Journal of Current Science and Technology, September - December 2023 Vol. 13 No. 3, pp. 630-641

ISSN 2630-0656 (Online)

Cite this article: Suwanamornlert, P., Taksima, T., & Thanyacharoen-anukul, P. (2023). Effects of plasticizers on characterization of biodegradable film based on tamarind kernel polysaccharide. *Journal of Current Science and Technology*, *13*(3), 630-641. https://doi.org/10.59796/jcst.V13N3.2023.814

Journal of Current Science and Technology

Journal homepage: https://jcst.rsu.ac.th

Effects of Plasticizers on Characterization of Biodegradable Film Based on Tamarind Kernel Polysaccharide

Panitee Suwanamornlert*, Takunrat Taksima and Pongsura Thanyacharoen-anukul

College of Integrative Medicine, Dhurakij Pundit University, Bangkok 10210, Thailand

*Corresponding author; E-mail: panitee.tip@dpu.ac.th

Received 20 March, 2023; Revised 22 April, 2023; Accepted 13 June, 2023;
Published online 30 August, 2023

Abstract

Tamarind kernel polysaccharide-based film was formulated to study the effects of polyethylene glycol 400, glycerol, sorbitol, tween 80, tween 40, tween 20 and span 20 on mechanical, optical, and thermal properties. Addition of plasticizers to tamarind kernel polysaccharide led to changes in tensile strength, elastic modulus, and elongation at break of the films. The tensile strength of films containing polyethylene glycol 400, glycerol, sorbitol, tween 20 and span 20 was lower than films containing tween 80, tween 40 and non-plasticized film. Sorbitol-plasticized film exhibited the best mechanical properties with lowest tensile strength, 6.07 MPa and highest elongation at break 5.91%. Film containing sorbitol also showed highest optical transparency, while polyethylene glycol 400 and glycerol exhibited lowest transparency and highest whiteness index. Differential scanning calorimetry (DSC) revealed that incorporation of plasticizers increased the mobility of the polymer chains. The addition of sorbitol, glycerol, and tween 20 reduced the glass transition temperature of tamarind kernel polysaccharide film from 49.81 to 20.97, 42.41 and 42.92 °C, respectively. Sorbitol proved to be an effective plasticizer for improving flexibility and enhancing the optical property of tamarind kernel polysaccharide film. As a result of the research, it was discovered that tamarind kernel polysaccharide film plasticized with sorbitol has the potential to be used in the creation of biopolymer film for culinary and biomedical applications.

Keywords**:** *biopolymer; flexibility; plasticization; tamarind kernel polysaccharide; thermal behavior; transparency*

1. Introduction

Bio-based polymers have recently emerged as a viable alternative to traditional petroleum-based polymers. These naturally available materials are derived from polysaccharides and have gained much attention in the development of food packaging and biomedical material due to their non-toxicity, film-forming ability, and biodegradability. Biopolymers obtained from polysaccharide, protein, lipid and other bio-materials are to be utilized to the food industry either in the form of films or coating materials (Gupta et al., 2022). The application of biofilms is intended to extend the shelf-life of food products by reducing moisture loss and gas exchange in fresh produces (Suresh et al., 2022). Reducing O_2 and increasing CO_2 delay respiration rate, oxidative reaction, and therefore extended shelf life. Biofilms may also serve as carriers of food additives such as antioxidants (Lin et al., 2023; Choi et al., 2023) and antimicrobial agents (Chou et al., 2023; Haghighatpanah et al., 2022) in food products such as fruit, chicken, meat, and pork. Tamarind kernel powder as an agricultural byproduct has evolved as a promising

novel raw material (Agarwal et al., 2020). Tamarind seed is an underutilized byproduct of the tamarind pulp industry. Decorticated tamarind kernel powder is considered as a promising biopolymer source due to its high availability, low cost, and desirable functional properties (Thivya et al., 2021). Tamarind kernel derived from tamarind seed (*Tamarindus indica*) is an appealing alternative heteropolysaccharide, possessing a backbone composed of 1,4-β-D-glucose with up to 70% of the glucose residues substituted with 1,6-α-D xylose. Some of these xylose groups may be attached to 1,2-β-D galactose residues (Kozioł et al., 2015). Tamarind kernel polysaccharide is water-soluble, non-ionic, hydrophilic, and branched in the form of a white to light yellow powder. It has recently been extensively studied for use as a promising biopolymer in the pharmaceutical, cosmetic and food industries (Malviya et al., 2021). Tamarind kernel polysaccharide has high viscosity, water binding capacity and film forming ability (Nagar et al., 2022), is commercially utilized as a food thickener, emulsifier, and stabilizer (Premalatha et al., 2016) and more recently in specific medical applications including drug delivery and in biomedical materials (Alpizar-Reyes et al., 2022; Qureshi et al., 2021).

Tamarind kernel polysaccharide is brittle and cracks during storage (Antoniou et al., 2014). Hence, polyethylene glycol, glycerol and sorbitol (Agarwal et al., 2020; Bergström et al., 2012) are typically added as plasticizers to improve film flexibility and processability by increasing the free volume or molecular mobility of the polymers. Plasticizers reduce intermolecular hydrogen bonding while increasing intermolecular spacing between the polymer chains (Suppakul, Jutakorn & Bangchokedee, 2010). In the past few years, copious research has been conducted on the properties of films produced from tamarind kernel polysaccharide (Agarwal et al., 2020; Ajovalasit et al., 2018; Alpizar-Reyes et al., 2022; Bergström et al., 2012; Hoque et al., 2022; Malviya et al., 2021; Mishra & Malhotra, 2009; Premalatha et al., 2016; Qureshi et al., 2021; Sudharsan et al., 2016; Suwanamornlert et al., 2020; Thivya et al., 2021). However, limited knowledge exists about the influence of commercial plasticizers such as tween and span on optical, mechanical, and thermal properties of films.

Thus, here, the effect of plasticizers such as polyethylene glycol 400, glycerol, sorbitol, tween 80, tween 40, tween 20 and span 20 was investigated on tamarind kernel polysaccharide-based film properties. Mechanical, optical, and thermal properties of the polysaccharide-based films were determined to explore the possibilities of using tamarind kernel polysaccharide to fabricate biomedical materials or biopolymer films in the food industry.

2. Objectives

The effects of various plasticizers on enhancing the flexibility of tamarind kernel polysaccharide films were evaluated and the optical properties and thermal behaviors of the developed films were investigated.

3. Materials and methods

3.1 Materials

Tamarind (*Tamarindus indica*) kernel powder was obtained from Freshy Thai Co., Ltd. (Chachoengsao Province, Thailand). Plasticizers (polyethylene glycol 400, glycerol, sorbitol, tween 80, tween 40, tween 20 and span 20), with properties shown in Table 1, and magnesium nitrate hexahydrate ($Mg(NO_3)_2 \cdot 6H_2O$) were purchased from Sigma-Aldrich (St Louis, MO, USA).

Table 1 Plasticizers selected for study and their properties

Plasticizer	Mol.wt. (g/mol)	HLB	Formula
Glycerol	92.09	10.2	$C_3H_8O_3$
Sorbitol	182	12.8	$C_6H_{14}O_6$
Span 20	346.46	8.6	$C_{18}H_{34}O_6$
PEG 400	400	11.6	$H(OCH_2CH_2)_nOH$ (n = 8.2 to 9.1)
Tween 20	1228	16.7	$C_{58}H_{114}O_{26}$
Tween 40	1277	15.6	$C_{62}H_{123}O_{26}$
Tween 80	1310	15	$C_{64}H_{124}O_{26}$

References: Antoniou et al. (2014), Lian et al. (2019), Garti et al. (1986)

3.2 Film preparation

Tamarind kernel polysaccharide (TKP) solution was prepared from tamarind kernel powder according to the method described in our previous study (Suwanamornlert, 2020) with some modifications. Briefly, tamarind kernel powder was dissolved in distilled water to obtain a 3% w/v aqueous solution and stirred with an overhead stirrer (IKA, RW20, Germany) on a hot plate at 80 °C for 10 min. The film-forming solution was then filtered through nylon filter cloth (mesh 60 micron) to remove undissolved matter. Polyethylene glycol 400, glycerol, sorbitol, tween 80, tween 40, tween 20 or span 20 (1% w/w film solution) were then added as plasticizers to the film-forming solution. Films were produced by the solution casting method. Before casting, the mixture was degassed using a bath-type sonicator to remove air bubbles for 20 min. The degassed film solution (30 g) was poured onto plastic petri dishes (200 mm diameter) and subsequently dried using a hot air oven (Binder, FD56, Germany) at 70 ± 2 °C for 24 hours. After dehydration, the dried thin films were peeled off the plastic petri dishes and conditioned in an environmental chamber at 25 ± 2 °C and 50% relative humidity for further studies. To establish 50% relative humidity, a saturated solution of $Mg(NO_3)_2 \cdot 6H_2O$ in distilled water was prepared and placed in a desiccator.

3.3 Film thickness

A hand-held micrometer caliper (Mitutoyo Corporation, Japan) was used to measure film thickness (mm) at 10 different points for each sample with average thickness reported.

3.4 Optical properties

Film specimens were cut into rectangular shapes and placed directly in a UV-Vis spectrophotometer test cell (Biochrom Libra S22, UK) with an incident wavelength of 600 nm (Rao et al., 2010). Percentage transmittance (%T) and absorbance (Abs) at 600 nm were reported. All measurements were performed in triplicate using air as the reference. The transparency and opacity of the films were calculated using equations (1) and (2), respectively according to the method described by Thivya et al. (2021).

$$\text{Transparency} = (\log\%T_{600})(x^{-1}) \quad (1)$$

$$\text{Opacity} = (Abs_{600})(x^{-1}) \quad (2)$$

where $\%T_{600}$ and Abs_{600} are percentage transmittance and absorbance at 600 nm, respectively and x is the film thickness (mm).

3.5 Color

The film color was determined using a colorimeter (Ultra Scan VIS, HunterLab, USA). L^* (black-white), a^* (+red to −green) and b^* (+yellow to −blue) values were averaged at six different points on each film. Total color difference (ΔE) was calculated using equation (3) and whiteness index (WI) of the film was obtained from equation (4) (Rao et al., 2010; Thivya et al., 2021).

$$\Delta E=\left[(\Delta L^*)^2+(\Delta a^*)^2+\left(\Delta b^*\right)^2\right]^{1/2} \quad (3)$$

$$WI=\left[(100-L^*)^2+(a^*)^2+\left(b^*\right)^2\right]^{1/2} \quad (4)$$

where $\Delta L^* = L^* - L_0^*$, $\Delta a^* = a^* - a_0^*$, $\Delta b^* = b^* - b_0^*$, where L_0^*, a_0^* and b_0^* are color values for TKP control films and L^*, a^* and b^* are color values for TKP films containing plasticizer.

3.6 Mechanical properties

Mechanical properties of all the film samples were determined as tensile strength (MPa), elastic modulus (MPa) and elongation at break (%) according to ASTM method D882-18. The films were cut into rectangular pieces (25 mm × 150 mm). Test specimens were conditioned at 25 ± 2 °C and 50% RH for at least 48 hours before testing. A Universal Testing Machine (model 5965, Instron, Norwood, MA, USA) equipped with a 5 kN load cell was performed at 50 mm/min speed with the gap between probes 100 mm. Tests were carried out as 10 replicates for each treatment.

3.7 Thermal analysis

Thermal properties of the samples were analyzed using differential scanning calorimetry (DSC) equipment (Perkin-Elmer, Waltham, MA, USA). Samples weighing 6-8 mg were sealed in 40 µL aluminum pans, with an empty pan used as a reference. In the preliminary study, the author found that glass transition temperature and melting temperature of tamarind kernel polysaccharide were showed at 50 °C and 93 °C, respectively. Hence, the samples were heated from 10 to 200 °C, cooled to 10 °C and then reheated to 200 °C according to a previous study (Premalatha et al., 2016). Heating and cooling rates were set at 10 °C/min under nitrogen atmosphere with a flow rate

of 10 mL/min. All treatments were run at least in duplicate. Glass transition temperature (T_g) and melting point temperature (T_m) were recorded.

3.8 Statistical analysis

Analysis of variance (ANOVA) and Duncan's new multiple range test were performed at a significant level of $p \leq 0.05$ using SPSS 10.0 statistical software.

4. Results and discussion

4.1 Film preparation

Film casting from film solutions plasticized with various types of plasticizers (Figure 1) exhibited higher or lower flexibility and transparency (Figure 2). Film prepared from tamarind kernel polysaccharide without plasticizer was very brittle and broke when peeled from the plastic petri dish. Phase separation occurred in the film containing 1% (w/w) polyethylene glycol 400, visible as a large molecule of polyethylene glycol 400 on the film surface, resulting in lower transparency compared to the other films. The films containing PEG-400 exhibited a white, opaque, and rough surface. This might be because of the high molecular weight and relatively low content of hydroxyl groups within PEG-400 chemical structure. The phenomena lead to phase separation and incompatibility of PEG-400 with tamarind kernel polysaccharide films. Similar observations have been found for film made from pea starch/guar gum, a polysaccharide-based polymer, plasticized with polyethylene glycol 400 (Saberi et al., 2017).

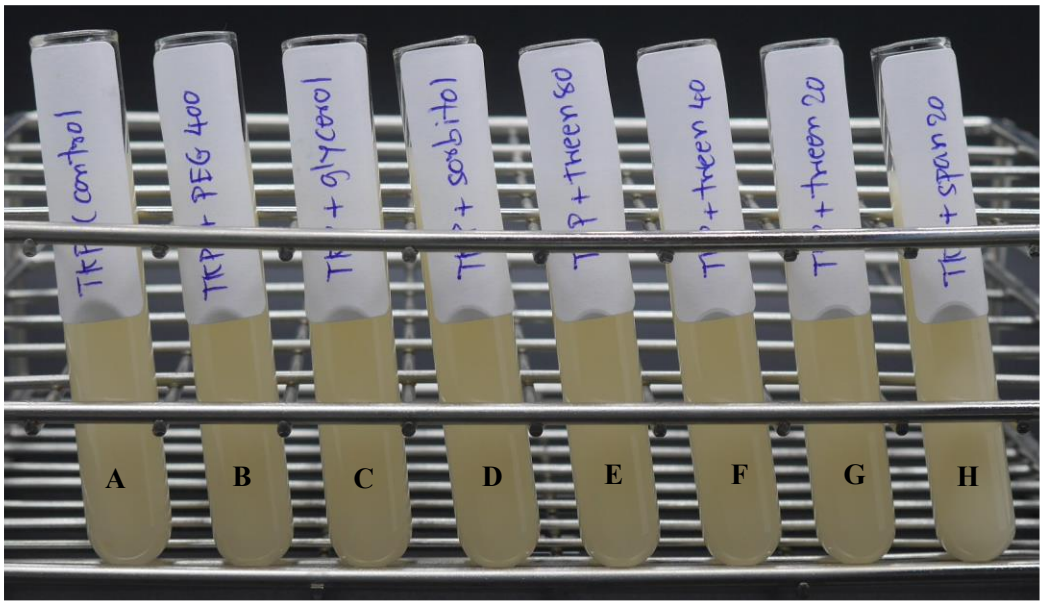

Figure 1 Tamarind kernel polysaccharide film solution containing 1% (w/w) plasticizer (A) control, (B) polyethylene glycol 400, (C) glycerol, (D) sorbitol, (E) tween 80, (F) tween 40, (G) tween 20 and (H) span 20.

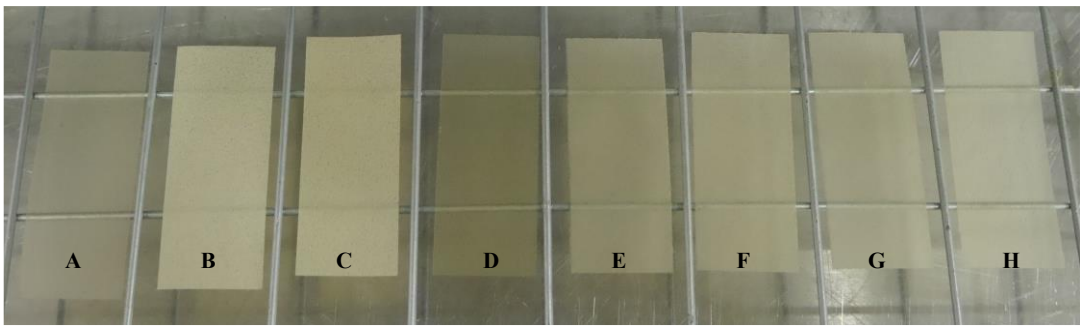

Figure 2 Tamarind kernel polysaccharide films containing 1% (w/w) plasticizer (A) control, (B) polyethylene glycol 400, (C) glycerol, (D) sorbitol, (E) tween 80, (F) tween 40, (G) tween 20 and (H) span 20.

4.2 Thickness and optical properties

Thickness and optical properties of tamarind kernel polysaccharide film containing plasticizers are shown in Table 2. The film thickness affects the mechanical and optical properties which depend on preparation method and drying condition. Thus, the amount of film solution in the petri dish, drying time and drying temperature must be controlled during the film preparation process. Tamarind kernel polysaccharide is a water-soluble polymer. Due to its hydrophilic property, drying temperature and time affects moisture content in the films (Effendi et al., 2022). The thickness decreased with increase in drying temperature and time and, therefore, the opacity is increased. The thicknesses of tamarind kernel polysaccharide and tamarind kernel polysaccharide plasticized films were 0.060 mm and ranged 0.061 to 0.074 mm, respectively. The thickness of tamarind kernel polysaccharide film was lower due to the compact polymer chains. Films comprising sorbitol showed significantly increased thickness ($p \leq 0.05$) because of greater disruption of the polymer chains by the plasticizer. No significant differences in thickness were observed between films containing sorbitol and the control.

Transparency and opacity are important characteristics of biopolymer materials when the films are exposed to visible light (Martins et al., 2012). Results showed decrease in transparency with increased greenness and yellowness for tamarind kernel polysaccharide plasticized with PEG 400, glycerol, tween 80, tween 40, tween 20 and span 20 compared to the control film (Table 2). The opacity of plasticized film samples ranged from 8.34 to 17.32, while opacity of the control film was 9.97. Film plasticized with sorbitol exhibited the highest transparency, while transparency values of films containing polyethylene glycol 400, glycerol, tween 80, tween 40, tween 20 and span 20 were significantly lower. The opacity and transparency of the sorbitol-plasticized film were lower than neat tamarind kernel polysaccharide film, but the differences were not significant. Films added with polyethylene glycol 400 and glycerol presented the highest opacity values compared to the other films. The decrease in transparency and increase in opacity of films containing polyethylene glycol 400 and glycerol were due to the inability of larger plasticizer molecules to insert between the polymer chains of gum (Aydinli et al., 2004).

.

Table 2 Effect of plasticizers on film transparency and opacity.

Film sample	Thickness (mm)	Optical properties	
		Transparency	Opacity
TKP (control)	$0.060^{a} \pm 3.25$	$25.90^{e} \pm 3.58$	$9.97^{ab} \pm 1.25$
TKP+PEG 400	$0.073^{b} \pm 1.80$	$13.18^{abc} \pm 3.02$	$15.58^{ef} \pm 2.80$
TKP+glycerol	$0.074^{b} \pm 2.21$	$13.70^{bc} \pm 2.37$	$17.32^{f} \pm 0.38$
TKP+sorbitol	$0.061^{a} \pm 1.04$	$24.41^{e} \pm 1.66$	$8.34^{a} \pm 0.62$
TKP+tween 80	$0.073^{b} \pm 0.64$	$12.41^{ab} \pm 2.24$	$11.92^{bc} \pm 0.75$
TKP+tween 40	$0.074^{b} \pm 4.54$	$9.71^{a} \pm 0.81$	$10.61^{b} \pm 1.46$
TKP+tween 20	$0.068^{b} \pm 2.93$	$17.37^{d} \pm 2.89$	$13.01^{cd} \pm 0.75$
TKP+span 20	$0.073^{b} \pm 9.15$	$16.73^{cd} \pm 1.84$	$14.35^{de} \pm 1.20$

[1]Values are mean with standard deviation (n = 3)

[2]TKP, tamarind kernel polysaccharide film; TKP-PEG400, TKP film plasticized with 1% (w/w) PEG 400; TKP-glycerol, TKP film plasticized with 1% (w/w) glycerol; TKP-sorbitol, TKP film plasticized with 1% (w/w) sorbitol; TKP-tween 80, TKP film plasticized with 1% (w/w) tween 80; TKP-tween 40, TKP film plasticized with 1% (w/w) tween 40; TKP-tween 20, TKP film plasticized with 1% (w/w) tween 20; TKP-span 20, TKP film plasticized with 1% (w/w) span 20.

[a–f]Means with different superscripts within a column indicate significant differences ($p \leq 0.05$).

Table 3 Color parameters of the films.

Film sample	Color parameters				
	L^*	a^*	b^*	ΔE	WI
TKP (control)	35.41^{b} ±0.57	-0.94^{bc} ±0.09	2.23^{a} ±0.55	-	35.33^{a} ±1.46
TKP+PEG 400	54.91^{f} ±0.14	-1.58^{a} ±0.32	8.79^{d} ±1.05	212.27^{d} ±5.60	62.85^{b} ±10.09
TKP+glycerol	52.43^{e} ±0.35	-1.29^{ab} ±0.50	9.78^{e} ±0.79	173.70^{c} ±8.40	72.77^{c} ±7.63
TKP+sorbitol	33.21^{a} ±0.67	-0.68^{c} ±0.07	2.79^{ab} ±0.36	2.80^{a} ±1.83	37.56^{a} ±1.34
TKP+tween 80	38.32^{c} ±1.17	-0.97^{bc} ±0.07	3.78^{b} ±0.29	5.92^{a} ±3.45	38.47^{a} ±1.55
TKP+tween 40	42.76^{d} ±2.31	-1.21^{ab} ±0.17	5.18^{c} ±0.13	33.18^{b} ±15.18	42.81^{a} ±1.80
TKP+tween 20	38.75^{c} ±0.67	-1.11^{bc} ±0.06	2.48^{a} ±0.37	5.81^{a} ±2.20	34.35^{a} ±1.19
TKP+span 20	37.71^{c} ±0.33	-1.19^{ab} ±0.16	3.60^{b} ±0.39	3.71^{a} ±1.12	38.38^{a} ±1.43

[1]Values are mean with standard deviation (n = 3)

[2]TKP, tamarind kernel polysaccharide film; TKP-PEG400, TKP film plasticized with 1% (w/w) PEG 400; TKP-glycerol, TKP film plasticized with 1% (w/w) glycerol; TKP-sorbitol, TKP film plasticized with 1% (w/w) sorbitol; TKP-tween 80, TKP film plasticized with 1% (w/w) tween 80; TKP-tween 40, TKP film plasticized with 1% (w/w) tween 40; TKP-tween 20, TKP film plasticized with 1% (w/w) tween 20; TKP-span 20, TKP film plasticized with 1% (w/w) span 20.

[a–f]Means with different superscripts within a column indicate significant differences ($p \leq 0.05$)

Color is also a critical parameter in terms of consumer acceptability of biopolymer films and depends on various factors including plasticizer incorporation, film production and condition of storage (Pérez et al., 2016). Effects of the plasticizer on color parameters of the films are shown in Table 3. Results showed an increase in ΔE values by incorporation of various plasticizers due to changes in L^*, a^* and b^* values of the films. The L^* values for polyethylene glycol 400, glycerol, tween 80, tween 40, tween 20 and span 20 were significantly higher compared to the control and sorbitol-plasticized films. This increment in L^* values led to an increase in the whiteness index (WI) of the films. The appearance of sorbitol-plasticized film was pale yellow color and translucent, whereas film plasticized with polyethylene glycol 400, and glycerol appeared as an opaque white surface, as shown by whiteness index values in Table 3. Similar results were obtained for pea starch-guar gum film with polyethylene glycol 400 (Saberi et al., 2017). They reported that the white opaque appearance was due to the high-molecular-weight plasticizer and relatively low content of hydroxyl groups within polyethylene glycol 400 that could not interact with the polymer chains and then escaped to the surface of the film. No significant differences in the whiteness index were observed between the tamarind kernel polysaccharide film and the films containing sorbitol, tween 80, tween 40, tween 20 and span 20

4.3 Mechanical properties

Mechanical properties of the plasticized tamarind kernel polysaccharide films including tensile strength, elastic modulus and elongation at break are summarized in Figures 3-5. Significant differences ($p \leq 0.05$) were observed between the mechanical properties of the film samples. Although tamarind kernel polysaccharide absorbs moisture and acts as a plasticizer, the neat tamarind kernel polysaccharide film was brittle, with high elastic modulus compared to the other plasticized films. The tensile strength and elongation at break of tamarind kernel polysaccharide-based films were 6.07 to 18.45 MPa and 1.36 to 5.91%, respectively depending on film formulation. Tamarind kernel polysaccharide film plasticized with sorbitol exhibited the lowest tensile strength and elastic modulus but showed the highest elongation at break compared to the other films. Tensile strengths of films containing polyethylene glycol 400, glycerol, tween 20 and span 20 were significantly lower than films plasticized with tween 80 and tween 40. No significant differences in elongation at break were observed between films containing polyethylene glycol 400, glycerol, tween 40, tween 20 and span 20. For films containing tween 80, tween 40 and tween 20, tensile strength and elastic modulus decreased with increasing HLB value. However, percentage elongation at break showed no significant difference ($p > 0.05$). HLB values of selected plasticizers were shown in Table 1. Plasticizers with lower HLB values exhibited the

main lipophilic balance in the molecular structure, whereas those with higher HLB values showed greater association of their hydrophilic fraction with the hydrophilic film matrix (Zhang et al., 2022). In this study, films plasticized with sorbitol and tween may bind to hydrophilic tamarind kernel polysaccharide, in order to improve the compatibility and conformity of film structure.

Polyethylene glycol 400, glycerol and sorbitol exhibited a higher plasticizing effect in tamarind kernel polysaccharide film than tween 80, tween 40, tween 20 and span 20. Sorbitol had a greater effect than polyethylene glycol 400 and glycerol due to lower molecular weight and higher water solubility that allowed better interaction between the plasticizer-polymer chains. A similar result was reported by Haq et al. (2014) for gum Cordia-based films plasticized with glycerol, sorbitol, polyethylene glycol 200 and polyethylene glycol 400. The observation was attributed to the inability of polyethylene glycol 400, which has higher molecular size, to penetrate and reduce the attraction between the polymer chain of gum film. A contrary result was reported by Antoniou et al. (2014) for glycerol-plasticized Tara gum films that exhibited higher percentage elongation at break than sorbitol and polyethylene glycol 400 plasticized films. Glycerol has a lower molecular weight and could easily fit between the polymer chains and improve the elasticity of the films (Mikkonen et al., 2007). The effectiveness of each plasticizer is associated with molecular weight, shape, number of free hydroxyl groups, spacing of oxygen atoms, water binding capacity and the compatibility of the plasticizer with the polymer (Saberi et al., 2017). Similar results were reported by Saberi et al. (2017) for pea starch-guar gum films with glycols, sugars, and polyols. They revealed that a greater number of hydroxyl groups in the plasticizer increased interaction between the carboxyl groups of starch and guar gum. These reactions lead to a reduction of movement between the chains of starch and guar gum.

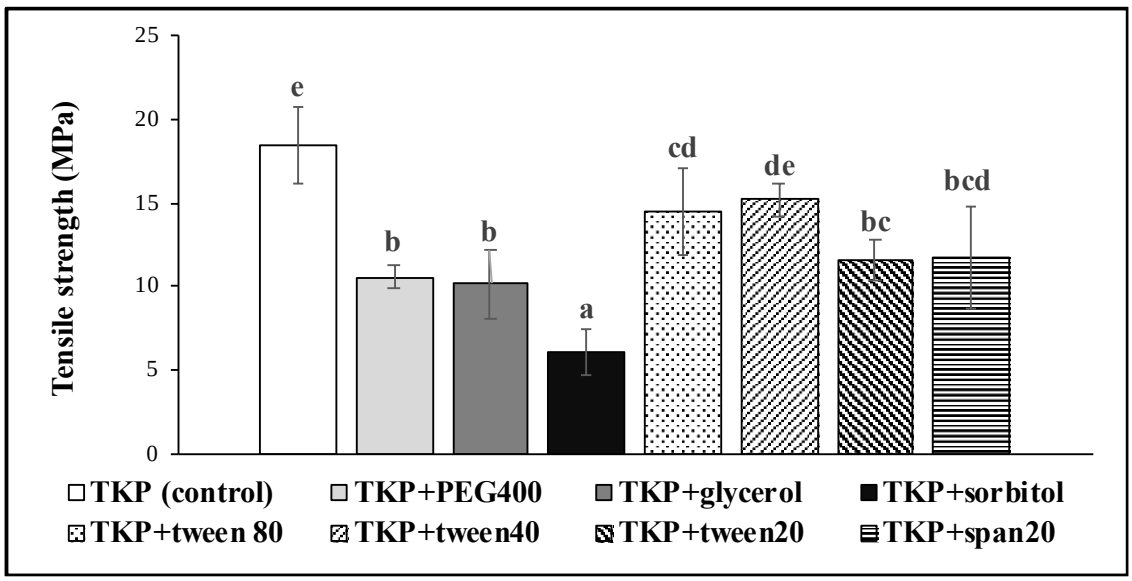

Figure 3 Tensile strength of tamarind kernel polysaccharide (TKP) film (□) compared to TKP films containing 1% w/w plasticizer (□) polyethylene glycol 400, (■) glycerol, (■) sorbitol, (⊡) tween 80, (▨) tween 40, (▧) tween 20 and (▤) span 20. [a–e] Means with different superscripts within a bar graph indicate significant differences ($p \leq 0.05$).

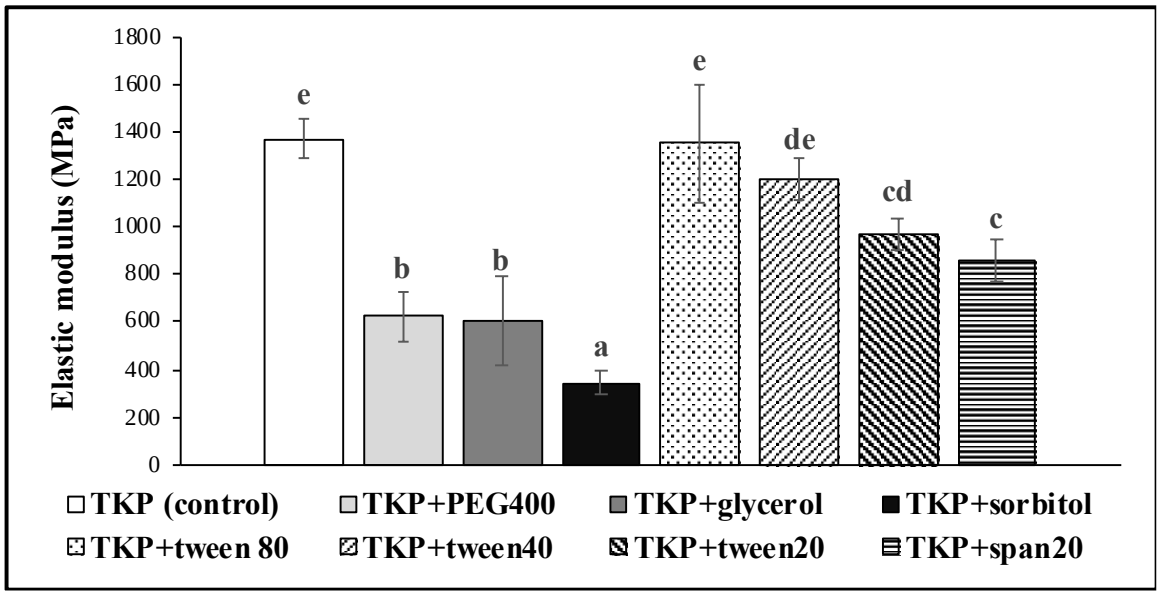

Figure 4 Elastic modulus of tamarind kernel polysaccharide (TKP) film (□) compared to TKP films containing 1% w/w plasticizer (□) polyethylene glycol 400, (■) glycerol, (■) sorbitol, (⊡) tween 80, (▨) tween 40, (▧) tween 20 and (▤) span 20. [a–e] Means with different superscripts within a bar graph indicate significant differences ($p \leq 0.05$).

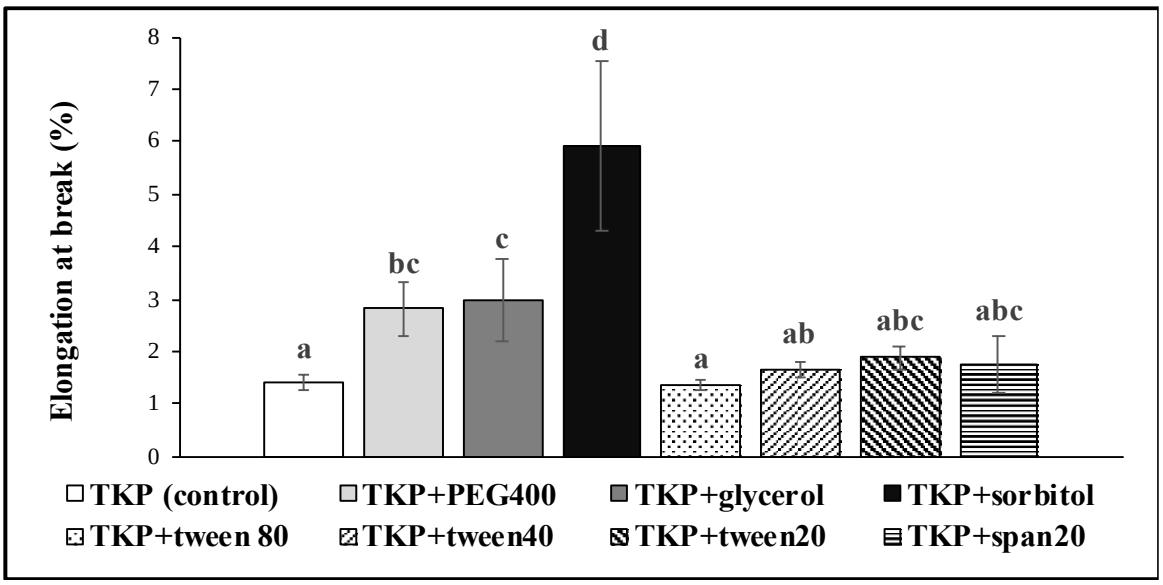

Figure 5 Elongation at break of tamarind kernel polysaccharide (TKP) film (□) compared to TKP films containing 1% w/w plasticizer (□) polyethylene glycol 400, (■) glycerol, (■) sorbitol, (⊡) tween 80, (▨) tween 40, (▧) tween 20 and (▤) span 20. [a–d] Means with different superscripts within a bar graph indicate significant differences ($p \leq 0.05$).

4.4 Thermal properties

Differential scanning calorimetry (DSC) was used to study the thermal characteristics of the tamarind kernel polysaccharide films. DSC thermograms of neat tamarind kernel polysaccharide and tamarind kernel polysaccharide films incorporated with the selected plasticizers (glycerol, sorbitol, and tween 20) are shown in Figure 6. The melting temperature (T_m) of tamarind kernel polysaccharide film was 92.88 °C corresponding to the occurrence of tamarind kernel-starch gelatinization (Sudharsan et al., 2016). The

thermogram of native tamarind kernel polysaccharide revealed a sharp endothermic peak at 305 °C, attributed to its melting (T_m) and degradation temperatures (T_d) (data not shown). This result was supported by Carvalho et al. (2022) who found a degradation temperature of tamarind gum at 299 °C by using thermogravimetric analysis. Tamarind kernel polysaccharide films have high T_m, indicating high thermal stability due to their complex structure (Thivya et al., 2021). The glass transition temperature (T_g) of neat tamarind kernel polysaccharide film was 49.81 °C. Tamarind kernel polysaccharide film plasticized with sorbitol exhibited the lowest glass transition temperature compared to the others, including tamarind kernel polysaccharide film, as shown in Figure 6. The addition of sorbitol decreased the T_g of tamarind kernel polysaccharide film from 49.81 °C to 20.97 °C, while films plasticized with glycerol and tween 20 exhibited T_g values at 42.41 and 42.92 °C, respectively. This result corresponded to reduction of tensile strength and extension of elongation at break of the sorbitol/tamarind kernel polysaccharide mixture, as shown in Figures 3 and 5. Both sorbitol and glycerol decreased the T_g of tamarind kernel polysaccharide film as expected, based on their compatibility with tamarind kernel polysaccharide. By contrast, Bergström et al. (2012) found that glycerol was more effective due to its lower T_g and molecular weight compared to sorbitol.

5. Conclusions

Polymer morphology plays an important role in the mechanical properties of the film, which consequently affects the processes utilized in food production. In this study, the presence of plasticizers affected the optical, mechanical, and thermal properties of tamarind kernel polysaccharide films. Addition of sorbitol resulted in the best mechanical and optical properties in tamarind kernel polysaccharide films and exhibited the highest elongation at break and transparency. Solvent casting of a tamarind kernel polysaccharide/sorbitol mixture produced transparent films with good mechanical properties. Differential scanning calorimetry measurements showed that incorporation of plasticizers increased the mobility of the system by decreasing the intermolecular forces between the polymer chains, therefore reducing both glass transition temperature and melting temperature of the films. Our results indicated that sorbitol-plasticized tamarind kernel polysaccharide has potential use in the manufacture of biomedical materials or biopolymer films in the food industry.

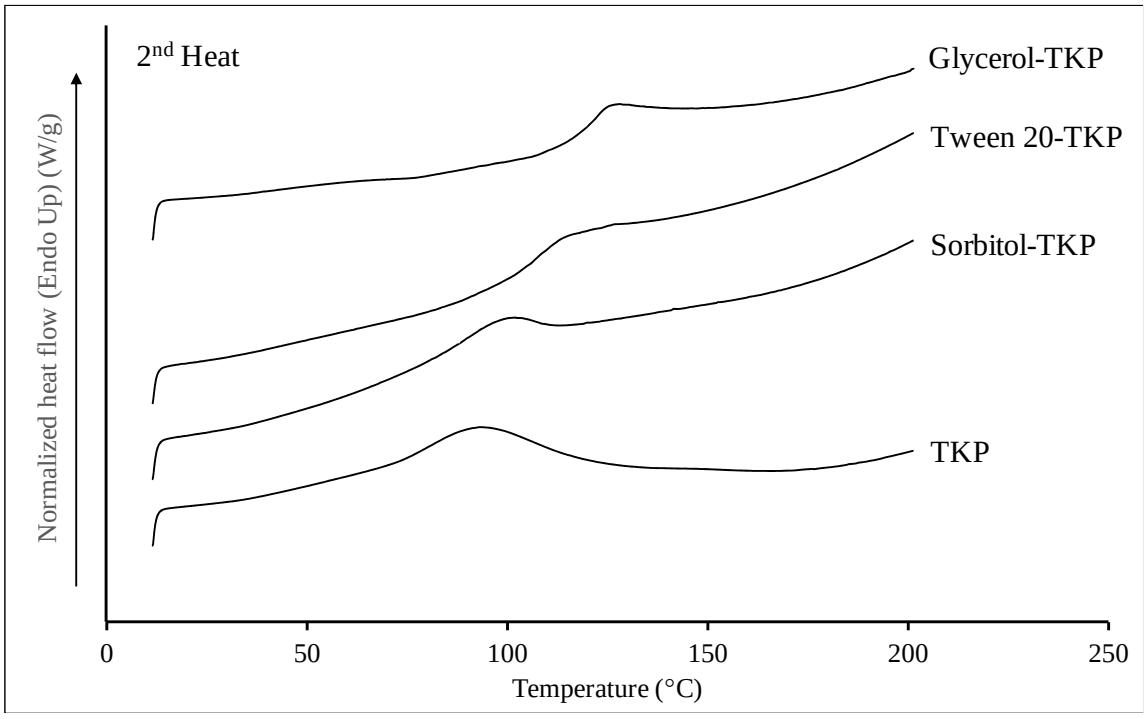

Figure 6 Differential scanning calorimetric analysis of plasticized-tamarind kernel polysaccharide

6. Acknowledgements

The authors would like to thank the Dhurakij Pundit University Research Development and Innovation (DPURDI), Dhurakij Pundit University, Bangkok 10210, Thailand for financial support.

7. References

Ajovalasit, A., Sabatino, M. A., Todaro, S., Alessi, S., Giacomazza, D., Picone, P., ... & Dispenza, C. (2018). Xyloglucan-based hydrogel films for wound dressing: Structure-property relationships. *Carbohydrate Polymers*, *179*, 262-272. https://doi.org/10.1016/j.carbpol.2017.09.092

ASTM (2018). *Standard test methods for tensile properties of thin plastic sheeting* (Vol. D882-18). West Conshohocken, PA: ASTM international.

Agarwal, S., Hoque, M., Bandara, N., Pal, K., & Sarkar, P. (2020). Synthesis and characterization of tamarind kernel powder-based antimicrobial edible films loaded with geraniol. *Food Packaging and Shelf life*, *26*, Article 100562. https://doi.org/10.1016/j.fpsl.2020.100562

Antoniou, J., Liu, F., Majeed, H., Qazi, H. J., & Zhong, F. (2014). Physicochemical and thermomechanical characterization of tara gum edible films: Effect of polyols as plasticizers. *Carbohydrate Polymers*, *111*, 359-365. https://doi.org/10.1016/j.carbpol.2014.04.005

Alpizar-Reyes, E., Cortés-Camargo, S., Román-Guerrero, A., & Pérez-Alonso, C. (2022). Tamarind gum as a wall material in the microencapsulation of drugs and natural products. In *Micro and Nano Technologies, Micro and nanoengineered gum-based biomaterials for drug delivery and biomedical applications.* (pp. 347-382). Amsterdam: Academic Press. https://doi.org/10.1016/B978-0-323-90986-0.00016-9

Aydinli, M., Tutas, M., & Bozdemir, O. A. (2004). Mechanical and light transmittance properties of locust bean gum based edible films. *Turkish Journal of Chemistry*, *28*(3), 163-172.

Bergström, E. M., Salmén, L., Kochumalayil, J., Berglund, L. (2012). Plasticized xyloglucan for improved toughness—Thermal and mechanical behaviour. *Carbohydrate Polymers*, *87*, 2532-2537. https://doi.org 10.1016/j.carbpol.2011.11.024

Carvalho, R. T. R., Pedrosa, C. D., Leal, A. C. P., Palermo, L. C. M., & Mansur, C. R. E. (2022). Extraction, characterization and rheological behavior of tamarind gum under high salinity. *Brazilian Journal of Analytical Chemistry*, *9*(35), 62-75. https://doi.org 10.30744/brjac.2179-3425.AR-104-2021

Choi, J., Lee, J., Han, J., & Chang, Y. (2023). Development of gelatin–sodium caseinate high-oxygen-barrier film containing elderberry (*Sambucus nigra* L.) extract and its antioxidant capacity on pork. *Food Bioscience*, *53*, Article 102617. https://doi.org/10.1016/j.fbio.2023.102617

Chou, M. Y., Osako, K., Lee, T. A., Wang, M. F., Lu, W. C., Wu, W. J., ... & Ho, J. H. (2023). Characterization and antibacterial properties of fish skin gelatin/guava leaf extract bio-composited films incorporated with catechin. *LWT*, *178* (15), Article 114568. https://doi.org/10.1016/j.lwt.2023.114568

Effendi, A. D., Jiaqi, L., Sia, C. W., Jasamai, M., & Sulaimon, A. A. (2022). Polysaccharides from *Tamarindus indica* L. as natural kinetic hydrates inhibitor at high subcooling environment. *Journal of Petroleum Exploration and Production Technology*, *12*(10), 2711-722. https://doi.org/10.1007/s13202-022-01477-2

Garti, N., Aserin, A., & Kopilovich, A. (1986). Transparent macroemulsions for cosmetic applications. *International Journal of Cosmetic Science*, *8*(1), 1-8. https://doi.org/10.1111/j.1467-2494.1986.tb00425.x

Gupta, I., Cherwoo, L., Bhatia, R., & Setia, H. (2022). Biopolymers: Implications and application in the food industry. *Biocatalysis and Agricultural Biotechnology*, *46*, Article 102534. https://doi.org/10.1016/j.bcab.2022.102534

Haghighatpanah, N., Omar-Aziz, M., Gharaghani, M., Khodaiyan, F., Hosseini, H. H., & Kennedy, J. H. (2022). Effect of mung bean protein isolate/pullulan films containing marjoram (*Origanum majorana* L.) essential oil on chemical and microbial properties of minced beef meat. *International Journal of Biological Macromolecules*, *201*, 318-329. https://doi.org/10.1016/j.ijbiomac.2022.01.023

Haq, M. A., Hasnain, A., & Azam, M. (2014). Characterization of edible gum cordia film: Effects of plasticizers. *LWT - Food Science*

and Technology, *55*, 163-169. https://doi.org/10.1016/j.lwt.2013.09.027

Hoque, M., Sarkar, P., & Ahmed, J. (2022). Preparation and characterization of tamarind kernel powder/ZnO nanoparticle-based food packaging films. *Industrial Crops and Products*, 178, Article 114670. https://doi.org/10.1016/j.indcrop.2022.114670

Kozioł, A., Cybulska, J., Pieczywek, P. M., & Zdunek, A. (2015). Evaluation of structure and assembly of xyloglucan from tamarind seed (*Tamarindus indica* L.) with Atomic Force Microscopy. *Food Biophysics*, *10*, 396-402. https://doi.org 10.1007/s11483-015-9395-2

Lian, H., Peng, Y., Shi, J., & Wang, Q. (2019). Effect of emulsifier hydrophilic-lipophilic balance (HLB) on the release of thyme essential oil from chitosan films. *Food Hydrocolloids*, *97*, Article 105213. https://doi.org/10.1016/j.foodhyd.2019.105213

Lin, L., Mei, C., Shi, C., Li, C., Abdel-Samie, M. A., & Cui, H. (2023). Preparation and characterization of gelatin active packaging film loaded with eugenol nanoparticles and its application in chicken preservation. *Food Bioscience*, *53*, 102778. https://doi.org/10.1016/j.fbio.2023.102778

Malviya, R., Sundram, S., Fuloria, S., Subramaniyan, V., Sathasivam, K. V., Azad, A. K., ... & Fuloria, N. K. (2021). Evaluation and characterization of tamarind gum polysaccharide: The biopolymer. *Polymers*, *13*(18), Article 3023. https://doi.org/10.3390/polym13183023

Martins, J. T., Cerqueira, M. A., & Vicente, A. A. (2012). In fluence of α-tocopherol on physicochemical properties of chitosan-based films. *Food Hydrocolloid*, *27*(1), 220-227. https://doi.org/10.1016/j.foodh yd.2011.06.011

Mikkonen, K. S., Rita, H., Helén, H., Talja, R. A., Hyvönen, L., & Tenkanen, M. (2007). Effect of polysaccharide structure on mechanical and thermal properties of galactomannan-based films. *Biomacromolecules*, *8*(10), 3198-3205. https://doi.org/10.1021/bm700538c.

Mishra, A., & Malhotra, A. V. (2009). Tamarind xyloglucan: A polysaccharide with versatile application potential. *Journal of Materials Chemistry*, *19*(45), 8528–8536. https://doi.org/10.1016/j.carbpol.2011.11.024

Nagar, C. K., Dash, S. K., & Rayaguru, K. (2022). Tamarind seed: Composition, applications, and value addition: A comprehensive review. *Journal of Food Processing and Preservation*, *46*(10), Article e16872. https://doi.org/10.1111/jfpp.16872

Pérez, L.M., Piccirilli, G.N., Delorenzi, N.J., & Verdini, R.A. (2016). Effect of different combinations of glycerol and/or trehalose on physical and structural properties of whey protein concentrate-based edible films. *Food Hydrocolloid*, *56*, 352-359. https://doi.org/10.1016/j.foodhyd.2015.12.037

Premalatha, M., Mathavan, T., Selvasekarapandian, S., Monisha, S., Vinoth Pandi, D., & Selvalakshmi, S. (2016). Investigations on proton conducting biopolymer membranes based on tamarind seed polysaccharide incorporated with ammonium thiocyanate. *Journal of Non-Crystalline Solids*, *453*, 131-140. https://doi.org/10.1016/j.jnoncrysol.2016.10.008

Qureshi, D., Behera, K. P., Mohanty, D., Mahapatra, S. K., Verma, S., Sukyai, P., ... & Pal, K. (2021). Synthesis of novel poly (vinyl alcohol)/tamarind gum/bentonite-based composite films for drug delivery applications. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, *613*, Article 126043. https://doi.org/10.1016/j.colsurfa.2020.126043

Rao, M. S., Kanatt, S. R., Chawla, S. P., & Sharma, A. (2010). Chitosan and guar gum composite films: Preparation, physical, mechanical and antimicrobial properties. *Carbohydrate Polymers*, *82*(4), 1243-1247. https://doi.org/10.1016/j.carbpol.2010.06.058

Saberi, B., Chockchaisawasdee, S., Golding, J. B., Scarlett, C. J., & Stathopoulos, C. E. (2017). Physical and mechanical properties of a new edible film made of pea starch and guar gum as affected by glycols, sugars and polyols. *International Journal of Biological Macromolecules*, *104*, 345-359. https://doi.org/10.1016/j.ijbiomac.2017.06.051

Sudharsan, K., Mohan, C. C., Babu, P. A. S., Archana, G., Sabina, K., Sivarajan, M., & Sukumar, M. (2016). Production and characterization of cellulose reinforced starch (CRT) films. *International Journal of Biological Macromolecules*, *83*, 385-395. https://doi.org/10.1016/j.ijbiomac.2015.11.037

Suppakul, P., Jutakorn, K., & Bangchokedee, Y. (2010). Efficacy of cellulose-based coating on enhancing the shelf life of fresh eggs. *Journal of Food Engineering*, *98*, 207–213.

https://doi.org/10.1016/j.jfoodeng.2009.12.027

Suresh, S. N., Puspharaj, C., & Subramani, R. (2022). Development of Almond gum/alginate composites to enhance the shelf-life of post-harvest Solanum Lycopersicum L. *Food Hydrocolloids for Health*, *2*, Article 100087. https://doi.org/10.1016/j.fhfh.2022.100087

Suwanamornlert, P. (2020). Application of tamarind kernel polysaccharide-soybean oil emulsion coating to preserve the internal quality and extend shelf-life of fresh eggs. *Journal of Current Science and Technology*, *10*(2), 99-107. https://doi.org/10.14456/jcst.2020.10

Thivya, P., Bhosale, Y. L., Anandakumar, S., Hema, V., & Sinija, V. R. (2021). Exploring the effective utilization of shallot stalk waste and tamarind seed for packaging film preparation. *Waste and Biomass Valorization*, *12*, 5579-5794. https://doi.org/10.1007/s12649-021-01402-4

Zhang, R., Zhai, X., Wang, W., & Hou, H. (2022). Preparation and evaluation of agar/maltodextrin-beeswax emulsion films with various hydrophilic-lipophilic balance emulsifiers. *Food Chemistry*, *384*, Article 132541. https://doi.org/10.1016/j.foodchem.2022.132541