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RESEARCH ARTICLE

Development and characterization of *Caesalpinia pulcherrima* seed gum-based films to determine their applicability in food packaging

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Abstract

Investigation of potential biopolymers for developing edible films as alternatives for plastic packaging is a recent trend. Among the biopolymers used in edible film making, galactomannan is a good film-forming agent. *Caesalpinia pulcherrima* seed gum is a rich source of galactomannan and could be used as an edible film making material. The main objective of this study was to identify the ability of *Caesalpinia pulcherrima* seed gum in making a food packaging film and assessing its performance. In this study, the gum was extracted from *Caesalpinia pulcherrima* seeds and dissolved in distilled water along with the plasticizer glycerol. In preparing film-forming solutions, a previously selected range of glycerol concentrations (0.0, 0.5, 1.0 and 1.5%) was used. After casting and drying, the resultant films were analyzed for their physical and mechanical properties. In the present study, the film prior to incorporating glycerol was brittle, less flexible and had a strong film matrix. With the increase of glycerol content, the film became more flexible, sticky and weak. Further, the physical properties namely thickness, moisture content, swelling index, water solubility of *Caesalpinia pulcherrima* seed gum-based films were increased from 0.039 ± 0.001 to 0.076 ± 0.001 mm, from 62.92 ± 0.51 to 69.40 ± 0.15%, from 5.39 ± 0.17 to 8.45 ± 0.17, from 55.69 ± 0.51 to 66.66 ± 0.45%, respectively, with the increase of glycerol content from 0.0 to 1.5%. Concerning the mechanical properties, the tensile strength and Young's module were decreased from 10.90 ± 0.08 to 2.11 ± 0.05 MPa and from 48.46 ± 0.24 to 3.47 ± 0.09 MPa, respectively, and the elongation was increased from 22.50 ± 0.05 to 60.84 ± 0.04% with increasing the glycerol content from 0.0 to 1.5%. Due to its physical and mechanical properties, *Caesalpinia pulcherrima* seed gum is a potential source for edible packaging film manufacture.

Keywords Food packaging · Edible films · *Caesalpinia pulcherrima* · Seed gum · Galactomannan

1 Introduction

Synthetic polymers known as plastics are the most common and persistent pollutant in the environment which has become a global threat (Moore 2008). Rigid and flexible plastic has gained a high market share within the worldwide packaging although their waste production is growing at a rate of 4.2% since 2010 and is expected to continue till 2024 (Ketelsen et al. 2020). As 73% of worldwide packaging accounts for food and beverage packaging (Ketelsen et al. 2020), there is a need to focus actions to minimize the packaging waste from this sector. Therefore, the development of biodegradable packaging materials from renewable sources has gained much attention in recent years. These biodegradable polymers are degraded by microorganisms through enzymatic catalysis ending up mainly has CO_2, CH_4 and water (Zhong et al. 2020). With the increasing demand for these biodegradable polymers in making environmentally friendly packaging materials, polymers of natural origin such as polysaccharides, proteins and lipids have gained much attention recently due to their applicability. The desirable properties of these edible polymers lead them to be applied not only in the food industry as a packaging material but also in the cosmetic and biomedical industries (Kouhi et al. 2020).

Among these biopolymers, natural gum and mucilage, known as plant hydrocolloids have attracted attention. Because of their low cost, availability, unique rheological properties and structural variations they are uniquely suitable

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for "green" material applications such as edible or biodegradable food packaging (Ahmad et al. 2019). The gum and mucilage obtained from these sources such as guar (Dhumal et al. 2019), tara (Chen et al. 2020), locust bean (Kurt et al. 2017), basil (Khazaei et al. 2014), cress (Jouki et al. 2013a), fenugreek (Salarbashi et al. 2019), cassia (Cao et al. 2018), quince (Jouki et al. 2013b) and chia (Urbizo-Reyes et al. 2020) seeds are currently being used or studied for potential edible film manufacture. Among these sources, guar, tara, locust bean and fenugreek seeds are considered the main sources of galactomannan (Kontogiorgos 2019) which is a thickening, stabilizing and emulsifying agent that has been already identified as a preferable alternative compound for edible film formation (Cerqueira et al. 2011). Galactomannan is identified as the major compound in *Caesalpinia pulcherrima* seed gum (Medeiros et al. 2020) which consists of a β-(1–4)-D-mannan backbone with α-(1–6) D-galactose molecule substitution at O-6 position of D-mannopyranosyl residues with a mannose to galactose ratio of 2.9:1.0 (Dos Santos et al. 2015; Wei et al. 2015; Mendes et al. 2017; Rodriguez-Canto et al. 2020).

Caesalpinia pulcherrima which is known as Pride of Barbados (Fig. 1) is in the family Fabaceae and the subfamily, Caesalpinioideae. This plant, native to Central America however is currently, widely distributed and widely grown in sub-tropical areas such as Myanmar, Brazil, Malayan, Peninsula, India, Sri Lanka, and Vietnam (Thombre and Gide 2013; Buriti et al. 2014; Marques et al. 2019). The plant is a small, perennial, well-branched tree that flowers with three colour varieties as pink, yellow and orange. The seeds (Fig. 2) which are in pods contain gum which accounts for 27% galactomannan of the seed mass that can be applied in food as a texture modifier, hydrocolloid and as a source of dietary fiber (Buriti et al. 2014; Medeiros et al. 2020). The ability of *Caesalpinia pulcherrima* seed gum to produce a highly viscous and stable solution leads to a widening of its applicability in film formation (Thombre and Gide 2013). Therefore, this study focused on developing an edible film from *Caesalpinia pulcherrima* seed gum with the aim of applying it in food packaging which may be beneficial in minimizing environmental pollution.

Fig. 1 *Caesalpinia pulcherrima* – a flowering tree

Fig. 2 *Caesalpinia pulcherrima* – seeds

2 Material and methods

2.1 Material

Pods of *Caesalpinia pulcherrima* were collected from selected plants in Gampaha, Sri Lanka (7.0840° N, 80.0098° E). All other chemicals and glycerol were obtained from Sigma Aldrich (St. Louis, MO, USA).

2.2 Extraction of *Caesalpinia pulcherrima* seed gum

Gum extraction from *Caesalpinia pulcherrima* seed was carried out based on a method described by Buriti et al. (2014) with some modifications. *Caesalpinia pulcherrima* seeds were removed from pods, cleaned, and mechanically ground using an AC400 grinder (Panasonic, Osaka, Japan). The endosperms were separated manually and soaked in 95% ethanol (endosperms to ethanol ratio was 1:3 v/v) for 15 min at 70 °C in a laboratory water bath (Memmert, Schwabach, Germany) in order to inactivate enzymes and eliminate low molecular weight compounds. After removing ethanol, distilled water was added to the endosperms (endosperm to distilled water ratio was 1:5 v/v) and kept overnight at room temperature. Thereafter, the volume of distilled water was increased to achieve a seed: water ratio of 1:10 and blended for five minutes in a laboratory blender (Panasonic, Osaka, Japan). The viscous solution obtained after blending was filtered through a muslin cloth and the gum was precipitated by adding 95% ethanol (solution to ethanol ratio was 1:2 v/v). The precipitate was washed with acetone, dried in a DZ-1BCII vacuum oven (Hinotek, Ningbo, China), mechanically ground and stored in sealed plastic bags for the subsequent use in the study.

2.3 Film formation

The film-forming solution was prepared by dissolving powdered *Caesalpinia pulcherrima* seed gum (1.0%) and glycerol (0.0, 0.5, 1.0 and 1.5, w/w%) in distilled water. The two ingredients were properly mixed in distilled water by mechanically stirring at 700 rpm for 1 h at room temperature (28–30 °C) using a hotplate stirrer (Lab Companion, Daejeon, Republic of Korea). Thereafter, the biopolymer solution was heated up to 80 °C and the temperature was maintained for 10 min, and allowed to cool to room temperature. The solution was degassed under vacuum and cast was done on Petri dishes (10.0 cm diameter) by pouring 35.0 ml of the solution (per Petri dish) and vacuum drying at 60 °C for 8 h.

2.4 Film characterization

2.4.1 Determination of physical properties

The color parameters of the films were measured with an LC100 colorimeter (Lovibond, Tintometer Ltd, UK) which was first calibrated with a white standard plate and colour reported as L*, a*, b*.

The film thickness was measured using a micrometre screw gauge (CH.MM.25D, China) with 0.001 mm accuracy. The thickness of six randomly selected locations from each film was measured and also four films from each treatment were used to measure the thickness.

The moisture content of films was determined using the oven drying method as described by Chen et al. (2020) with some modifications. The films were cut into 2 × 2 cm pieces and all film samples were weighed (W_0). Thereafter, the samples were dried at 105 °C until constant weight, cooled to room temperature in a desiccator and reweighed (W_1); this was repeated until constant weight. The moisture content (%) was calculated using the following equation:

$$Moisture\ Content = [(W0 - W1)/W0] \times 100$$

The swelling index of the films was determined using the method described by Chen et al. (2020) with some modifications. The films were cut into 2 × 2 cm pieces and all film samples were weighed (W_0). Thereafter, the samples were immersed in distilled water (50 ml) for 24 h, after which excess water was removed by filtering through Whatman 1 filter paper and the samples were reweighed (W2). The swelling index was calculated using the following equation.

$$Swelling\ Index = (W2 - W0)/W0$$

The water solubility of the films was determined according to a method described by Zareie et al. (2020) with some modifications. Films were cut into 2 × 2 cm pieces, weighed to the nearest 0.0001 g and immersed in 50 ml of distilled water for 6 h at room temperature while agitating continuously. The residual films were dried at 105 °C to a constant weight (W_3) and recorded. The initial dry weights of film samples were obtained by drying 2 × 2 cm films at 105 °C for 24 h and reweighing (W_1). The water solubility (%) of films was calculated using the following equation:

$$Water\ Solubility = [(W1 - W3)/W1] \times 100$$

2.4.2 Determination of mechanical properties

Mechanical properties of the films were determined according to the method described by Zareie et al. (2020) and were carried out using an HT-7 T Tensile tester (Hem Techsys Pvt Ltd, Vadodara, India).

2.5 Statistical analysis

The quantitative data were presented as mean ± SD of the replicated determinations ($n = 4$) and significant differences ($p < 0.05$) determined via ANOVA on Minitab 16 software. The means were compared using Tukey's multiple comparison test at a significance of 95%.

3 Results and discussion

3.1 Physical properties of films

The concentration of glycerol to be used as the plasticizer for *Caesalpinia pulcherrima* seed gum-based edible film (Fig. 3) preparation was determined based on the preliminary trials. The film formed without adding the plasticizer was brittle. To achieve a good film network, mixing plasticizers with the polysaccharide is necessary. However, the increase in the plasticizer content resulted in more flexible, transparent films with smooth surfaces owing to the plasticizers capability to reduce the intermolecular forces and enhance the mobility of polymeric chains (Cao et al. 2018). Although water molecules are also considered as a good plasticizer, its function is limited as it can be dehydrated at low relative humidity conditions. Hence, the incorporation of plasticizers such as glycerol or sorbitol is important in making film matrices with excellent packaging applicability

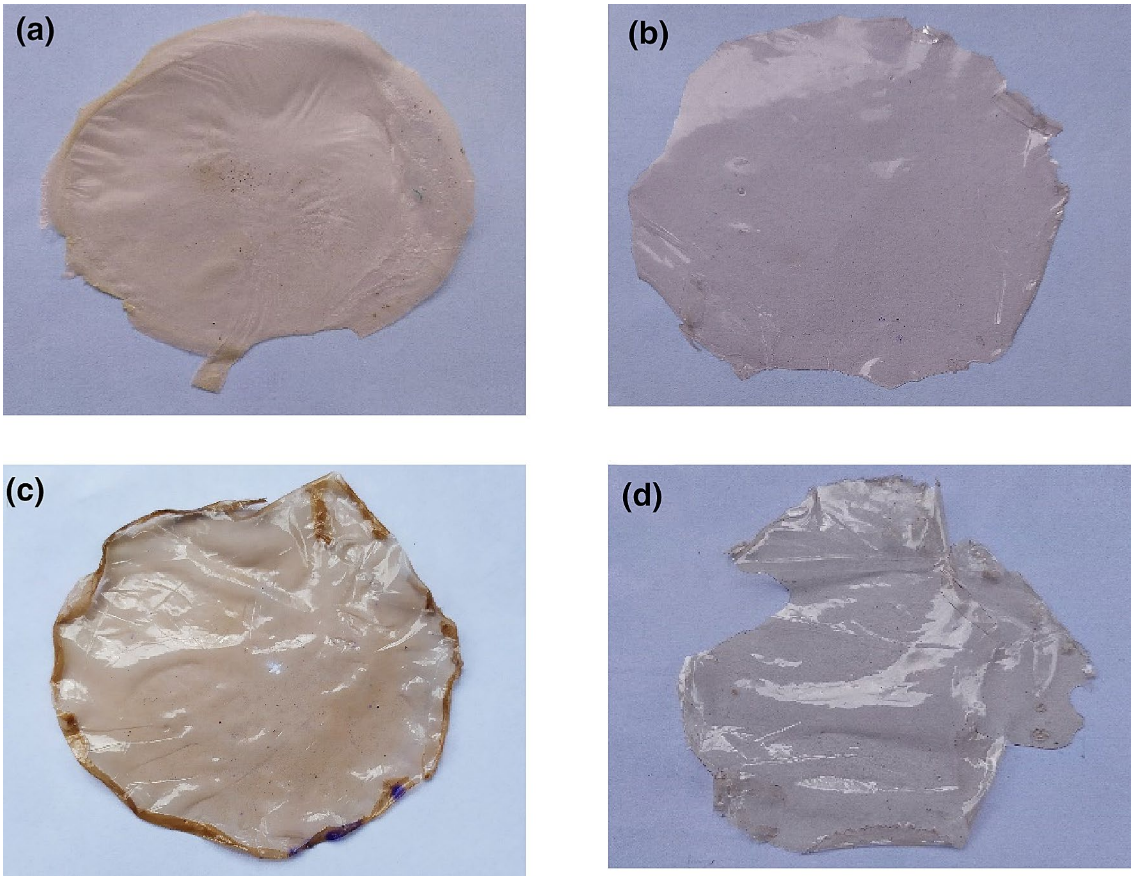

Fig. 3 *Caesalpinia pulcherrima* seed gum-based edible films plasticized with different contents of glycerol—0.0% (**a**), 0.5% (**b**), 1.0% (**c**), 1.5% (**d**)

(Zhang et al. 2016). According to the physical appearance of resultant films (Fig. 3), the film prepared by adding 0.5% glycerol imparted the most appropriate properties for a packaging film. However, further increase of plasticizer content may result in a weaker film matrix due to the weaker intermolecular interactions. The films formulated by adding 1.0 and 1.5% of glycerol were sticky and difficult to remove from the casting plate. Therefore, it is important to determine the most desirable plasticizer content for a packaging film in order to enhance its applicability as a packaging material.

The packaging color is important for consumer's acceptability as it can impact the appearance of the product. The determination of color parameters of formulated films was carried out by measuring the L* (lightness), a* (green–red) and b* (blue–yellow) values (Table 1). The lightness of the film (L*) increased with the increase in added glycerol concentration up to 1%. However, there is no significant difference between the films formulated with 1.0% and 1.5% glycerol levels ($p > 0.05$). Similarly, a* value of *Caesalpinia pulcherrima* seed gum films increased with the increase in glycerol concentration, however, no significant difference was observed between 1.0% and 1.5% ($p > 0.05$) films. The yellowness of the films (b*) were not affected by the added glycerol whereas a significant difference was observed only in the film with no plasticizer. The color values for *Caesalpinia pulcherrima* seed gum-based films obtained in this study are also compatible with the published research studies on the color parameters of different types of biopolymer-based films (Jouki et al. 2013a; Razavi et al. 2015; Nafchi et al. 2017).

Table 1 Effect of glycerol content on color parameters of films

Glycerol %	L*	a*	b*
0.0	37.6 ± 0.2^c	2.6 ± 0.1^c	4.3 ± 0.1^b
0.5	38.5 ± 0.1^b	2.8 ± 0.1^b	4.6 ± 0.1^a
1.0	39.5 ± 0.7^a	3.0 ± 0.1^a	4.5 ± 0.1^a
1.5	40.0 ± 0.2^a	3.2 ± 0.1^a	4.6 ± 0.1^a

Values represent the mean ± standard deviation of four replicates for all tests. Values in a column having different superscript letters are significantly different ($p < 0.05$)

The impact of glycerol concentration on the physical properties of the films is given in Table 2. The thickness of the films increased significantly with the increment of glycerol content. Accordingly, the films produced with high glycerol contents absorb more moisture that led to an increase in the thickness because of swelling (Dick et al. 2015). This phenomenon itself may cause limited applicability in food packaging. These findings are also consistent with other research studies that have been carried out for the film thickness and the plasticizer concentration (Jouki et al. 2013a; Khazaei et al. 2014; Razavi et al. 2015).

The hydrophilic nature of the *Caesalpinia pulcherrima* seed gum-based films also increased with the increment of glycerol content (Table 2) and similar findings have been reported by previous research studies focused on seed gum-based edible films (Jouki et al. 2013a; Zhang et al. 2016). Moisture content and the swelling index consistently increased with the thickness of the film as a result of increasing glycerol concentration in the film formulation. Due to the replacement of intermolecular interactions between the polysaccharide chains from the hydrogen bonds with the plasticizer, the increased plasticizer content adds more hydrophilicity to the film which increases the moisture absorbance ability (Dick et al. 2015). Further, it can minimize the applicability of film in food packaging as it can negatively affect the properties of the food that is going to be packaged and reduce the shelf life of both food products and packaging film. The study which was carried out by Jouki et al. (2013a) on the impact of glycerol content on the cress seed gum-based edible film also revealed that the moisture content of films was directly increased with the glycerol content. According to the results of the study, increasing glycerol content caused increased water solubility of films. The water solubility of *Caesalpinia pulcherrima* seed gum-based films may increase its packaging applications in several food products such as meat products, instant tea bags, flavoring sachets etc. Similar findings have been reported by Nafchi et al. (2017), on *Alyssum homolocarpum* seed gum-based films in which increasing the glycerol content in film formulations suggested increased water solubility owing to the increase of water attraction to the polysaccharide-based film network that creates a high mobility of polymeric chains with high interchain distances (Dick et al. 2015).

3.2 Mechanical properties of films

Mechanical properties of food packaging films are important because they determine the behavior of the films towards the force applied on their surface, which is an indication of the films ability to minimize losses in storage as well as in distribution. The formulated films are characterized by their mechanical properties such as strength, flexibility and stiffness in terms of the tensile strength (TS), elongation at break (EAB) and Young's modulus (YM). The TS of a film accounts for the maximum cross-sectional deformation to the breaking point. The EAB gives the percentage increase in the length of the film at breaking point when subjected to an opposing force. YM refers to the ratio of tensile strength and elongation at break (Martins et al. 2019). The impact of mechanical properties on the *Caesalpinia pulcherrima* seed gum-based films is illustrated in Fig. 4.

The TS and YM of *Caesalpinia pulcherrima* seed gum-based films significantly decreased ($p < 0.05$) with the incorporation of glycerol from 0.0% (10.90 ± 0.08 and 48.46 ± 0.24 MPa) to 1.5% (2.11 ± 0.05 and 3.47 ± 0.09 MPa). The EAB values significantly increased ($p < 0.05$) with the addition of glycerol from 0.0% (22.50 ± 0.05) to 1.5% (60.84 ± 0.04%). Thus, the results obtained for the mechanical properties of *Caesalpinia pulcherrima* seed gum-based films are compatible with the findings that have been extensively discussed in literature. The increase in the plasticizer content contributes to a decrease in the strength of the film matrix while significantly increasing their flexibility. Zhang et al. (2016) investigated the feasibility of gum ghatti for edible film making and reported similar behavior of mechanical properties with the increase of plasticizer content. The plasticizer content similarly affected the mechanical properties of tara gum-based edible film (Antoniou et al. 2014). The increase in plasticizer content once again contributed to a decrease in the strength of the film matrix while significantly increasing their flexibility. This means that the plasticizer concentration needs to be optimized to deliver the optimum strength and flexibility of these types of gum used in edible film making.

Table 2 Effect of glycerol content on the physical properties of films

Glycerol %	Thickness (mm)	Moisture content (%)	Swelling index	Water solubility (%)
0.0	0.039 ± 0.001^{d}	62.92 ± 0.51^{c}	5.39 ± 0.17^{d}	55.69 ± 0.51^{d}
0.5	0.050 ± 0.001^{c}	66.60 ± 0.75^{b}	6.76 ± 0.21^{c}	59.19 ± 0.33^{c}
1.0	0.065 ± 0.001^{b}	68.43 ± 0.15^{a}	7.55 ± 0.03^{b}	62.42 ± 0.08^{b}
1.5	0.076 ± 0.001^{a}	69.40 ± 0.15^{a}	8.45 ± 0.17^{a}	66.66 ± 0.45^{a}

Values represent the mean ± standard deviation of four replicates for all tests. Values in a column having different superscript letters are significantly different ($p < 0.05$)

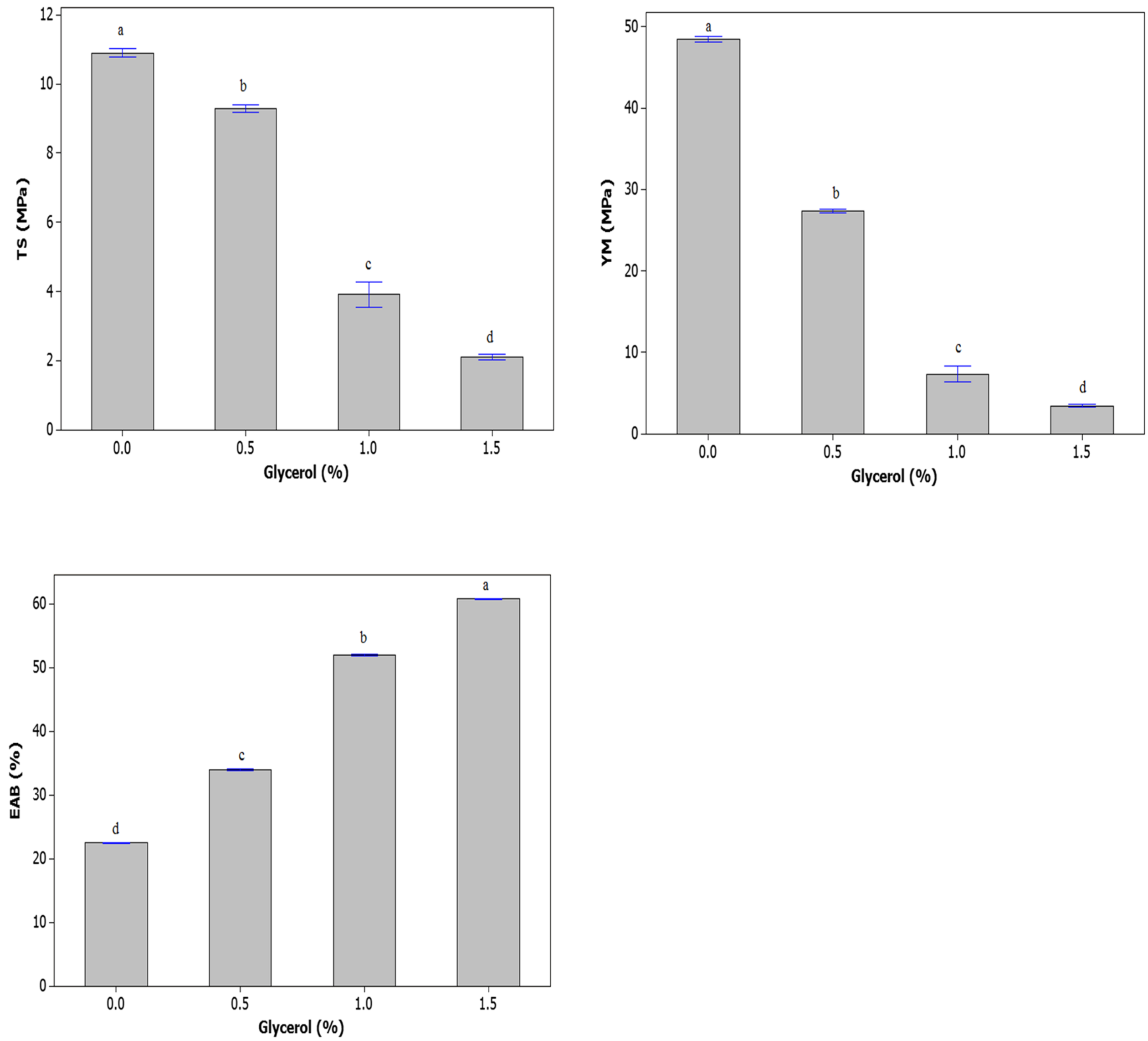

Fig. 4 Effect of glycerol content on the mechanical properties of films. The values indicated by bars represent the mean ± standard deviation of four replicates for all tests. Bars with different letters are significantly different ($p < 0.05$)

4 Conclusion

The findings of this study revealed the suitability of *Caesalpinia pulcherrima* seed gum as an edible film making material. The incorporation of glycerol as a plasticizer in making films from *Caesalpinia pulcherrima* seed gum leads to flexible but sticky films with higher hydrophilicity, elongation and lower TS and YM. *Caesalpinia pulcherrima* seed gum could be a potential source of edible films which can be used as a water-soluble packaging material. However, further scientific studies are needed to identify the barrier properties of *Caesalpinia pulcherrima* seed gum-based films and their impact on product characteristics to widen their applicability as a food packaging material.

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Authors' contributions All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Sandunika Senarathna. The first draft of the manuscript was written by Sandunika Senarathna and Seneviratne Navaratne, Indira Wickramasinghe, Ranil Coorey commented on previous versions of the manuscript. All authors read and approved the final manuscript.

Declarations

Conflict of interest The authors declare that they have no conflict of interest.

Availability of data and material The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Code availability Not applicable.

Ethics approval Not applicable.

Consent to participate Not applicable.

Consent for publication Not applicable.

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