

Research Article

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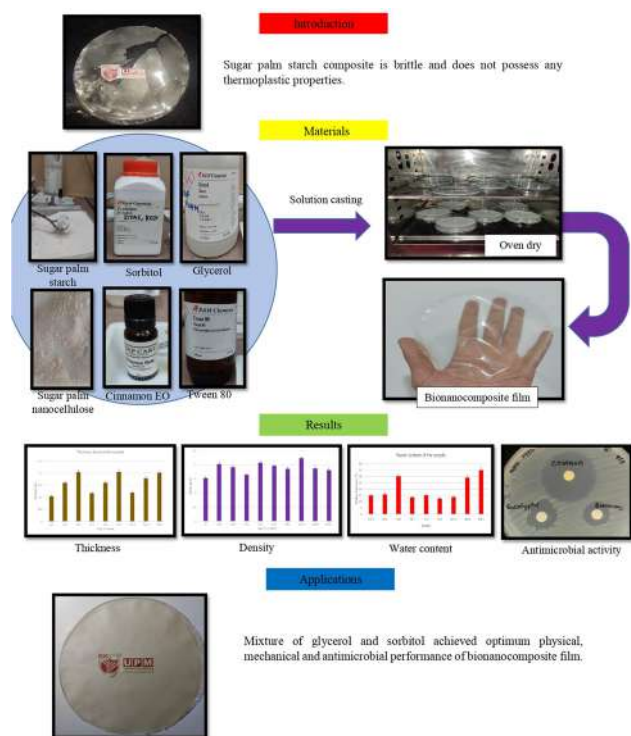
Effect of plasticizers on the properties of sugar palm nanocellulose/cinnamon essential oil reinforced starch bionanocomposite films

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Abstract: This work examines the effects of plasticizer type and concentration on mechanical, physical, and antibacterial characteristics of sugar palm nanocellulose/sugar palm starch (SPS)/cinnamon essential oil bionanocomposite films. In this research, the preparation of SPS films were conducted using glycerol (G), sorbitol (S), and their blend (GS) as plasticizers at ratios of 1.5, 3.0, and 4.5 wt%. The bionanocomposite films were developed by the solution casting method. Plasticizers were added to the SPS film-forming solutions to help overcome the fragile and brittle nature of the unplasticized SPS films. Increasing plasticizer contents resulted in an increase in film thickness and moisture contents. On the contrary, the increase in plasticizer concentrations resulted in the decrease of the densities of the plasticized films. The

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Graphical abstract: Processing of bionanocomposite film with various types and concentrations of plasticizers.

increase in the plasticizer content from 1.5 to 4.5% revealed less influence towards the moisture content of S-plasticised films. For glycerol and glycerol-sorbitol plasticized (G and GS) films, higher moisture content was observed compared to S-plasticised films. Various plasticizer types did not significantly modify the antibacterial activity of bionanocomposite films. The findings of this study showed significant improvement in the properties of bionanocomposite films with different types and concentrations of plasticizers and their potential for food packaging applications was enhanced.

Keywords: plasticizer types, plasticizer concentration, sorbitol, sugar palm biopolymer, bionanocomposites, glycerol, physical properties

1 Introduction

Petroleum-based polymers are broadly utilized in the packaging industry due to their remarkable flexibility, barrier, and mechanical capabilities [1–5]. Despite their various benefits, petroleum-based plastics is among the major sources of pollutions for being non-biodegradable. However, as they originate from non-renewable resources including toxic and harmful substances, they pose severe health and environmental risks [6–8]. Non-biodegradable polymer packaging products are very resistant to microbial attacks that stay in the environment for decades after disposal. Because of their nonbiodegradability and non-renewable sources, petroleum-based plastics are regarded as one of the leading causes of solid waste formation and deposition in the environment. The rapid petroleum reserves depletion, along with the fact that petroleum-based polymers are nonbiodegradable, has raised concerns about their widespread use in packaging. The plastic polymers advantages, *e.g.* clear optical, low cost, easy availability, mechanical qualities, heat sealability, and resistance to water and grease have proved them to be the most practical and cost-effective alternative for packaging applications [1,7,9,10].

Responding to this problem, various research groups and industrial organizations around the world are presently working on new eco-friendly packaging solutions that take advantage of biopolymer's "ecological" benefits in applications such as food packaging [11]. New materials made from renewable resources have exploded in popularity during the last decade [12]. Most of the research focused on polysaccharides, proteins, and lipids to be used in edible films. The technological properties could be improved *via* chemical reactions (*e.g.* cross-linking) or physical treatments (*e.g.* ultrasound, heat, or radiation) to change their molecular structures [13,14]. Various studies confirmed the growing interest in the utilization of starch derived from numerous sources incorporated with essential oils (EOs) to prepare biobased packaging films. The rigidity of bionanocomposite films is contributed by the strong interactions between amylose–amylopectin or amylose–amylose in the polymer matrix [15–18]. By adding a plasticizer, these films' inherent brittleness and rigidity are reduced, while their malleability, ductility, and flexibility are increased. In literature studies, the addition of nanofillers into a biopolymer matrix led to an improvement in the properties of biopolymer-based packaging films including enhanced mechanical, thermal, barrier, and physicochemical properties [19–21]. On the other hand, it is not very common to study the effect of the plasticizer type and concentration on the films of sugar palm nanocellulose reinforced sugar palm starch (SPS)/cinnamon

essential oil (CEO) bionanocomposite films. Recent studies by Ilyas *et al.* [13], Syafiq *et al.* [1], and Sanyang *et al.* [7] have highlighted the sugar palm tree as a promising source of starch for biopolymer use. Sugar palm (*Arenga pinnata*), a versatile tree, is native to many tropical countries, most notably South-East Asia. The tree is well-known for its various economic applications [22–24]. Additionally, starch can be collected from the trunk of the tree, especially when the tree no longer produced fruits and sugar [25]. Traditionally, SPS is utilized as a feedstock for materials [26]. According to Ilyas *et al.* [27] and Nazrin *et al.* [28], SPS film containing 0.5% sugar palm nanocellulose revealed improved tensile strength of 140% [27] and water vapour permeability of 19.94% [28]. Nevertheless, it still has not gained the attention it requires to be developed as an industrial starch biopolymer. Starch is regarded among the most promising biopolymers owing to their benefits, *e.g.* renewability, biodegradability, low cost, and abundant availability [1,7,29,30]. Starch is also the primary source of carbohydrates in tuber and cereal plants, *e.g.* cassava and corn, respectively, comprising two macromolecules, which are amylose and amylopectin. It is extensively used to develop biodegradable film due to the advantages it offers, *e.g.* affordability, availability, and excellent film-forming ability [31,32]. The proportion of linear homopolymer amylose to highly branched amylopectin is dependent on the plant starch source and could influence their processing behaviour and final product characteristics [7].

From the packaging industry perspective, starch-based materials are gaining traction in the bio-based polymers market for a variety of packaging applications. Native starches, on the other hand, are brittle and lack thermoplastic properties. As a result, products derived from native starches crumble easily when dried in environmental conditions. The starches' brittle nature is a result of the strong intermolecular hydrogen bonds between the macromolecular network chains of amylose and amylopectin [27,33,34]. Plasticizers combined with higher temperatures and shear improve the native starches' flexibility to a level comparable to that of conventional thermoplastic polymers [11,35].

Antibacterial packaging is making inroads in the food industry due to its ability to inhibit microbial activity in foods during their processing and storage, consequently, lengthening their shelf life [29,36]. EOs are shown to be effective as preservatives and antibacterials in food, suggesting that they may be a viable substitute to synthetic compounds [1,37]. The majority of the antibacterial substances found in EOs are shown to be beneficial for microbial population control *via* targeting foodborne microorganisms, resulting in higher-quality and safer

food products [38]. Syafiq *et al.* [1] and Mith *et al.* [39] determined the antibacterial performances of commercial EOs derived from cinnamon, eucalyptus, thyme, and rosemary against food-borne pathogenic bacteria and bacteria associated with food spoilage. According to the findings of the aforementioned studies, bioactive components, *e.g.* carvacrol, cinnamaldehyde, eugenol, and thymol, demonstrated significant antibacterial activities against all bacteria tested [39]. The EOs' antibacterial activity is associated with the presence of bioactive volatile components that are classified into two major groups: terpenes and aromatic compounds [40]. Bioactive components may adhere to the surface of the cell and then penetrate the bacterial membranes, impairing the structural integrity and metabolism of the cell [40,41]. The cinnamon (*Cinnamomum zeylanicum* Boiss.) essential oil (CEO), a member of the Lauraceae family native to southern Asia, has the highest antibacterial activity of all EOs of Lauraceae plants [12,42,43]. Zhang *et al.* [44] and Syafiq *et al.* [29] found that the extract solution containing CEO inhibited the growth of *E. coli* and *S. aureus* and revealed high antibacterial activity for biopolymer packaging, respectively.

Currently, there are limited studies conducted on the effects of types and concentration of plasticizers on the films of sugar palm nanocellulose reinforced SPS/CEO bionanocomposite. Thus, this study aims to investigate the effects of different plasticizer types (glycerol, sorbitol, and glycerol/sorbitol) and concentrations (1.5, 3.0, and 4.5 wt%) on the mechanical, physical, and antibacterial characteristics of sugar palm nanocellulose reinforced SPS/CEO bionanocomposite films.

2 Materials and methods

2.1 Materials

SPS and fibre utilized in this study were extracted from sugar palm trunks planted in Kuala Jempol, Negeri Sembilan (Malaysia). Glycerol (99% purity, food-grade) with a density of 1.26 g/cm³ and a molar mass of 92.09 g/mol and D-sorbitol plasticizers (99% purity) with a density of 1.49 g/cm³ and a molar mass of 182.17 g/mol, CEO, and Tween 80 were supplied by Evergreen Sdn. Bhd., Semenyih (Selangor, Malaysia).

2.2 Methods

2.2.1 SPS extraction and preparation

A chainsaw (Stihl, USA) was used to remove SPS from the inside stem of a mature sugar palm tree. Following that,

the starch powder was washed using water that was used to extract the starch from the mixture. The mixture was then filtered through a sieve (<300 µm), with the fibre remaining at the top of the sieve and the starch granules flowing into the container with the water. Separating the starch from water required slow pouring of the water so that the starch would not drain out. The by-product of the process was the fibre that was removed from the wet starch. Next, the wet starch was sun-dried for 30 min following oven drying for 24 h at 120°C [45].

2.2.2 Preparation of sugar palm nanocellulose/SPS/CEO bionanocomposite films

The sugar palm-based films were prepared by the conventional solution-casting method. The plasticizers used were glycerol (G), sorbitol (S), and their 1:1 combination ratio (GS) to study the effect of each plasticizer on SPS films. To begin with, a gelatinized SPS with 10% wt aqueous dispersion was prepared *via* heating of the film-forming solution for 15 min at a temperature of 95 ± 2°C in a hot water bath with constant stirring, a crucial step to disintegrate starch granules to obtain a homogenous solution. Next, different plasticizers at 0, 1.5, 3.0, or 4.5% (w/w, starch basis) were added to the dispersion (Table 1). The solution was further heated for 15 min at 95 ± 2°C. Next, cinnamon EO, SPS nanocellulose, and emulsifier (Tween 80) were added into the dispersions at 2, 0.05, and 0.05 wt%, respectively. Then, the solution was placed in a sonicator (VCX 500-W, Vibra-Cell™, USA) for 30 min using 50% amplitude and 05 pulse to disperse the SPS nanocellulose. Then, the heating process was continued for 15 min at 95°C. Before casting the film-forming solutions in glass Petri dishes, they were allowed to cool to room temperature. The glass Petri dishes were used as the casting surfaces that provided

Table 1: Formulation of plasticizer used in the preparation of bionanocomposite films

Samples	Plasticizer type	Plasticizer concentration (wt%)
SPS	—	0
G1.5	Glycerol	1.5
G3.0	Glycerol	3.0
G4.5	Glycerol	4.5
S1.5	Sorbitol	1.5
S3.0	Sorbitol	3.0
S4.5	Sorbitol	4.5
GS1.5	Glycerol/sorbitol	1.5:1.5
GS3.0	Glycerol/sorbitol	3.0:3.0
GS4.5	Glycerol/sorbitol	4.5:4.5

a smooth and flat surface for the film formation. The newly casted films were dried in an oven (Lab Companion Oven, Model: ON-11E, Korea) (40°C). The preparation of all films was conducted in triplicate. Upon the completion of

drying for 24 h, the films were removed from the Petri dishes and placed at 53 ± 1% relative humidity (RH) in desiccators. Figure 1 shows the flowchart of preparing the bionanocomposite film.



Figure 1: Processing flowchart of the bionanocomposite films.

2.2.3 Film thickness

The film thickness was measured using a digital micrometre (Mitutoyo Co., Japan) with 0.001 mm sensitivity. Five different areas of each film were used to obtain the thickness. The final thickness was recorded as the value of the mean measurements of each film.

2.2.4 Density and physical appearance

A densimeter (Mettler-Toledo (M) Sdn. Bhd., Shah Alam, Selangor, Malaysia) was employed for the density measurement of the prepared films. The physical appearance of the bionanocomposite films was evaluated based on their transparency, rigidity, brittleness, flexibility, and difficulty in peeling the bionanocomposite films. Xylene was used as the immersing liquid to substitute distilled water to prevent uptake of water by the hydrophilic films. Additionally, the liquid must have a lower density than the film to prevent the film from floating, making xylene the best option. Each film sample was weighed (m) prior to the xylene immersion and the volume of xylene displaced after the immersion was denoted V . The test was conducted in quadruplicate for each sample. The density (ρ) was calculated as follows:

$$\rho = m/V. \quad (1)$$

2.2.5 Water content

The term “water content” refers to the amount of water contained within a material. The water uptake capacities of the films were determined in accordance with ASTM D 644 [46]. The initial film weight (W_i) was recorded and the film was oven-dried for 24 h at a temperature of 105°C. Upon completion, the dried sample was reweighed, obtaining the final weight (W_f). The weight loss percentage from the drying process was determined using equation (2). The mean value of the water content from 10 specimen sets was calculated.

$$\text{Water content (\%)} = [(W_i - W_f)/W_i] \times 100. \quad (2)$$

2.2.6 Mechanical properties

All samples were stored in a climate-controlled room at a temperature of $23 \pm 2^\circ\text{C}$ and RH of $53 \pm 1\%$ for 72 h or until a steady weight within 2% weight changes was obtained. The tensile properties of the samples were determined using an Instron 3365 universal testing machine (High

Wycombe, England) equipped with a 30 kg loading cell. The elongation at break and the tensile strength of 10 mm \times 70 mm samples were determined in accordance with the standard technique of ASTM D 882-02 [47]. A 2 mm/min crosshead speed was used to pull the film strips. The average measurement value from 10 film samples was calculated.

2.2.7 Antibacterial activity using disc diffusion method (DDM) and agar disc method (ADM)

Three samples were chosen for plastic packaging after considering the optimum elongation at break, the tensile strength and modulus, and low water content and density. This test was carried out in triplicate. The antibacterial activity was determined using two bacteria strains: (a) Gram-positive *Bacillus subtilis* B29 (*B. subtilis*) and (b) Gram-negative *Escherichia coli* ATCC 2592 (*E. coli*). Both of the strains were supplied by the Laboratory of Molecular Biomedicine, Institute of Bioscience, Universiti Putra Malaysia, Serdang, Malaysia. The antibacterial activity of the films was determined using ADM, in which films were punched into 6 mm size using a sterile paper puncher. After that, the films were immersed in 0.5 mL of McFarland broth standard agar at a concentration of 1×10^8 CFU/mL. All samples were cultured in a variety of microbes that completely covered the plate's surface. The plates were then inverted and incubated at 37°C for 24 h. After the incubation was completed, the inhibition zone was measured.

The CEO was tested with two other types of EOs, rosemary, and eucalyptus, to determine the inhibition zone using DDM. About 20 μL of the EO was dropped onto a 6 mm paper disc to inhibit the two types of bacteria, *B. subtilis* and *E. coli*. The disc was then placed onto the plate that was spread with the bacteria. The plates were incubated for 24 h at a temperature of 37°C. The inhibition zones were photographed and the diameters were measured.

2.2.8 Statistical analysis

Duncan's multiple range test was employed to perform the comparison of the mean at a 0.05 significance level ($p < 0.05$) by using SPSS software to determine the analysis of variance (ANOVA) on the experimental findings. The test was conducted in triplicate unless otherwise stated. The significance of the added EOs based on the inhibition area was determined using ANOVA.

3 Results and discussion

3.1 Film thickness

Figure 2 shows the increase in the film thickness from 0.118 to 0.206 mm, 0.104 to 0.204 mm, and 0.12 to 0.202 mm, resulting from the increase in G, S, and GS concentrations from 1.5 to 4.5 wt%, respectively. Increasing the plasticizer concentration resulted in a considerable increase in the film thickness, depending on the plasticizer type. This could be due to the involvement of plasticizers in dissolving and structuring intermolecular polymer chain networks, resulting in greater free volumes and thus thicker films. A similar plasticizer concentration effect on the film thickness was described by Sanyang *et al.* [7], Syafiq *et al.* [1], Razavi *et al.* [48], Hazrol *et al.* [49], and Ilyas *et al.* [27]. Furthermore, using different plasticizer types revealed a significant effect on the film thickness, as presented in Figure 2. S-plasticized films were thicker than G-plasticized films, while GS-plasticized films were the thickest for a concentration of 1.5 wt%. The changes in the film thickness using various plasticizers could be attributed to their molar masses since the formulation of the film-forming solution was constant. G-plasticized (molar mass 92.09 g/mol) films possessed a lower film thickness than the S-plasticized (molar mass 182.17 g/mol) films at a constant concentration, which could be due to the lower molar mass of the G-plasticizer. Sanyang *et al.* [7], Aitboulahsen *et al.*

[6], and Ghasemlou *et al.* [50] also found that S-plasticized films produce higher thickness than the G-plasticized films.

3.2 Density of bionanocomposite films

Figure 3 shows the effects of the plasticizer type and concentration on the density of SPS films incorporated with the CEO. Increasing the concentration of plasticizers from 1.5 to 4.5 wt% caused a partial decrease in the density of S- (2.085–1.864 g/cm³), G- (2.038–0.990 g/cm³), and GS-plasticized films (2.233–1.822 g/cm³). It was clear that increasing the amount of plasticizers from 1.5 to 4.5 wt% decreased the density of the films significantly regardless of the plasticizer type. The findings of G-plasticized films were aligned with those described by Sanyang *et al.* [7], Sahari *et al.* [26], and Hazrol *et al.* [49], who plasticized SPS with glycerol, sorbitol, and glycerol/sorbitol at a 1:1 ratio.

The density values amongst the various plasticizer types did not show much of a difference. However, at the same plasticizer concentration, the order of density reduction is as follows: GS > S > G-plasticized films. The changes in the density and molecular weight of the plasticizers might be ascribed to this phenomenon. The molecular weight of the plasticizers followed a similar reduction pattern with density. Thakur *et al.* [51], who researched the factor that affected the properties of the starch-based film,

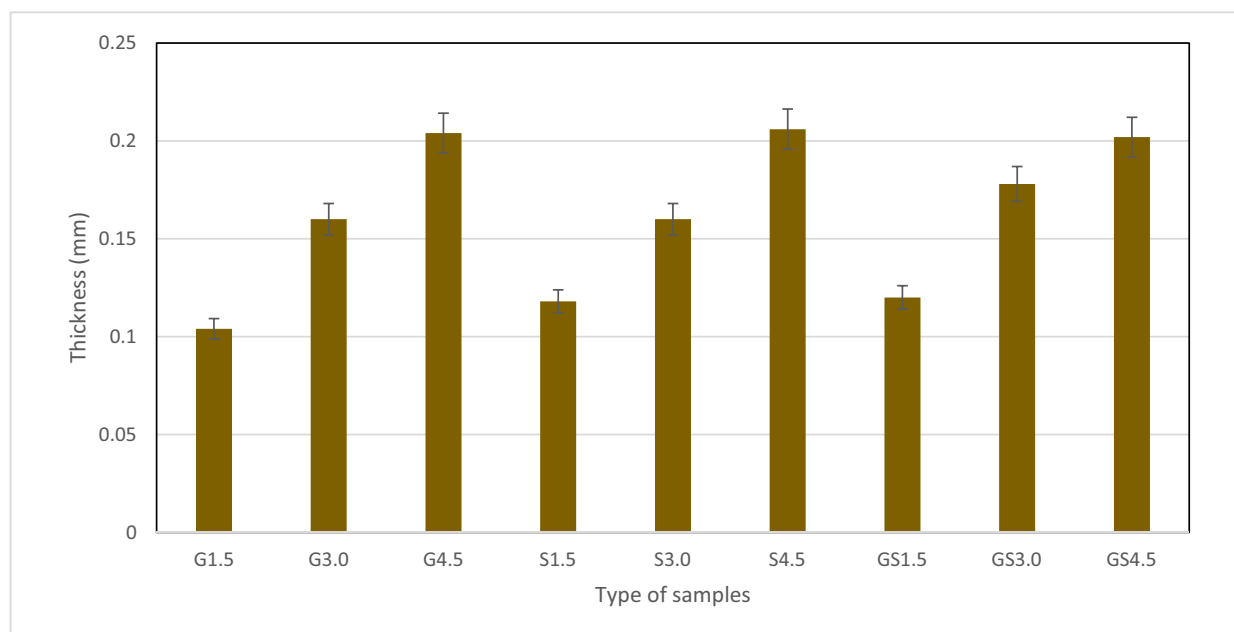


Figure 2: Thickness of the samples.

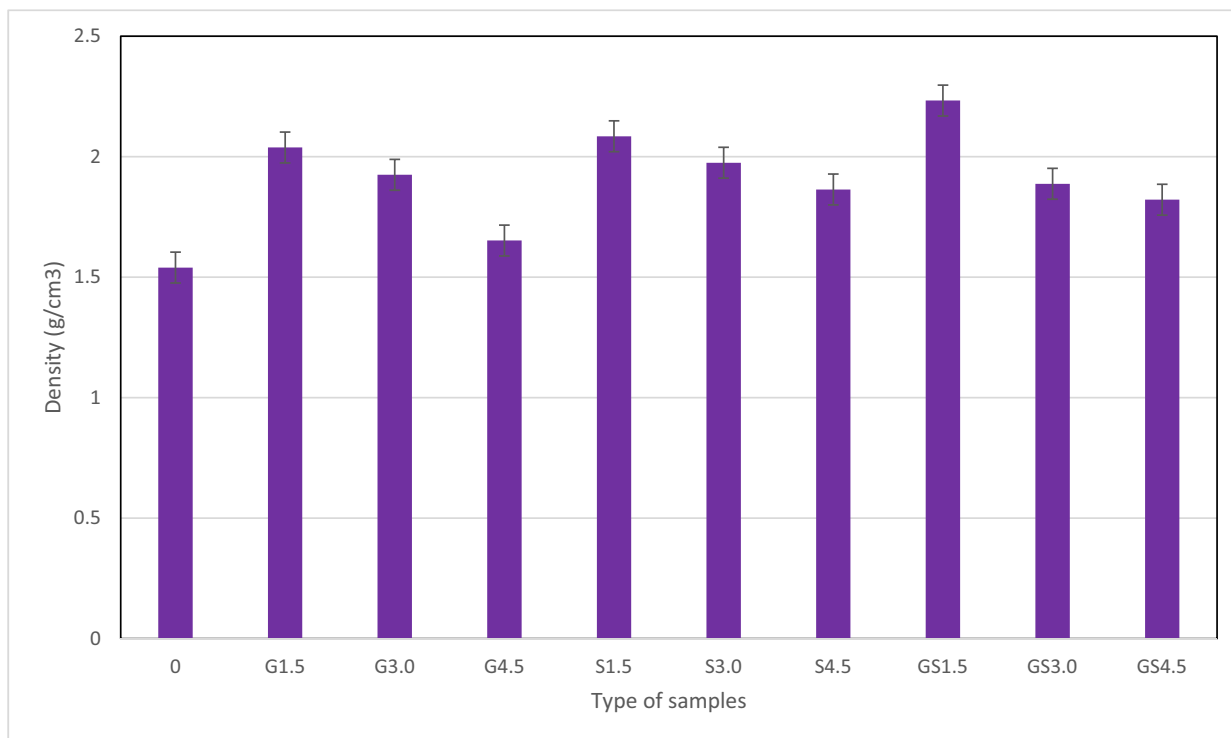


Figure 3: Density of bionanocomposite films.

reported that increasing the content of glycerol in starches slowed crystallization kinetics, resulting in low density.

Figure 4a and b and Table 2 show the images and describe the visual appearance of the obtained bionanocomposite films with and without a plasticizer. It was observed that all films were slightly brownish in colour and clearly transparent (Figure 4a and b). Bionanocomposite films without plasticizers were wavy, brittle, fragile, and rigid (Figure 4a). Many cracks were also observed on the surface of bionanocomposite films without a plasticizer (Figure 4a). It was also hard to be peeled off and handle because it broke into small pieces. Strong inter/intramolecular hydrogen bonds of SPS provided less mobility to the macromolecular chains, causing the film to be cracked, rigid, and brittle [7,27]. This result was in agreement with Hazrati *et al.* [52], Hazrol *et al.* [49], and Tarique *et al.* [53], who studies bionanocomposite films from *Dioscorea hispida*, corn, and arrowroot (*Maranta arundinacea*) starches, respectively.

The use of plasticizers altered SPS films to become more flexible with smooth and homogenous surfaces (Figure 4b). SPS films with 4.5 wt% plasticizers were found to be more flexible than the film with a lower plasticizer concentration. The flexibility reduced as the plasticizer concentration decreased. The low molecular weight of plasticizers allowed them to creep into the polymer chains' intermolecular gaps, weakening intermolecular hydrogen

bonds and enhancing molecular mobility. As the plasticizer concentration increased from 1.5 to 4.5 wt%, the intermolecular hydrogen bonds of SPS films deteriorated. Nevertheless, changes in the plasticizer type had a considerable impact on the flexibility of their resultant films at similar plasticizer concentrations. Consequently, GS-plasticized bionanocomposite films revealed higher flexibility than G- and S-plasticized films. Figure 4c. shows the mechanism of reinforcement and the role of plasticizers in bionanocomposite films.

3.3 Moisture content

Figure 5 shows the significant moisture content increase as the plasticizer concentration was increased from 1.5 to 4.5 wt%, except for the S-plasticized films. In general, as the concentration of the plasticizer increases, starch-based films become more hydrophilic. Thus, several studies have demonstrated that increasing the amount of the plasticizer in hydrocolloid films increased the moisture content [7,50]. Even so, sorbitol had a lesser influence on the moisture content of SPS films for the S-plasticized films. Figure 5 illustrates the stable moisture content of G- and GS-plasticized films as the plasticizer concentrations were increased, which was in line with the findings from Sanyang *et al.* [7]

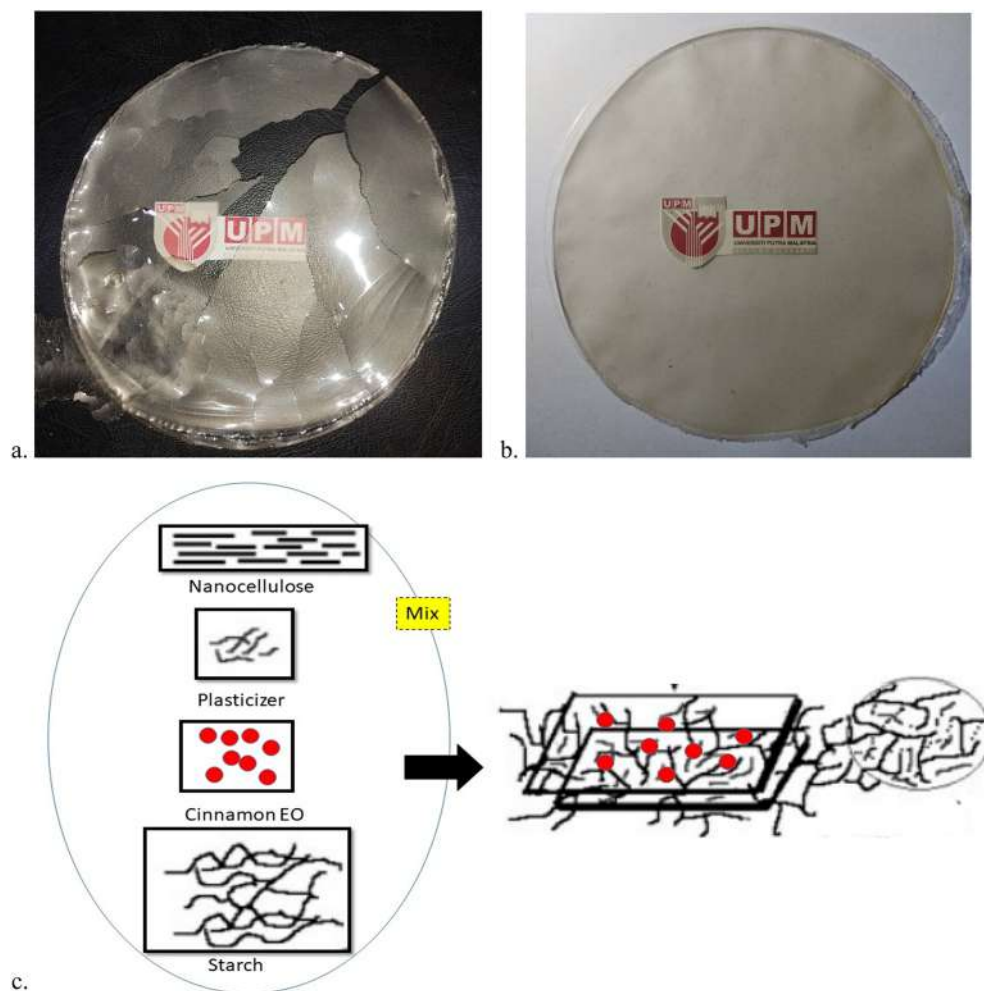


Figure 4: (a) SPS film without a plasticizer and (b) sugar palm bionanocomposites with a plasticizer. (c) Mechanism of reinforcement and the role of plasticizers.

and Aguirre *et al.* [54]. The lower moisture content of S-plasticized films in comparison to glycerol-containing films (e.g. G-and GS-plasticized films) could be explained by the high similarity in the molecular structure of glucose units to sorbitol, resulting in stronger molecular interactions

between the intermolecular polymer chains and sorbitol. As a result, the possibility for sorbitol interaction with water molecules had decreased. On the other hand, glycerol's hydroxyl groups had a strong affinity for water molecules, allowing glycerol-containing films to easily hold water by

Table 2: Description of bionanocomposite films with and without a plasticizer

Samples	Description of films
SPS	Clear transparent, cracked, broke into pieces, brittle and fragile, difficult to peel, and rigid
G1.5	Clear transparent, not fragile, not brittle, no cracks, flexible, not sticky, and peelable
G3.0	Clear transparent, more flexible than G1.5, and slightly sticky
G4.5	Clear transparent, more flexible than G3.0, slightly elastic, sticky, and easy to peel
S1.5	More transparent than G1.5 and GS1.5, brittle and fragile, less flexible than G1.5, not sticky, and slightly difficult to peel
S3.0	More transparent than G3.0 and GS3.0, not brittle and fragile, more flexible than S1.5 but less than G3.0, not sticky, and peelable
S4.5	More transparent than G4.5 and GS4.5, not brittle and fragile, flexible, not sticky, peelable, and easy to handle
GS1.5	Transparent, not brittle and fragile, not rigid, flexible, peelable, and not sticky
GS3.0	Transparent, more flexible than G3.0, and stickier than G3.0
GS4.5	Transparent, more flexible than G4.5, and stickier than G3.0 and G4.5

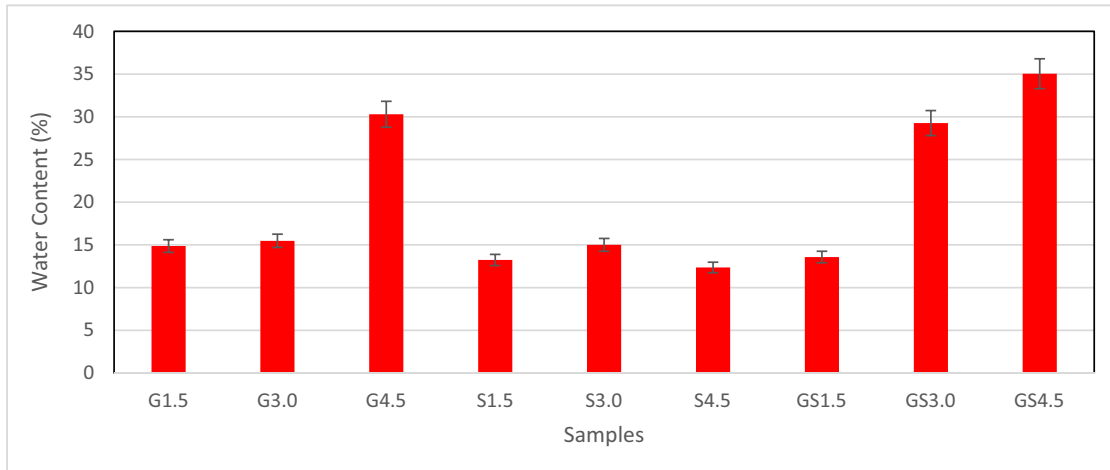


Figure 5: Water content of the samples.

forming hydrogen bonds within their matrix [7,55]. As a result, glycerol functions as a water-binding agent, whereas sorbitol has a lower affinity for water molecules.

3.4 Mechanical properties

Figures 6 and 7 and and 8 illustrate the mechanical properties of G-, S- and GS-plasticized films, respectively. A lower plasticizer concentration of 1.5 wt% resulted in high tensile strength of 12.37 MPa for G-plasticized

films, 12.26 MPa for S-plasticized films, and 8.75 MPa for GS-plasticized films. The high tensile strengths at low plasticizer concentrations could be explained by the dominance of stronger hydrogen bonds established by a starch–starch intermolecular interaction over starch–plasticizer attraction. However, adding plasticizers at concentrations ranging from 1.5 to 4.5 wt% resulted in a substantial decrease in the films' tensile strength regardless of the type of plasticizer. The tensile strength of the G-plasticized films decreased significantly from 12.37 to 2.20 MPa, while that of the S-plasticized films decreased from 12.26 to

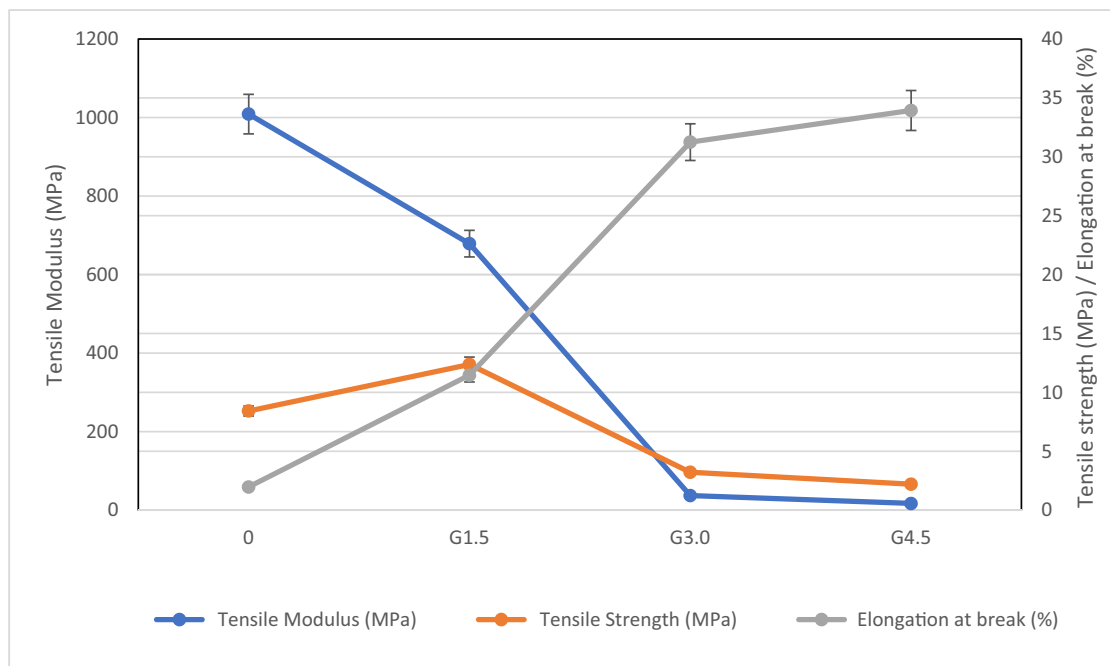


Figure 6: Mechanical properties of G-plasticized films.

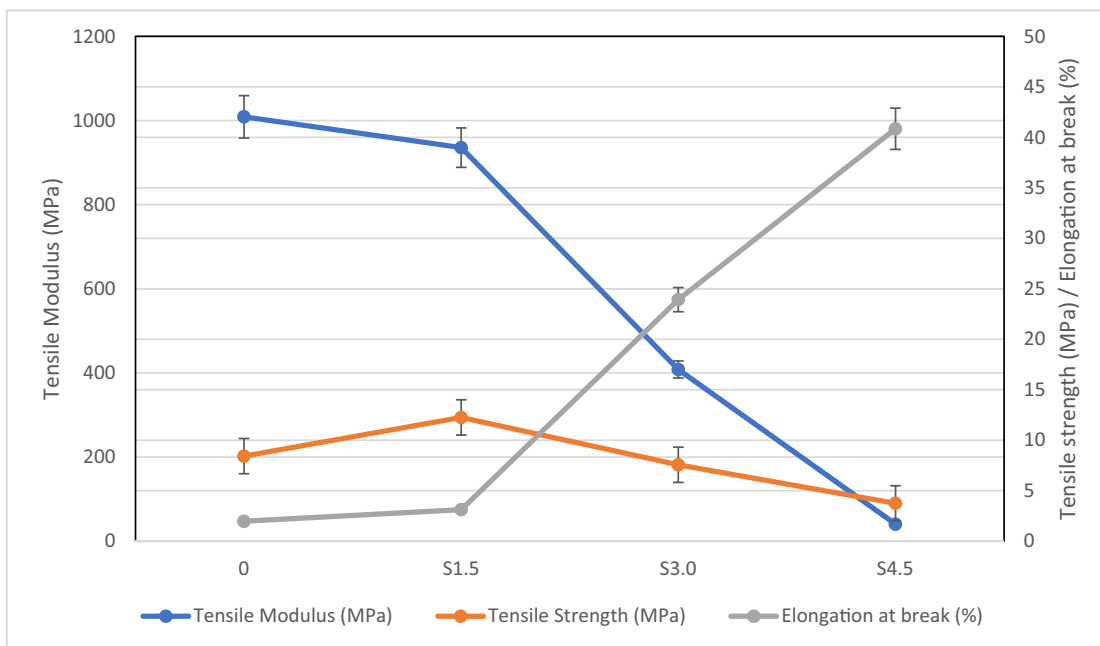


Figure 7: Mechanical properties of S-plasticized films.

2.20 MPa as the plasticizer concentration was increased from 1.5 to 4.5 wt%. The films' tensile strength decreased from 8.75 to 0.83 MPa in the case of GS-plasticized films at the same plasticizer concentration range. Numerous authors reported a decrease in the tensile strength of starch-based films as the plasticizer concentration increased [11,56,57].

This phenomenon was a result of the plasticizers' role in weakening the strong intramolecular attraction between the starch molecules chain, thereby promoting the hydrogen bond formation between starch molecules and plasticizers. Thus, it weakened the hydrogen bonds between starch chains, lowering the tensile strength of SPS plasticized films [11,58].

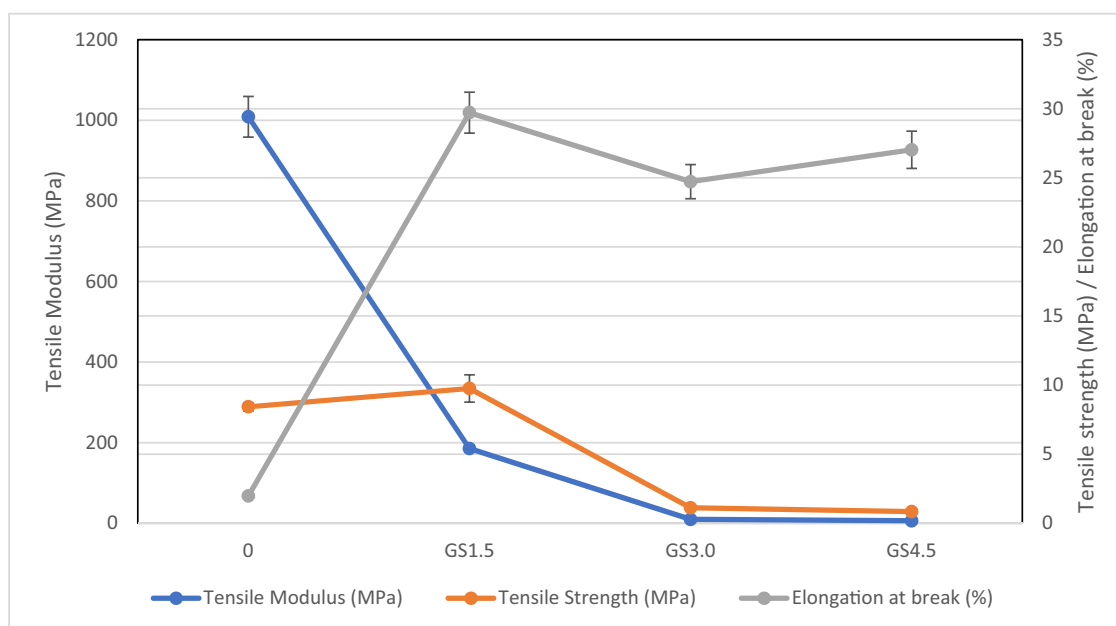


Figure 8: Mechanical properties of GS-plasticized films.

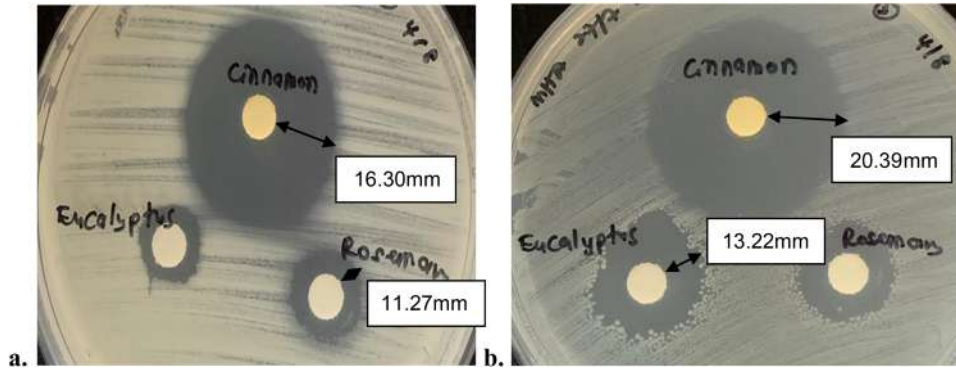


Figure 9: (a) Image of inhibition zone against *B. subtilis* and (b) inhibition zone against *E. coli*.

When the concentration of glycerol was increased from 1.5 to 4.5 wt%, the G-plasticized films demonstrated the highest decrease in the tensile strength than S- and GS-plasticized films. These findings indicated that glycerol was more effective at plasticizing SPS films than sorbitol. Sanyang *et al.* [11], Razavi *et al.* [48], and Muscat *et al.* [57] reported parallel findings that glycerol caused a greater decrease in the tensile strength than other polyols. This tendency could be associated with the a lower molar mass of 92.0928 g/mol of glycerol compared to sorbitol of 182 g/mol that facilitated easier glycerol–starch molecular chains interaction. Sanyang *et al.* [7] Tapia-Blácido *et al.* [59] also stated that a more effective plasticizer for the majority of edible films is glycerol.

The extendibility of a film's length from its starting length to the breaking point is known as elongation at break, which is also known as the films' ability to deform before breaking [11,60]. This parameter ($E\%$) is used to calculate a film's flexibility and stretchability. Flexibility requirements for biopackaging films vary according to their intended use and subsequent transit, handling, and storage of packed food products. The effect of plasticizer concentration (1.5–4.5 wt%) on the elongation of SPS plasticized films was inversely proportional to the films' tensile strength. As expected, increasing the plasticizer concentration from 1.5 to 4.5 wt% resulted in a remarkable increase in film elongation: 11.46–33.94% for G-plasticized films and 3.12–40.84% for S-plasticized films. Meanwhile, GS-plasticized film experienced a slight reduction from 29.72 to 27.02%. Similar film elongation phenomena were reported by Sanyang *et al.* [11], Kurt and Kahyaoglu [61], and Suppakul *et al.* [62]. The observed increase in the film elongation might be due to plasticizers weakening the intermolecular connections between amylose, amylopectin, and amylose–amylopectin molecules in the starch matrix and substituting hydrogen bonds established between plasticizer and starch molecules.

The disruption and reconstruction of starch molecular chains might reduce the rigidity and promote films' flexibility by allowing more chain mobility.

The Young's modulus in Figures 6–8 shows the significant film stiffness' determinant. The stiffness of a bionanocomposite film is proportional to its Young's modulus; the high modulus is associated with high stiffness. Films with a higher concentration of the plasticizer showed lower Young's moduli, which showed lower stiffness. Previously, several studies observed a decrease in stiffness when the hydrophilic concentration of the plasticizer was increased in the films [49,52,63]. This phenomenon could be explained by the changes in the structure of the starch network as plasticizers were added, causing the film matrix to become less compact [11,53]. This also clarified the considerable decrease in the tensile strength and modulus observed in all bionanocomposite films as plasticizer concentrations were increased from 1.5 to 3.0 wt%, with a maximum of 4.5 wt%.

3.5 Antibacterial activity using DDM and ADM

Antibacterial activity testing is required to study membrane activity against pathogen bacteria for various applications. To make a comparison of the effectiveness of the CEO, two

Table 3: Data of inhibition zone for 3 EOs against *B. subtilis* and *E. coli*

EOs	Test strain	
	Inhibition zone (mm)	
	<i>B. subtilis</i> (B29) (a.)	<i>E. coli</i> (ATCC25922) (b.)
Cinnamon	16.30	20.39
Rosemary	11.27	11.35
Eucalyptus	10.44	13.22

Table 4: Data of inhibition zone for three selected bionanocomposite films incorporated with CEO

Sample(s)	Test strain	
	Inhibition zone (mm)	
	<i>E. coli</i> (ATCC25922)	<i>B. subtilis</i> (B29)
1 (G1.5)	6.82	3.35
2 (S1.5)	6.83	3.32
3 (GS1.5)	6.85	3.34

other EOs (rosemary and eucalyptus) were also tested for the inhibition zone. In Figure 9a (*B. subtilis*) and 9b (*E. coli*) show the antibacterial effect of the EOs towards the *B. subtilis* and *E. coli* bacteria. The presence of phenolic (cinnamaldehyde) compounds has been linked to EOs' antibacterial action [41]. About 20 μ L of CEO could inhibit *E. coli* and *B. subtilis* bacteria resulting in 20.39 and 16.30 mm inhibition diameter, on average, respectively (Table 3). The CEO showed the highest inhibition activity compared to rosemary and eucalyptus EOs. The various inhibitory effects of EOs might be due to the biological characteristics of their major constituents. The most important constituent in CEO was cinnamaldehyde while the major constituents in eucalyptus and rosemary EOs were 1,8-cineol/ α -pinene and 1,8-cineol/ α -pinene/camphor, respectively.

Three samples of bionanocomposite films were chosen to test the inhibition zone, which were the mixture of G1.5, S1.5, and GS 1.5. The antibacterial performance of the films and the EOs were evaluated against *B. subtilis* and *E. coli* via ADM and DDM. Table 3 shows the inhibition zone of bionanocomposite films for *B. subtilis* and *E. coli*. Table 4 presents the inhibition zone of the films on *B. subtilis* and *E. coli*. According to the different types of plasticizers that were chosen, all samples showed no significant change of their inhibition zones, which were in the range of 6.82–6.85 mm for *E. coli* and 3.32–3.35 mm for *B. subtilis*. The antibacterial activity of bionanocomposite films was not significantly affected by the various types of plasticizers, which showed the same range of inhibition as showed by Syafiq *et al.* [1], Ghoshal *et al.* [64] and El Fawal *et al.* [65], who researched bionanocomposites incorporated with cinnamon, eucalyptus, and rosemary EOs, respectively.

4 Conclusion

The effects of various plasticizer types and concentrations on the physical, mechanical, and antibacterial properties of sugar palm nanocellulose/SPS/CEO bionanocomposite films were studied. SPS films were brittle, with many

visible cracks, and were hard to be peeled off the casting surfaces without a plasticizer. Therefore, the introduction of plasticizers aided to overcome the brittleness and improve the flexibility and peelable ability of SPS films. The findings revealed that the concentration and type of the plasticizer influenced the density, thickness, tensile strength, moisture content, surface structure, and elongation at the break of the films. The antibacterial activity of the bionanocomposite films was not significantly influenced by the type of the plasticizer, which showed that the effectiveness of the inhibition zone was maintained. Gradual increase in the plasticizer concentration from 1.5 to 4.5 wt% decreased the films' density but increased the moisture content and thickness of the films, irrespective of the plasticizer type used. Overall, GS-plasticized films demonstrated the best performance in terms of physical and mechanical properties. The plasticizing effect of various concentrations of the plasticizer was attributable to the weakening of hydrogen bonds between the starch intermolecular chains caused by the development of starch (amylose)–plasticizer complexes. At a concentration of 4.5 wt% plasticizer, an antiplasticization effect was found for the G- and GS-plasticized films, whereas plasticization behaviour was observed at lower concentrations of the plasticizer. Fascinatingly, GS-plasticized films improved the G-plasticized films' tensile strength while reducing S-plasticized films' brittleness. Sugar palm nanocellulose/SPS/CEO bionanocomposite films plasticized using a mix of glycerol and sorbitol achieved optimum physical, mechanical, and antibacterial performance. However, the effects of different plasticizer types and concentrations on the solubility, water absorption, water vapour permeability, and barrier characteristics of SPS-based films should be investigated to discover the optimal combination to develop biodegradable food packaging films.

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