, respectively. This might be a result of the formation of a strong interaction between the molecules of PS and gelatin.

4. CONCLUSION

The PS/gelatin green blend films were successfully prepared by casting method. The ternary blend containing 30% gelatin content resulted in films with high TS when compared to other films. The elongation at break, moisture, and water solubility of the blend films increased gradually with increasing gelatin content. These results indicated that the addition of gelatin could improve the mechanical properties of the pure PS film and change the water resistance of pure PS film. Further, the thermal stability of PS/gelatin blend films was found to increase on gelatin content. The results TGA indicated a good compatibility between the molecules of PS and gelatin at the 30% of PS/gelatin blend films. This might be because of the intense interaction between PS and gelatin at the PS/gelatin blend films and SEM micrographs visibly confirmed the homogeneous structure of PS-gelatin blended films. The approaches used in this study enabled a mechanistic understanding of the effects of gelatin on the structure and function of PS blended films.

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