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The Structural and Mechanical Properties of The Tamarind Seed Polysaccharide Plasticized with Deep Eutectic Solvent for Food Packaging

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ABSTRACT. Tamarind seed polysaccharides (TSP) exhibit significant potential as bioplastic films for food packaging, attributed to their biodegradability, renewability, cost-effectiveness, and biocompatibility. Nonetheless, the brittleness of TSP film, attributed to hydrogen bonding between the chains, will hinder its application. In this study, a deep eutectic solvent (DES) consisting of choline chloride and ethylene glycol was integrated into the TSP matrix at concentrations of 0.4 and 0.8 wt%, and the films were fabricated using the solvent casting technique. The structural and mechanical properties of the DES-plasticized TSP films were examined using Fourier transform infrared spectroscopy (FTIR) and tensile testing, respectively. The results indicated that the DES effectively enhanced the flexibility of TSP films by elevating the tensile strain value. The peak displacement in FTIR analysis demonstrated that TSP-DES interactions have markedly diminished the formation of hydrogen bonds among TSP chains. The ideal concentration of DES was 0.4 wt%, as this sample exhibited the maximum tensile strain of 10.35%. These findings underscore the potential of DES as an eco-friendly plasticizer for bioplastic films, warranting additional research on its thermal properties.

Key words: Polysaccharides, deep eutectic solvent, food packaging, bioplastic, flexible film

1. INTRODUCTION

Food packaging is an integral component of food processing, vital for the protection, preservation, and convenience of products. Petrochemical plastics are predominantly utilized in food packaging industries due to their low cost, excellent tensile properties, and effective barrier capabilities against oxygen, carbon dioxide, and water vapor (Ncube et al., 2020). Plastics frequently utilized for food packaging comprise thermoplastics, including low-density polyethylene (LDPE), high-density polyethylene (HDPE), polyvinyl chloride (PVC), polyethylene terephthalate (PET), polystyrene (PS), and polypropylene (PP) (Ferreira et al., 2016).

Annually, approximately 400 million tons of plastics are manufactured worldwide, with nearly 46% originating from the packaging industry (Wei et al., 2023). Greenhouse gas emissions associated with plastics are projected to surpass 650 million tons per year by 2050, with approximately seven billion tons of plastics produced from 1950 to 2017 ultimately becoming waste (Fayshal, 2024). Additionally, plastic products exacerbate numerous environmental and health concerns. Some plastics release various toxic substances, including Bisphenol A (BPA). Certain studies indicate that plastics may release BPA molecules when beverage and food containers are utilized for extended periods.

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Women exposed to BPA exhibit compromised health conditions, including polycystic ovarian syndrome, obesity, recurrent miscarriages, endometrial hyperplasia, and infertility. The exposure of children and women of reproductive age to elevated concentrations of BPA constitutes a significant public health concern due to the heightened susceptibility of children and developing fetuses to BPA compared to adults exposed to similar levels (Lin et al., 2017). Plastic consumption can also lethally affect marine mammals through ingestion, as they may confuse it with food.

Alternatively, to address these challenges, plastics derived from bio-based polymers are extensively researched. This study concentrated on the fabrication of bioplastic film utilizing tamarind seed polysaccharides (TSP) owing to their biodegradability, renewability, cost-effectiveness, and biocompatibility for food packaging applications. Nonetheless, a challenge encountered in this process is the brittleness of the TSP film, attributed to hydrogen bonding among polymer chains (Fauzee et al., 2024). Numerous prior studies employed polymer blending techniques utilizing TSP alongside other polymers, including berry seed starch (Chowdhury et al., 2022) and poly (butylene adipate-co-terephthalate) (PBAT) (Preethi et al., 2024), to mitigate the brittleness of TSP. Nonetheless, polymer blending may result in an inhomogeneous mixture.

Previously, deep eutectic solvents (DESs) such as choline chloride/gallic acid, choline chloride/glycerol, and choline chloride/anhydrous oxalic acid have been utilized as plasticizers in bioplastic films due to the inadequate mechanical properties of natural polymers including chitosan (Rolińska et al., 2024), carrageenan (Ramli et al., 2024), and lignocellulose-bamboo leaf powder (Liu et al., 2024a), respectively. The integration of DES into the natural polymer matrix has effectively enhanced the flexibility and elasticity of the bioplastic films. The inclusion of DES has diminished the crystallinity of the films by disturbing the internal structural order of the bioplastics. In this study, flexible bioplastic film was produced by incorporating varying weight percentages of deep eutectic solvent (DES), specifically 0.4 and 0.8 wt%, composed of choline chloride and ethylene glycol, to function as a plasticizer by filling the interstitial spaces between TSP chains and inhibiting hydrogen bonding formation. The structural and mechanical properties of the DES-plasticized TSP films were examined.

2. METHODOLOGY

2.1. Preparation of the DES- Plasticized TSP Films

The solvent casting method, derived from the research conducted by Fauzee et al. (2024), was employed to fabricate the film samples. Initially, 2.0000 g of TSP was dissolved in 100 mL of distilled water at 80 °C using a magnetic stirrer for 10 minutes. The TSP solution was deposited into the petri dish and subsequently dried in a hot air oven at 60 °C for 24 hours until films were formed. The films were preserved in a desiccator to enable peeling prior to being sectioned into multiple pieces for additional characterization. The identical procedures were reiterated with the incorporation of DES for the fabrication of DES-plasticized TSP films. Table 1 enumerates the composition of the DES-plasticized TSP films.

Table 1. Composition of the DES-plasticized TSP films

Mass of TSP (g)	Weight percentage of DES (wt%)	Mass of DES (g)	Sample designations
2.000	0	0.0000	TSP
	0.4	0.0080	TSPDES0.4
	0.8	0.0161	TSPDES0.8

2.2. Characterization Methods

2.2.1. Fourier Transform Infrared Spectroscopy (FTIR)

The spectra of the film samples were acquired at ambient temperature utilizing FTIR (Thermo Fisher Scientific Nicolet iS 10) with Attenuated Total Reflectance (ATR). The objective of the FTIR analysis was to determine the interaction between TSP and DES. Prior to conducting transmittance mode measurements at a resolution of 2 cm^{-1} with 16 scans across the frequency range of 4000 cm^{-1} to 600 cm^{-1}, the samples were positioned directly onto the crystal.

2.2.2. Tensile Test

The mechanical properties of the film samples, including tensile strain and tensile stress, were assessed using the Instron Universal Testing Instrument (3365, Instron USA). In compliance with ASTM D882, each specimen was sectioned into 1.0 cm x 7.0 cm segments and assessed at a crosshead speed of 25 cm/min. Each sample formulation was measured in triplicate.

3. RESULTS AND DISCUSSION

3.1. Formation of DES-Plasticized TSP films.

Figure 1 illustrates the films of TSP and DES-plasticized TSP films (specifically, TSPDES0.4 and TSPDES0.8). The formation of DES-plasticized TSP films was examined and analyzed. Figure 1(a) indicates that the TSP film is transparent, has a rough surface, and displays brittleness. The observed brittleness of the TSP film may result from interchain crosslinking through hydrogen bonding, potentially causing diminished flexibility and heightened rigidity in the polymer matrix. Fauzee et al. (2024) similarly documented this observation regarding TSP film. The incorporation of 0.4 and 0.8 wt% of DES produced transparent films exhibiting enhanced elasticity and smoother surfaces (Figure 1(b) - (c)). This may result from DES's capacity to infiltrate the interstitial spaces of polysaccharide chains, thereby inhibiting hydrogen bonding formation. This observation is further corroborated by the study conducted by Nardecchia et al. (2012), which reported the enhancement of brittle polypeptides following the incorporation of DES.

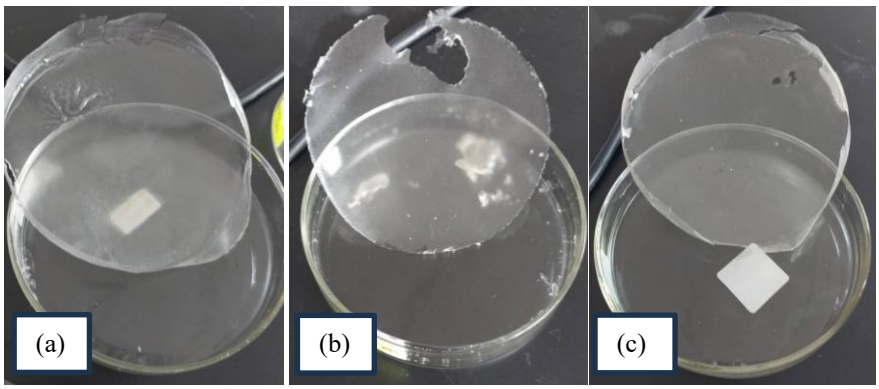

Figure 1. The films for (a) pure TSP, (b) TSP0.4DES, (c) TSP0.8DES

3.2. Structural Studies of DES-plasticized TSP films.

Figure 2 depicts the FTIR spectra of TSP and DES-plasticized TSP films. Concurrently, the significant peaks are enumerated in Table 2. The FTIR peaks for TSP were observed at 3343, 2893, 1645, 1365, 1008, 942, and 895 cm^{-1}, corresponding to OH stretching, CH_2 stretching, C=O stretching, CH_2 bending, C-O-C stretching, C-H bending, and CH stretching, respectively.

The introduction of DES resulted in peaks at 663 cm^{-1}, indicative of CH stretching. This signifies the successful integration of DES into the TSP matrix. The OH stretching peak shifted to lower wavenumbers from 3343 cm^{-1} to 3300 cm^{-1} and 3292 cm^{-1} upon the addition of DES in samples TSPDES0.4 and TSPDES0.8, respectively. The alteration in peak intensity was likewise noted. The transition to higher wavenumbers was also noted for the C-O-C stretching peak, shifting from 1008 cm^{-1} to 1012 cm^{-1} (TSPDES0.4) and 1016 cm^{-1} (TSPDES0.8) upon the addition of DES, accompanied by alterations in peak intensities. These observations suggest the interaction between TSP and DES. The interaction among the constituents in the materials can be validated through the wavenumber shift and variations in peak intensity of the FTIR spectra (Pasieczna-Patkowska et al., 2025). Pavia et al. (2008) reported that energy is directly proportional to the wavenumber. Consequently, a shift of the band to a higher wavenumber indicates a more robust interaction, while a shift to a lower wavenumber implies a diminished interaction. Fauzee et al. (2024) similarly reported the effect of ChCl/1,4-butanediol DES on TSP. The FTIR analysis substantiates the TSP-DES interaction, anticipated to inhibit hydrogen bonding among TSP chains and diminish the brittleness of the bioplastic film.

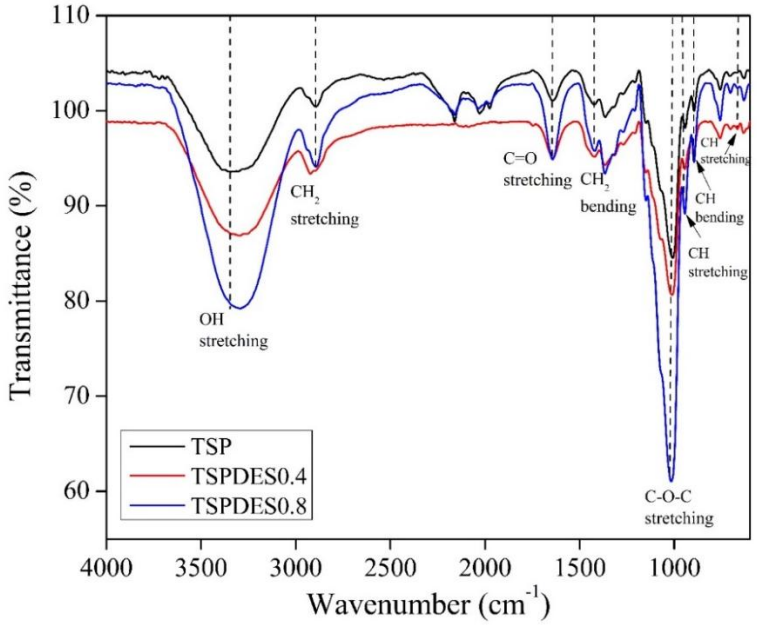

Figure 2. FTIR spectra of TSP and DES- plasticized TSP films

Table 2. FTIR important peaks of TSP and DES-plasticized TSP films

TSP	TSP DES 0.4	TSP DES 0.8	Spectral band assignment	References
3343	3300	3292	OH stretching	Fauzee et al. (2024)
2893	2922	2892	CH_2 stretching	Fauzee et al. (2024)
1645	1644	1643	C=O stretching	Shao et al. (2019)
1365	1366	1366	CH_2 bending	Fauzee et al. (2024)
1008	1012	1016	C-O-C stretching	Chawananorasest et al. (2016)
942	944	944	C-H bending	Fauzee et al. (2024)
895	895	895	CH stretching	Liu et al. (2024b)
-	663	663	CH stretching	Ren et al. (2016)

3.3. Mechanical Studies of DES-plasticized TSP films.

Before conducting mechanical studies, the film thickness was measured. Table 3 enumerates the thickness of TSP and DES-plasticized TSP films.

Table 3. Thickness (mm) of TSP and DES-plasticized films

Sample File	Thickness (mm)
TSP	0.246± 0.0896
TSP0.4DES	0.150 ± 0.0163
TSP0.8DES	0.130 ±0.0216

The thickness of TSP is 0.246 mm ± 0.0896 mm. Upon the addition of 0.4 and 0.8 wt% of DES, the thickness diminished to 0.150 ± 0.0163 mm and 0.130 ± 0.0216 mm, respectively. This results from increased polymer chain mobility, a plasticizing effect, and diminished hydrogen bonding, which facilitate a more compact structure, as noted by Murthy (2006).

Tensile strain is defined as the relative elongation of a specimen subjected to a tensile force (Qin, 2015). Figure 3 illustrates the graph of tensile strain versus weight percentage of DES. The tensile strain of TSP is 1.83%. Upon the addition of 0.4 wt% of DES, the tensile strain elevated to 10.35%. This signifies enhanced flexibility of the film. This may be attributed to the successful occupation of DES among the polysaccharide chains, thereby inhibiting the formation of hydrogen bonds. The TSP-DES interaction is further corroborated by structural studies utilizing FTIR. Fauzee et al. (2024) similarly reported this observation when 0.4 wt% of ChCl/1,4-butanediol DES was incorporated into the TSP matrix. Conversely, the addition of 0.8 wt% of DES resulted in a decrease in tensile strain to 3.12%, signifying a reduction in the film's flexibility. This may result from an accumulation of excess DES particles. Shamsuri and Daik (2012) documented a similar finding for the agarose/ChCl-urea system at 70 wt% of ChCl/Urea.

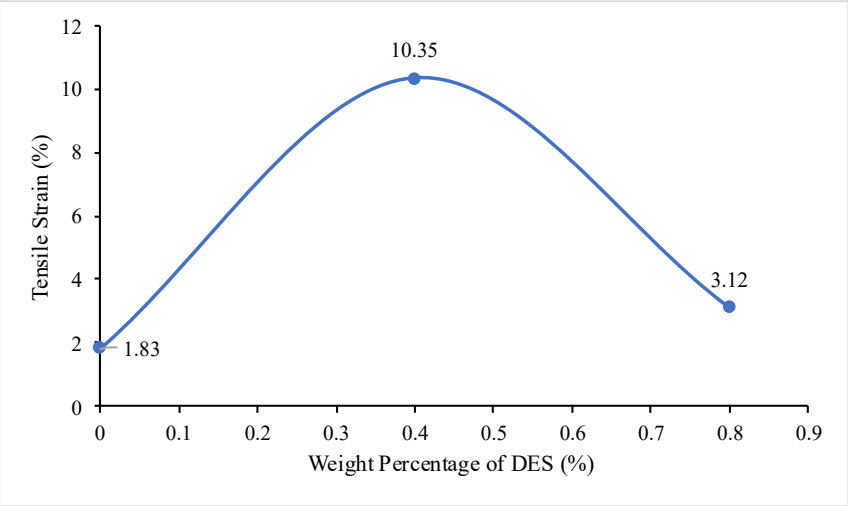

Figure 3. The graph of tensile strain against wt% of DES

Tensile stress is defined as the ratio of a stretching force applied to a material to the material's cross-sectional area (Gdoutos & Konsta-Gdoutos, 2024). Figure 4 depicts the graph of tensile stress versus weight percentage of DES. The tensile stress is expected to exhibit an inverse relationship with tensile strain. The tensile stress of TSP measures 13.55 MPa. Upon the addition of 0.4 wt% of DES, the tensile stress diminishes to 12.27 MPa. This may be attributed to the reduction in the material's resistance to deformation, leading to diminished stiffness of the sample. Upon the addition of 0.8 wt% of DES, the tensile stress diminishes to 7.03 MPa, despite a reduction in flexibility. The presence of excessive DES particles in TSPDES0.8 may result in reduced film stiffness. Fauzee et al. (2024) made a similar observation when 0.6 wt% of ChCl/1,4-butanediol was incorporated into the TSP matrix.

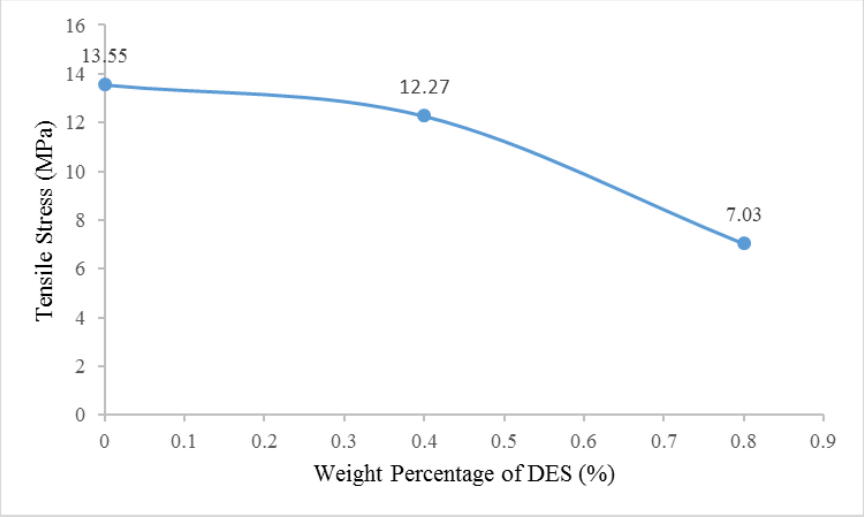

Figure 4. The graph of tensile stress against wt% of DES

4. CONCLUSION

This study successfully prepared flexible films by incorporating 0.4 and 0.8 wt% of DES into the TSP matrix. This may be attributed to the DES that occupied the interstitial space between the TSP chains, inhibiting the formation of hydrogen bonds. This can be further substantiated by the TSP-DES interaction as evidenced by FTIR analyses. The optimal DES utilized as an effective plasticizer was 0.4 wt%, as the film exhibited the greatest flexibility, evidenced by its maximum tensile strain of 10.35%. The reduction in flexibility upon the addition of 0.8 wt% of DES is attributable to the excessive presence of DES particles. The results indicate the potential of DES as an eco-friendly plasticizer for bioplastic films. It is recommended to examine the thermal properties of the film to further explore the potential of these bioplastic films at elevated temperatures.

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AUTHOR CONTRIBUTIONS

Nur Syamimi Azman, Nurdamia Farhana, Nurnajihah Norrizam and Nurul Adlina Ridzuan are responsible for data collection and analysis as well as writing the original draft. Nabilah Akemal is responsible for supervising, administrating the project and editing the final manuscript. Atikah Wan Nafi and Faiezah Hashim is responsible for editing the final manuscript.

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COMPETING INTEREST

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

COMPLIANCE OF ETHICAL STANDARDS

Not applicable.

SUPPLEMENTARY MATERIAL

Not available.

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