

Production of Thermoplastic Starch-Aloe Vera Gel Film Embedded with Polyethylene for Improved Tensile Strength and Water Absorption

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ABSTRACT

The water absorption characteristics of thermoplastic starch (TPS) reveal its ability to interact with and absorb water, influencing its overall performance in various applications. The introduction of aloe vera (AV) gel into the TPS polymeric matrix film has proven beneficial in enhancing tensile strength but a notable deficiency is observed in lowering water absorption percentage. Therefore, this paper aims to reduce the water absorption of the TPS30AV polymer matrix film by utilizing polyethylene (PE) without reducing the tensile strength performance of the TPS30AV film. The TPS30AV film was produced using melt-blending and hot-press techniques. Six samples were presented consisting of a control film (TPS30AV) and TPS30AV film with different concentrations of PE resins (5%, 10%, 20%, 30%, and 40%). The film's performance was assessed by examining its mechanical properties, water absorption, and water solubility. The finding shows that TPS30AV with 40% PE has the highest improvement in water absorption percentage which reduces from 309.86% to 34.16% but a tremendous decrement in tensile strength performance. However, TPS30AV with 10% PE has the highest tensile strength, 9.65 MPa with a 20.99% improvement in water absorption reduction. The properties obtained for TPS30AV10PE were also good in

Young's modulus and water solubility percentage. It shows that the film formulated is comparable with previous studies, and improvement has been achieved. As a conclusion, adding PE improved the polymeric structure of TPS30AV and can be used as a biodegradable food-packaging film.

Keywords: *Thermoplastic Starch; Aloe Vera; Polyethylene; Thin Film; Polymer Blend, Tensile Strength*

Introduction

In Malaysia, the challenge of plastic waste involves the widespread use of single-use plastics, insufficient waste management infrastructure, and issues associated with the illicit import of plastic [1]. The short lifespan of plastic products such as single-use plastics contributes to a significant volume of plastic waste generated [2]. Olefin such as polyethylene (PE), is obtained from fossil fuels commonly used in the production of single-use plastics. It is the main raw material used which is resistant to harmful environments causing poor degradation performance and leading to plastic waste accumulation. Thus, 30 years ago, researchers started blending PE with a biopolymer that has good degradation ability for the goal of having a film plastic that degraded faster compared to petroleum-based plastic [3].

Starch, a biopolymer, shows great potential as a substitute for petroleum-based plastics. This is because starch possesses the desirable qualities of being fully biodegradable and readily available in large quantities in nature [4]. Thermoplastic starch (TPS), a type of bioplastic made from starch, is widely utilized in packaging materials. Starch mixture with plasticizer produced TPS is a promising material because of its abundance of availability, eco-friendly, and biocompatible [5]-[8]. Unfortunately, cellulose and hemicellulose in TPS absorb moisture in the environment and cause strength reduction of the polymer [9]. Previous researchers tried improving the TPS properties by adding filler or biopolymer instead of combining them with PE. Aloe vera (AV), sugarcane, microalgae, ahpa peels baggase, and polylactic acid are among the fillers or additives being used for bioplastic applications [10]-[14]. The incorporation of these materials causes an improvement in the mechanical and barrier properties of TPS. Unfortunately, a previous study found AV addition into TPS caused a high water absorption percentage which restricts TPS usage in applications [10].

Blending TPS with PE would help in reducing the water absorption percentage of TPS due to the hydrophobic character of PE [15]-[16]. Plus, adding PE causes the mechanical properties of TPS/PE blends to improve, making them appropriate for sustainable applications and expanding the scope of TPS's potential applications [17]. Researchers tried various ranges of PE ratios which between 60% to 95% of PE in the TPS/PE blend. Some

papers mentioned that TPS's addition to low-density PE (LDPE) caused a decrement in tensile strength [18]-[19]. Previously, the addition of 60% TPS to PE reduced tensile strength by 50% [20]. Flexibility loss was also experienced by linear low-density PE/TPS/green tea film, in which the tensile strength and elongation at break were reduced by 62.2% - 87.3% and 31.2% - 93.5%, respectively [16]. The loss was due to poor hydrogen bonding formation in the polymer blend. However, contradicting results obtained by others, the tensile strength of linear low-density PE increased with TPS [21]. The literature suggests a more significant proportion of PE is still required to maintain the mechanical performance of the polymer mix.

Thus, a specific percentage of PE was needed to reduce the water absorption percentage of the TPS film with the addition of filler or biopolymer. In this paper, TPS with AV gel was used as the control film to determine the appropriate range of PE suitable in the TPS/AV blend. The range of PE added in this study was set between 5% and 40% because starch with 30% AV in 70% PE resulted in poor interfacial adhesion through starch accumulation [22]. PE was predicted to reduce the film's water absorption capacity while maintaining or improving its mechanical qualities of TPS with 30% AV film because PE has a hydrophobic characteristic (water resistant) and is insoluble in water [23]. Therefore, this work further investigated the inclusion of PE at a lower range ratio to find the optimal PE concentration for TPS with 30% AV.

Materials and Methodology

Materials

The powdered potato soluble starch as the primary polymer-based film brand Bendosen was obtained from Laupik Chemical, glycerol acted as plasticizer from Chemiz, Malaysia, AV gel was bought from Chemieconnex, Malaysia, and PE resin from Lotte Chemical, Titans, Malaysia.

Film preparation

Modified melt-blending and hot-press techniques were used in preparing the film [10]. Figure 1 shows the film preparation procedure started with the starch and PE was dried in an oven at 70 °C for 24 hours to remove moisture. The starch and glycerol were mixed at a 70:30 ratio to produce TPS. The melt-blending process involved AV gel and PE resin added to an internal mixer (Haake Polylab OS RheoDrive7, Thermo Scientific) that was thermally heated with TPS to form a solid resin. The internal mixer operated at 170 °C, 60 rpm, for 30 minutes. The solid resin produced from the internal mixer was then crushed to form smaller resin using an HMRV50-19, Rexmac compact crusher before being subjected to a QC-602A, Cometech hot-press machine to form a thin film. The thin film produced was then stored in a desiccator to

prevent direct contact with the moisture before further characterization was conducted. Six samples consisting of TPS with 30% AV (control film) and a combination of TPS with 30% AV film with different PE percentages were prepared for characterization as presented in Table 1.

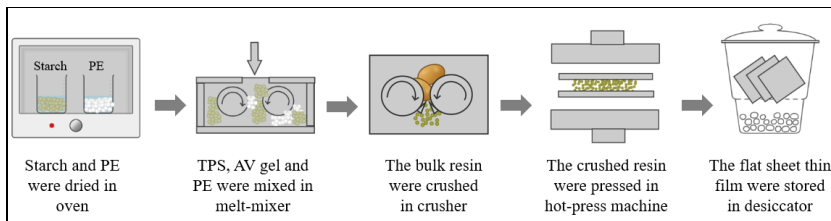


Figure 1: Film preparation procedure

Table 1: Film formulation

Sample Name	TPSAV (g)	PE (g)
TPS30AV	30	0.0
TPS30AV5PE	30	1.5 (5% PE addition)
TPS30AV10PE	30	3.0 (10% PE addition)
TPS30AV20PE	30	6.0 (20% PE addition)
TPS30AV30PE	30	9.0 (30% PE addition)
TPS30AV40PE	30	12.0 (40% PE addition)

Tensile strength, elongation at break, and Young's modulus

Tinius Olsen H50KT, Instron 3382 universal testing equipment, was used to determine the film's mechanical properties, which covered tensile strength, elongation at break, and Young's modulus. The mechanical properties of polymeric films demonstrate the film's suitability for various applications [24]. The samples were prepared and tested following ASTM D882-02, the standard test method for thin-film sheeting tensile properties. The strip specimen measured 100 mm in length and 25 mm in width. The test specimens were chosen for consistent thickness within 5% of the film's thickness between 0.25 mm and 1.00 mm. The films were strained at 25 mm/min at room temperature with 50% relative humidity. Five repeated data points were collected because the film is an isotropic material, and its properties are identical in all directions. The minimum tensile strength required for the produced film was 7.78 MPa [25], representing the tensile strength of pure PE film.

Water absorption and water solubility

Water absorption analysis is required to predict the stability and quality of packaging materials during the loading, transporting, and storing of various

foodstuffs [26]. The extent of water absorption into the polymeric matrix depends on the polymer's chemical structure and shape. Water absorption and whether diffusion takes place on the surface are also affected by the volume fraction and condition of the polymer. Solubility in water was measured by the amount of the film's dry matter dissolved after 24 hours in water [27]. The method applied was referred to as ASTM D570-63. The samples were cut at 50.8×50.8 mm for a thickness of less than 0.5 mm. Samples were dried for 24 hours to remove the physisorbed water in a universal oven (ULE 600, Memmert) at 50 °C. After drying, the first weight was measured (W_0). The dried samples underwent 24 hours of immersion in a beaker holding 250 mL of distilled water. Then, the samples were drained, dried using a paper towel, and weighed (W_1). Next, the sample was dried in the same oven with the same procedure. Then, the samples were weighed again (W_2). Equation (1) was used to determine the value of water absorption [21], and the water solubility percentage was calculated by using Equation (2) [28].

$$WA (\%) = \frac{w_1 - w_2}{w_0} \times 100 \quad (1)$$

$$WS (\%) = \frac{w_0 - w_2}{w_0} \times 100 \quad (2)$$

Results And Discussion

Mechanical properties of the films

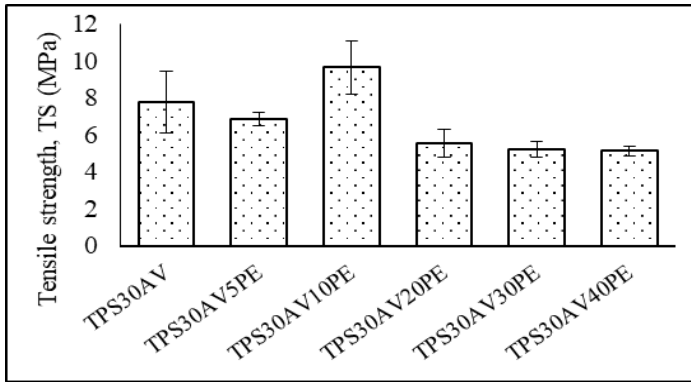
Figure 2(a) shows the tensile strength of TPS30AV film added with different percentages of PE (5% to 40%). Adding 10% PE increased the tensile strength of TPS30AV by 24%, from 7.78 MPa to 9.65 MPa. Previously, the modification of starch enhanced the tensile strength of the LDPE/starch blend even at a higher ratio of modified-starch content in the LDPE/starch blend [29]. Chemical bonding between 10% PE and TPS30AV might occur by hydrogen abstraction reactions. Heat applied during the melt-blending process generated free radicals in the PE molecule and TPS30AV, grafting between PE and TPS30AV simultaneously, increasing the tensile strength performance. This agrees with a previous study that found 3.5% grafting formation in an internal mixer between 50% LDPE and 50% silicone rubber due to C-H cleavage that creates macroradicals in LDPE [30]. Free radicals occurred in TPS30AV films even without an initiator because of reactive functional groups such as the carboxyl group at leucine and anhydride group at acemannan chemical structure. No external assistance from an initiator or a catalyst is needed to allow free radicals if the starting material consists of two reactive functional groups [31].

However, when the proportion of PE increased, the tensile strength decreased to 5.56 MPa, 5.24 MPa, and 5.15 MPa for 20%, 30%, and 40% PE, respectively. These two materials repel one another because of their polarities and surface energies. The hydrophobic nature of PE and the hydrophilic nature of TPS30AV led to a decrement in tensile strength at a higher PE ratio due to incompatibility. Strong hydrogen bonds between TPS and 30% AV led to the accumulation of TPS30AV on the PE matrix and decreased tensile strength when 20% to 40% PE was added, consequently decreasing the compatibility between TPS30AV and PE. This decrease in tensile strength is significantly influenced by the interfacial tension caused by the hydrophobic PE and hydrophilic starch [29]. Other studies obtained a lower tensile strength at a higher starch concentration in LDPE film due to the non-homogeneity of the starch and LDPE, which caused the formation of pores and voids [32]. This is consistent with the literature demonstrating that high LDPE starch content has minimal interfacial adhesion [33]. Incompatibility between PE and TPS leads to the aggregation of TPS particles and creates a microcrack initiation region [17].

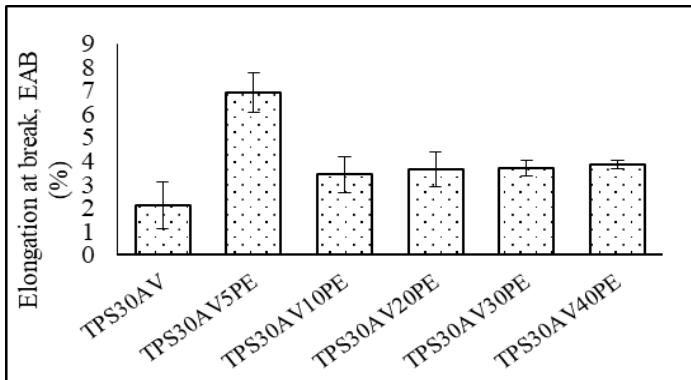
Figure 2(b) shows the elongation at the break of the TPS30AV film with the addition of different percentages of PE. Elongation at break is a determinant factor for interpreting the rupture performance of the films [34]. The film with 5% PE had the highest elongation at break at 6.91%, while other films with 10% - 40% PE addition showed no significant changes. TPS30AV5PE recorded the highest elongation at the break due to free electrons trapped on the film's surface. The electrostatic repulsion resulting from 5% PE addition on TPS30AV film might hinder adhesion between the two surfaces simultaneously, increasing the elongation at break, while no electrostatic repulsion occurred at 10% - 40% PE addition. Panrong et al. stated that poor interfacial adhesion between materials in the polymer blend contributes to the high elongation at break [35]. However, at 10% - 40% PE, there is no significant effect experienced by TPS30AV, which suggests that the two components are immiscible and have incompatible secondary phases. This can result in phase separation, where the TPS30AV and PE form distinct phases within the blend. A previous study shows a similar trend, with nanoclay's reinforcing and stiffening effect reducing polymer chain mobility because it builds up a network [34]. Other studies have also demonstrated a decrement in elongation at break with the addition of TPS to the PE matrix due to the lack of interfacial adhesion between PE and TPS [33], [36]. Even 10% PE addition resulted in the highest tensile strength, it did not result in the highest elongation at break. Tensile strength and elongation at break represent different aspects of a material's performance under tension and cannot be used interchangeably.

An increase in Young's modulus indicates that the stiffness of the films improved [34]. Young's modulus provides information on the film's ability to lengthen during stretched and shrink when compressed. A high Young's

modulus material can withstand more initial force, repeated pressure, and release without deforming. As shown in Figure 2(c), the highest Young's modulus was obtained by TPS30AV10PE (513 MPa), comparable to Young's modulus of TPS30AV. The 10% PE restricted the mobility and deformation of the matrix polymer. Young's modulus decreased at 5% PE and consistently decreased as the PE content was increased to 20% or higher. The non-homogeneous dispersion of TPS30AV with the addition of PE caused incompatibility of polymer structure, thus reducing Young's modulus. Previously, starch particle aggregation in LDPE/TPS contributed to a lower Young's modulus [17]. Similarly, the incompatibility of date seed extract, ethylenediaminetetraacetic acid (EDTA), and LDPE caused a reduction in Young's modulus [34]. Young's modulus decrement is also caused by the softening effect of the compatibilizer [37].



(a)



(b)

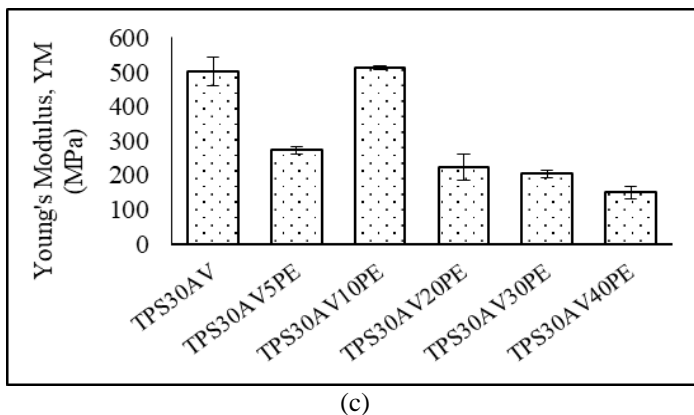


Figure 2: (a) Tensile strength, (b) elongation at break, and (c) Young's modulus of TPS30AV film at different percentages of PE. Data presented as mean \pm SD ($n = 3$). Different alphabets between columns represent significance at $p < 0.05$ after applying post hoc Tukey's test

Water absorption and water solubility of the films

Figure 3 depicts the water absorption percentage of TPS30AV with different percentages of PE. Adding PE reduces the water absorption percentage and increases the film application range [17]. The trend of the water absorption illustrates that the percentage dropped from 309.61% to 34.16%, with 40% PE for TPS30AV film. However, a slight increase in water absorption percentage was observed at 5% PE, with around a 40% increase compared to the TPS30AV film water absorption percentage. Less exposed hydrophilic TPS30AV film surface area led to a lower water absorption percentage that reduced the formation of more fine pores, including the increment of lumps (agglomeration). The presence of PE in TPS30AV film was expected to reduce the water absorption due to the hydrophobic nature of PE. The higher hydrophobicity delays the moisture transition into the composite. This is consistent with a previous finding, which observed LDPE as a hydrophobic polymer that restricted water penetration into the TPS matrix [17]. Others also stated that the presence of hydrophobic hemiacetal groups produced due to crosslinking might reduce the water absorption percentage [38]. Gray et al. [39] stated that the presence of cellulose nanocrystals as a crosslinker between LDPE and TPS significantly reduced moisture absorption, which was attributed to the lower mobility of starch chains due to the higher crystallinity of cellulose nanocrystals.

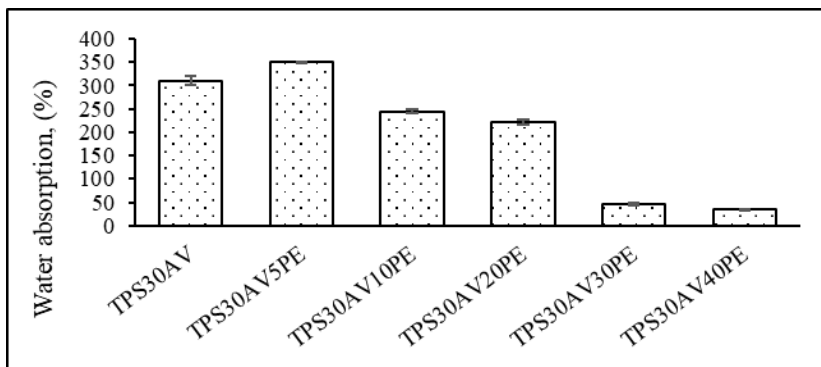


Figure 3: The percentage of water absorption of TPS30AV film at different percentages of PE. Data presented as mean \pm SD (n = 3). Different alphabets between columns represent significance at $p < 0.05$ after applying post hoc Tukey's test

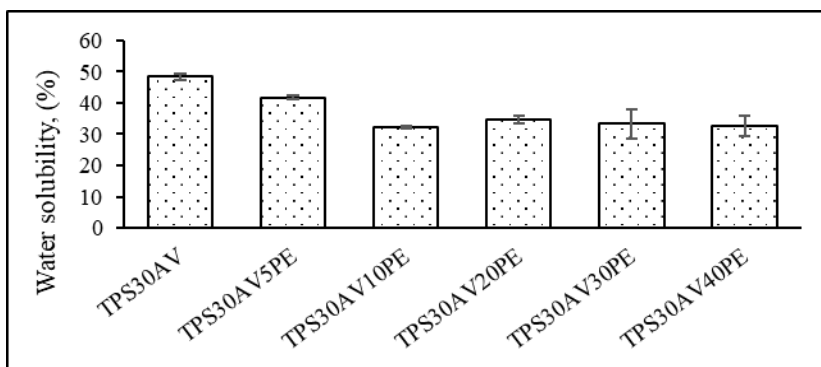


Figure 4: The percentage of water solubility of TPS30AV film at different percentages of PE. Data presented as mean \pm SD (n = 3). Different alphabets between columns represent significance at $p < 0.05$ after applying post hoc Tukey's test

Figure 4 shows the water solubility percentage of TPS30AV film with different percentages of PE. The result demonstrates some decrement in water solubility percentage from 48.46% to 32.39% with the addition of 10% PE. The value is almost consistent for 20% - 40% PE added into TPS30AV film. These findings are consistent with the literature: PE is water-repellent, keeping moisture out and preventing it from contacting TPS [17]. The reaction between TPS30AV and PE reduced the interfacial tension due to

irreversible covalent bonds between hydroxyl groups of TPS30AV and a polar group of PE. More agglomerated TPS30AV adheres on the film surface, reducing the number of O–H groups available on the surface of the polymer matrix, thus causing low interaction with the water molecules. Even though poor compatibility was obtained between TPS30AV with a higher percentage of PE, the water migration was still poor, presumably due to the coverage of more PE components on the surface structure of the polymer matrix, which gradually changed the surface from hydrophilic to hydrophobic. The interfaces enhance the cohesive properties of the polymer matrix and cause water molecules not to break the strong bonds [40]. The hydrogen bonding in a polymer matrix hinders water diffusion through the bulk, reducing the water solubility [41].

Conclusion

In conclusion, this study revealed addition of PE into TPS30AV reduced the water absorption percentage of TPS30AV film, aligning with PE characteristics which is hydrophobic. In addition, the best formulation decided was TPS30AV10PE due to having the highest tensile strength, and yet water absorption was also reduced. The significant findings is the tensile strength improvement proved that interaction occurred between TPS30AV and 10% PE which caused significant improvement in lower water absorption and water solubility percentage. The outcome suggested 10% PE addition improved the TPS30AV film's ability to be stored at room temperature. This research offers valuable insights into producing a bioplastic made from a TPS/PE blend with a low percentage of PE which is good for the environment. In addition, the best formulation decided was TPS30AV10PE due to having the highest tensile strength, and yet water absorption was also reduced.

Contributions of Authors

The authors confirm the equal contribution in each part of this work. All authors reviewed and approved the final version of this work.

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Conflict of Interests

All authors declare that they have no conflicts of interest.

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