



Alginate/aloë vera films reinforced with tragacanth gum

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ABSTRACT

The objective of present study was to investigate the effect of incorporation of varying concentrations (2% to 14%) of Tragacanth gum (TG) to alginate/aloë vera composite films to enhance their functional properties. The resulting films were investigated for their mechanical, barrier, optical properties and biodegradability. The WVP, swelling capacity and thickness of films increased significantly by the addition of TG while film solubility was dropped at higher concentration of TG. It was observed that TG acted as an efficient reinforcing agent for enhancing the strength and flexibility of the films. The tensile strength (TS) of films increased more than threefold as compared to control, reaching a maximum value 67.64 N/mm² at 12% concentration of TG. Colour properties were affected by the addition of TG as the higher the concentration, the darker the films.

1. Introduction

Recent researches have focussed on utilization of natural resources to develop biodegradable and edible food packaging films and boosting their properties to obtain the required characteristics of petroleum-based plastics like lightness, softness, water resistance and mechanical strength. Edible biopolymers which include proteins, polysaccharides and lipids can be a better replacement of synthetic packaging materials (Beikzadeh, Khezerlou, Jafari, Pilevar, & Mortazavian, 2020).

Sodium alginate (SA), a natural biopolymer extracted from brown algae is made up of alternative blocks of guluronic and mannuronic acid residues. The various patterns of arrangement for these two uronic acids are M–M, G–G and M–G. Sodium alginate forms irreversible gels by reacting with many divalent and polyvalent metal ions except divalent magnesium cations. Sodium alginate was found to be a promising material in many industrial applications in cosmetics, pharmaceutical, food and other chemical industries as a stabilizing, thickening and gelling agent owing to its distinguished properties like biocompatibility, biodegradability, non-toxic nature and low cost (Bergonzi et al., 2020). Alginates are also being used to develop films as they are biocompatible, non toxic and possess good film forming properties. However, it has some disadvantages like its great hydrophilic nature and low mechanical properties, due to which films based on sodium alginate alone lack the required strength. This drawback of alginate has restricted its use in the food packaging films. To overcome these shortcomings, films can be prepared by compounding other biopolymer and/or natural additives with sodium alginate and thus composite films of improved properties

could be obtained. Several researchers have tried to improve the quality of alginate films by mixing different types of gums and fibers (Cheng et al., 2020; Thomas et al., 2020). (Lu et al., 2021) claimed to improve the tensile strength of sodium alginate based films by the addition of oregano essential oil and mesoporous nano silica. Similarly, another study reported that the addition of gallnut extract enhanced the tensile strength and elongation at break of films based on sodium alginate (Aloui, Deshmukh, Khomlaem, & Kim, 2021). Incorporation of thymol into sodium alginate films, reported by Chen et al. (Chen et al., 2021), was also found to improve the mechanical, barrier and optical properties of the resulting composite films compared with standard sodium alginate film without thymol.

Aloe vera (AV) is a xerophyte succulent plant which grows in tropical areas of the world. It is a natural product of interest which has been utilized in edible films for enhancing the storage life and maintaining the nature of different fruits and vegetables. Aloe vera gel was added not only as a basic component of the composite films to improve the physicochemical and mechanical properties but also to make it active against oxidation and bacterial proliferation in order to enhance its role for food preservation. Furthermore, AV gel also behaves as a plasticizer to improve the flexibility of the films along with typical plasticizer (Bajer, Janczak, & Bajer, 2020). For further enhancement of physical and chemical properties of biopolymer films, many other types of polysaccharides such as natural fibers have been studied as film forming materials for food packaging. Tragacanth gum (TG) is an example of natural fibers and is extensively used as dietary fiber, in packaging films, thickening agents, stabilizers, emulsifiers, and coating agent etc.

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Tragacanth gum is a natural gum obtained from Middle Eastern legumes of the genus *Astragalus*. It is highly branched anionic polysaccharide. Its major components are tragacanthin and bassorin. The highly soluble tragacanthin forms a mucilaginous colloid. The major component bassorin is 60 to 70 percent of the total gum and is much less soluble but has a high swelling capacity in water to form a gel (Gorji, Gorji, Mohamadifar, & Zargaraan, 2014). Since TG has both hydroxyl and carboxyl groups, it is expected to form linkages with various functional groups like amino, hydroxyl, carboxyl and sulfonic acids (Sharma et al., 2021). Due to this reason ionic cross linking agents and organic monomers have been utilized to form ionic linkage with carboxyl groups present in TG (Qasemi & Ghaemy, 2020; Sahraei & Ghaemy, 2017). Usually a single biopolymer is not capable enough to form a good film by itself alone. For this reason various composite films based on ternary systems like whey protein isolate, gelatine and sodium alginate (Wang, Auty, & Kerry, 2010), k-carrageenan, i-carrageenan, and alginate (Paula et al., 2015) and cassava starch, carnauba wax and cashew tree gum (Rodrigues et al., 2014) have been developed and studied. The literature survey showed that TG, when used alone exhibited poor film properties, therefore mostly it is used with other hydrocolloids like whey protein isolate (WPI), where it improved the flexibility and WVP of films as reported earlier (Tonyali, Cikrikci, & Oztop, 2018). Therefore the present study also aimed to investigate and characterize the ternary biopolymer system based on alginate, aloe vera and tragacanth gum to develop composite films.

2. Materials and methods

2.1. Materials

The algal material was collected from Karachi seashore. The brown seaweeds (*Sargassum* spp.) were then washed, dried and crushed into fragments, and were kept in fridge before use. Aloe vera (*Aloe barbadensis* Miller) growing in the premises of the University of Karachi, was utilized in this study. Tragacanth gum (TG) was purchased from local market. Analytical grade laboratory chemicals and sorbitol was procured from Sigma-Aldrich Chemical Ltd.

2.2. Extraction of sodium alginate

The method of Gholamipoor, Ghanavati, Oromiehie, and Mohammadi (2013) was followed for extraction of sodium alginate from brown seaweed. 2% formaldehyde solution was used as extractant in which 25 g dry algal material was placed for 24 h at room temperature. The material was then washed with distilled water and immersed in 800 ml HCl 0.2 M solution for 24 h. After washing again it was stirred with 2% sodium carbonate for 3 h at 50 °C. The resulting slurry was centrifuged at 3500 rpm for 20 min. 95% ethanol was added to the supernatant liquid and sodium alginate was obtained as precipitate, which was washed with acetone, dried and again dissolved in 100 ml of distilled water. The alginate was re-precipitated from this aqueous solution by adding 95% ethanol and dried at 45 °C in an oven (Gholamipoor, Ghanavati, Oromiehie, & Mohammadi, 2013).

2.3. Extraction of aloe vera gel

Aloe vera gel was extracted from fresh plant leaves after removing the outer skin. The gel was mixed at slow speed in a blender and filtered through a 200 mesh sieve. Foam and callus was removed from the filtrate by centrifuging at 4800 rpm for 10 min. The supernatant so obtained was utilized in formation of composite films.

2.4. Alginate/aloe vera/tragacanth composite films preparation

Casting technique was used in films preparation. Alginate/Aloe Vera (AAV) composite films were prepared by first dissolving sodium alginate

(3% w/w) in distilled water at 60–70 °C on a hot plate for 30 min under stirring. Then 10% aloe vera extract (w/w of sodium alginate db) was added to the alginate solution, and homogenized again using magnetic stirrer. 50% sorbitol (w/w of sodium alginate db) was used as plasticizer and it was added after complete dissolution (Hadi, Nawab, Alam, & Zehra, 2021). Film based on sodium alginate alone was taken as control.

For AAV-TG films different concentrations of TG (2%, 4%, 6%, 8%, 10%, 12% and 14%) was selected. The selection of these tragacanth concentrations was based on preliminary trials wherein the concentration was gradually increased from a low level of 2% to a maximum of 14%. Increasing the concentration further was detrimental as the films started getting brittle. This range of tragacanth concentration (2% to 14%) was found to be optimal as it produced non sticky, smooth and flexible films. Different concentrations of TG was added to the previously prepared AAV solution. The solution was heated again on a hot plate under stirring for additional 30 min. After complete homogenization the film forming solution was then casted onto petri dishes and dried in an oven (Binder GmbH, Tuttlingen, Germany) at 40 °C for 24 h. The blend films were then peeled off from the petri dishes and stored before characterization.

2.5. Film thickness

The thickness of the film was determined by taking readings at five different points in the film. Average film thickness was calculated from these five values of thickness, measured with a digital micrometer (Mitutoyo, Tokyo, Japan) with a precision of 0.001 mm. Film's mechanical and barrier properties were investigated using this mean thickness value.

2.6. Moisture content

Moisture content of the films was determined with the method of Norajit, Kim, and Ryu (2010) (Norajit et al., 2010). Film samples were weighed and then dried at 110 °C for 24 h. After keeping in a desiccator for 30 min, the samples were weighed again to calculate the moisture content according to equation..

$$MC = \frac{W_i - W_d}{W_i} \times 100 \quad (1)$$

2.7. Swelling power

The method of Sarheed et al. was used to determine swelling power of the films (Sarheed, Rasool, Abu-Gharbieh, & Aziz, 2015) with some modifications. Square pieces of the film were weighed and immersed in distilled water at room temperature for 30 min. The extra water was removed by absorption with filter paper and the film pieces were weighed again. The swelling capacity was calculated by the equation..

$$S = \left[\frac{W_s - W_d}{W_d} \right] \times 100 \quad (2)$$

2.8. Solubility

The method of Jangchud & Chinnan (Jangchud & Chinnan, 1999), after some modifications, was followed for film's solubility determination. Films were cut into 20x20 mm sample pieces. After drying these pieces were weighed and then immersed in 20 ml distilled water in 50 ml Erlenmeyer flasks, which were shaken in a water bath at 25°C for 24 h. The undissolved film residues were filtered and dried at 80°C in oven. The film solubility was calculated as per the equation..

$$\text{Solubility (\%)} = \frac{\text{Mass of film} - \text{Mass of residue}}{\text{Mass of film sample}} \times 100 \quad (3)$$

2.9. Scanning electron microscopy (SEM)

The scanning electron microscope (JSM, 6380A, Jeol, Japan) was used to study the microstructure of AAV-TG composite films at an accelerated voltage of 10 kV.

2.10. Mechanical properties

The mechanical tests were performed according to the method described by Hadi et al. (Hadi et al., 2021).

2.11. Water vapor permeability

The films Water vapour permeability (WVP) was conducted using the method described by Nawab et al. (Nawab, Alam, Haq, Lutfi, & Hasnain, 2017).

2.12. Optical properties

The colour analysis of the films was performed as reported by (Yang et al., 2015). The a^* , b^* and L^* values were measured in triplicate. Opacity of the films was analysed by spectrophotometer (JASCO Corporation, Tokyo, Japan) by following the method described previously (Ebrahimi, Koocheki, Milani, & Mohebbi, 2016). Film opacity was estimated by using the equation:

$$\text{Opacity} = \frac{A_{600}}{X} \quad (4)$$

2.13. Biodegradation

Films biodegradation was carried out by the soil burial test as described by Martucci (Martucci & Ruseckaite, 2009). Films were cut into rectangular pieces, oven dried at 105 °C for 24 h and weighed (W_i). The pieces of films were buried under compost to undergo decomposition. The water was added periodically to maintain a 35% relative humidity (RH) at a temperature of 32°C. At different time intervals the film samples were taken out, oven dried at 105°C for 12 h and were weighed (W_t) to measure the percent weight loss (WL) as follows:

$$\text{WL}\% = \frac{W_i - W_t}{W_i} \times 100 \quad (5)$$

2.14. Statistical analysis

The data from triplicate analysis of samples was subjected to one way analysis of variance (ANOVA) and Duncan's multiple range tests to differentiate the treatments at $p < 0.05$. SPSS version 17.01 for windows program (SPSS Inc., Chicago, IL, USA) was utilized for statistical analyses.

3. Results and discussion

3.1. Thickness and moisture content of films

The thickness of the film is an important criterion as the physico-chemical and mechanical properties of the film largely depend on the film thickness. The results of the film thickness are presented in Table 1. The thickness varied from 0.069 to 0.085 mm. Due to the fact that tragacanth gum has gel like property, thus it absorbs moisture and swells that eventually resulted in increased film thickness. These results are in conformity with recent study conducted by Pirsra et al. (Pirsra, Mohtar-ami, & Kalantari, 2020).

The results showed that the total moisture content of the film increased from 3% in (control) to 23% at highest concentration of TG (TG14) as shown in Fig. 1. The reason for this increase could be the higher affinity of TG towards water as mentioned earlier and also its

Table 1

Effect of tragacanth gum (TG) on the mechanical and water vapour permeability of alginate aloe vera films.

Film Concentration (%)	Thickness (mm)	WVP × 10 ⁻¹⁰ (g/Pa.m.s)	Tensile strength N/mm ²	Elongation at break (%)
A	0.069 ± 0.25 ^a	1.25 ± 0.03 ^a	20.92 ± 0.54 ^e	0.7 ± 0.02 ^a
AAV	0.071 ± 0.38 ^c	1.27 ± 0.05 ^b	21.56 ± 0.23 ^a	0.9 ± 0.04 ^a
TG2	0.074 ± 0.54 ^e	2.37 ± 0.07 ^b	25.58 ± 0.48 ^d	2.41 ± 0.08 ^b
TG4	0.076 ± 0.61 ^f	2.84 ± 0.12 ^c	36.45 ± 0.31 ^c	2.45 ± 0.10 ^b
TG6	0.078 ± 0.29 ^b	3.54 ± 0.14 ^c	38.28 ± 0.46 ^d	3.15 ± 0.12 ^b
TG8	0.079 ± 0.49 ^e	3.68 ± 0.16 ^d	40.18 ± 0.73 ^g	3.48 ± 0.16 ^c
TG10	0.081 ± 0.43 ^d	4.99 ± 0.24 ^e	43.75 ± 0.75 ^g	5.36 ± 0.24 ^d
TG12	0.083 ± 0.58 ^f	7.25 ± 0.36 ^f	67.49 ± 0.66 ^f	5.50 ± 0.26 ^e
TG14	0.085 ± 0.42 ^d	8.31 ± 0.41 ^g	65.34 ± 0.29 ^b	5.42 ± 0.21 ^d

Values are means ± SD of triplicates.

Values in the same column with different superscript are significantly different ($P < 0.05$).

Abbreviations: A (alginate) , AV (aloe vera) , TG (tragacanth gum).

high ability to retain greater amount of water. Pirsra and co-workers (Pirsra et al., 2020) also claimed that a composite film of starch/tragacanth gum/nanoclay showed increased moisture content on increasing the concentration of tragacanth gum. Similar results were also reported in other study conducted by Saberi et al. (Saberi et al., 2016) using pea starch and guar gum.

Moisture content of the films was determined with the previously reported method of Norajit et al. (2010) (Norajit et al., 2010) that did not report any thermal degradation or other remarkable change during drying of the film samples. For the accurate determination of the moisture content of the films, the present method required for placing the films at 110 °C for 24 h, to achieve complete removal of moisture from the films. We also observed the same as the previous study that the temperature and length of time did not cause any thermal degradation of the film samples as indicated by the absence of any change in the color and flexibility. Moreover, sorbitol also enhanced the thermal stability of these composite films by providing additional plasticizing effect along-with aloe vera gel. The previous study conducted by (Sánchez et al., 2020) also stressed that the thermal stability of fish gelatin films enhanced by the addition of aloe vera gel (Sánchez et al., 2020).

3.2. Swelling capacity

The swelling behaviour is an important parameter which affects the mechanical and barrier properties of the films. The stability and quality changes in the packaged food during storage will depend on the swelling characteristics of the film. The results showed that swelling capacity of composite films increases with increasing concentration of TG. The swelling capacity of control was found to be 103.8 % which increased to 451.11% when highest concentration of gum was added i.e. 14% (TG14) as shown in Fig. 1. The reason for increase in swelling capacity of the films with increasing concentration of TG is due to highly branched polysaccharide structure of TG which contributes to its high swelling and gelling properties (Mostafavi, Kadkhodae, Emadzadeh, & Koocheki, 2016). Moreover, gum tragacanth has a very high capacity to absorb water which enables it to absorb water several times its weight and form a gel. Some recent studies have also reported the increase in swelling capacity of polysaccharide based composite films by the addition of gums (Cheng et al., 2020; Khodaei, Oltrogge, & Hamidi-Esfahani,

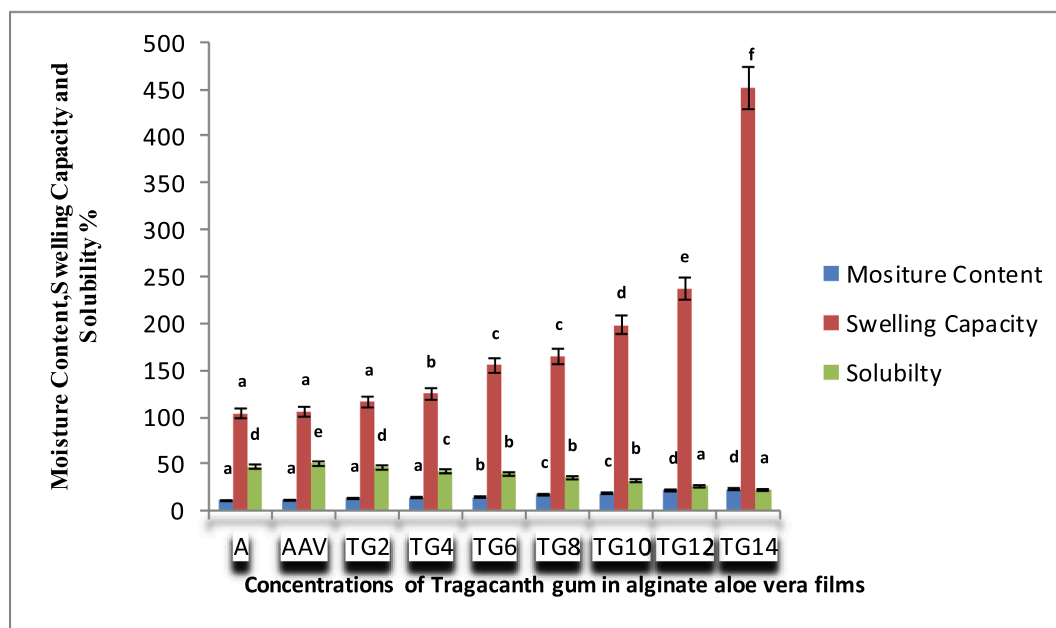


Fig. 1. Effect of TG on moisture content, swelling capacity and solubility of composite films.

2020).

3.3. Solubility and water vapor permeability (WVP)

Solubility and WVP, of the biodegradable food packaging films are two important factors which determine the ability of the film to protect the packaged food from decay. Addition of TG to AAV composite films reduced the film solubility from 47% (control) to 22% (TG14) as shown in Fig. 1. Similar studies were conducted by Pirsra and co-workers (Pirsra et al., 2020) where the authors claimed that the reduction in solubility of starch films by the addition of TG was due to the fact that tragacanth gum contains a substance called bassorin, which is insoluble in water and lowers the solubility of the film in water. Another study conducted on effect of TG on whey protein isolate (WPI) by Tonyali et al. (Pirsra et al., 2020; Tonyali et al., 2018) also reported the improved water resistance of WPI films with the addition of TG. The authors claimed that the heating step during film formation could have initiated the Maillard reaction which caused TG to crosslink with WPI, thus reducing the film solubility. For solubility determination the undissolved residual film samples were dried in an oven at 80C. There was no indication of a maillard reaction at this temperature, because as discussed earlier, even at 110C no change in colour of the films was observed. Browning is a typical outcome of maillard reaction, as is observed in the browning of bread and cookies during cooking (Karangwa et al., 2016). Maillard reactions begin to occur above 140C, and are also defined as non-enzymatic browning reactions (Jiang et al., 2018). TG is remarkably stable in a wide range of temperature and pH, which makes it suitable for use as an emulsifier in food and drug industry (Mohamadnia, Zohuriaan-Mehr, Kabiri, & Razavi-Nouri, 2008; Zohuriaan & Shokrolahi, 2004). The heat stability of TG also rules out the possibility of maillard reaction.

The results presented in Table 1 showed that the addition of TG to AAV composite films increased the WVP from $1.25 \times 10^{-10} \text{ g m}^{-1} \text{ s}^{-1} \text{ Pa}^{-1}$ (control) to $8.31 \times 10^{-10} \text{ g m}^{-1} \text{ s}^{-1} \text{ Pa}^{-1}$ in (TG14). The high swelling capacity of TG and its ability to form a gel, is the reason for this increase as already mentioned earlier. Addition of TG to the film makes it swell creating a gap between polymer chains of AAV films that resulted in increased WVP. This trend is also in accordance with a recent study carried out on starch/TG/nanoclay films (Pirsra et al., 2020).

3.4. Scanning electron microscopy (SEM)

The microstructure of AAV composite films reinforced with TG was studied by scanning electron microscopy (SEM). The morphological microstructure of the composite films as well as their physical, optical, mechanical and barrier properties largely depend on the interaction, miscibility and compatibility between the composite film forming system. In the present study the micrographs of the surface of AAV and TG reinforced composite films are uniform and smooth showing a homogenous film network. The addition of TG to AAV matrix does not cause any irregularity, cracks, breaks or openings in the film structure. This indicates the compatibility of the reinforcing agent, that is gum tragacanth with the film forming matrix of alginate/aloe vera. The varying concentrations of TG were uniformly dispersed in the film matrix without deforming the surface microstructure of the films. The uniformity and homogenous structure of the films is an indication of the cross linking between various functional groups of TG and alginate/aloe vera components. The cross-section micrographs of films also do not show any cracks or breaks in the film microstructure, again indicating the compatibility of the reinforcing TG with AAV composite films. The findings of this study are consistent with a previously reported study (Shahvalizadeh et al., 2021) (Fig. 3)

3.5. Mechanical properties

Edible and biodegradable films based on biopolymers have been developed for application in food packaging. Being environment friendly they are capable of solving the pollution problem. But the main drawback of these films was their inferior mechanical properties as compared to synthetic plastic films. Their low TS and the ability to bear stress restrained them from efficiently replacing the synthetic plastic films in food packaging. The present study aims to enhance the mechanical strength of these biodegradable films based on biopolymers which are inherently low in tensile strength.

Significant improvement in TS of the AAV composite films was achieved by the addition of TG to film forming solution. Table 1 shows that TG has a drastic effect on the TS of these composite films as the value increased from 20.92 MPa (Control) to 67.49 MPa (TG12). There was more than threefold increase in the TS at near maximum concentration of TG i.e.12%. This shows that 12% concentration of TG was its

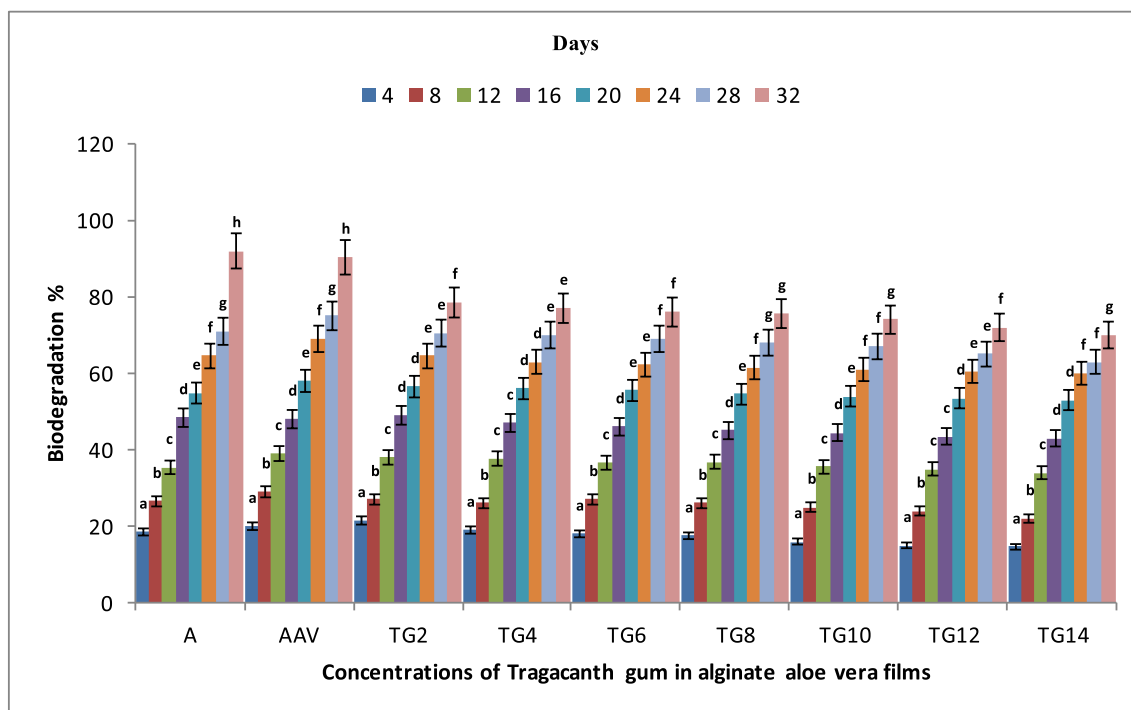


Fig. 2. Effect of TG on biodegradation of alginate-aloe vera composite films.

threshold value as further increase in concentration caused a slight decrease in TS. The reason for this increase is a synergistic interaction between polymer chains of TG and alginate which contributes to the cohesive forces working inside the film matrix. The similar effect of adding TG to the starch films was reported recently (Pirsa et al., 2020) where the authors suggested that the increase in TS of starch film by adding TG was due to the fact that TG inserted between the starch polymer chains that strengthen the van der Waal forces between polymer chains and eventually resulted in higher TS.

Incorporation of TG into AAV composite films also contributed positively to the flexibility of the films. Elasticity of the films improved with the increasing concentration of TG (Table 1). The values increased from 0.7% (control) to 5.50% (TG12). The ability of TG to form gel by absorbing water contributes to its plasticizing effect on the films. The increasing trend of EB values with increasing gum concentration in composite films has been reported previously (Pirsa et al., 2020; Tonyali et al., 2018).

3.6. Optical properties

Visual appearance and attraction of the packaging film is important for the marketability of the packaged food product. Natural polymer films with high transparency and colourlessness having a resemblance to synthetic plastic film like polyethylene have a better chance of acceptance and the potential to replace the synthetic materials. The colour properties of the packaging films are a function of their composition and the process used in their preparation. Biodegradable films developed from natural polymers may be clear, slightly hazy or have little coloured appearance. Indicator a^* measures the redness or greenness of the films, with positive numbers to show the redness and negative numbers represents the greenness. Whereas the b^* is a measure of yellow or blue colour of the film, with negative b showing blue background and a positive b pointing to the yellowness of the films. L shows the lightness or the intensity of the colour.

In the present study it was observed that the composite AAV films were transparent, clear and colourless. However the addition of TG increased the opacity of films significantly which increased further with

the increasing concentration of TG. This was due to the fact that TG being opaque in nature having a milky or greyish colour. Silva et al. (Silva, Mauro, Gonçalves, & Rocha, 2016) and Sui et al. (Sui, Zhang, Ye, Liu, & Yu, 2016) also reported the increased opacity of whey protein and soy protein isolate based films with the addition of locust bean gum and guar gum respectively. Similarly Ebrahimi et al. (Ebrahimi et al., 2016) found increased opacity of films prepared by higher gum ratio in pea protein based films which proved that all these gums being coloured in nature were responsible for the increment in film opacity.

The L^* values also showed slight decrease in colour of film with the addition of TG. No significant trend was observed in a^* and b^* values (Table 2). Similar effect of decreasing lightness value by the addition of guar and xanthan gum to mango kernel starch films were reported by Nawab and co-workers (Nawab et al., 2017). Arismendi et al. (Arismendi et al., 2013) reported that the L values were higher when the concentration of hydrocolloids was lower in the composite xanthan-tapioca starch films. Another study conducted by Ebrahimi et al. (Ebrahimi et al., 2016) reported the decreasing lightness values with increasing protein proportions which was also in accordance with the results of our study.

3.7. Biodegradation

Biodegradability is the most important requirement for the food packaging films of the future so that they can replace the synthetic plastic packaging materials which are non biodegradable and are a source of environmental pollution. Biodegradation of the control as well as composite films (AAV-TG) having various concentrations of TG was studied and the results are presented in Fig. 2. The in vitro degradation of the films was performed in order to explore the influence of addition of TG at various concentration to the AAV composites, on the degradation profile over a period of 32 days. The biodegradability of the films was determined in terms of percent weight loss of the films during this period. It was observed that during early stages of degradation i.e. on 4th day, there was no significant difference in percent weight loss between AAV films and TG6 films. At the highest concentration of TG (TG14) weight loss was 14.6%. At the end of degradation period the

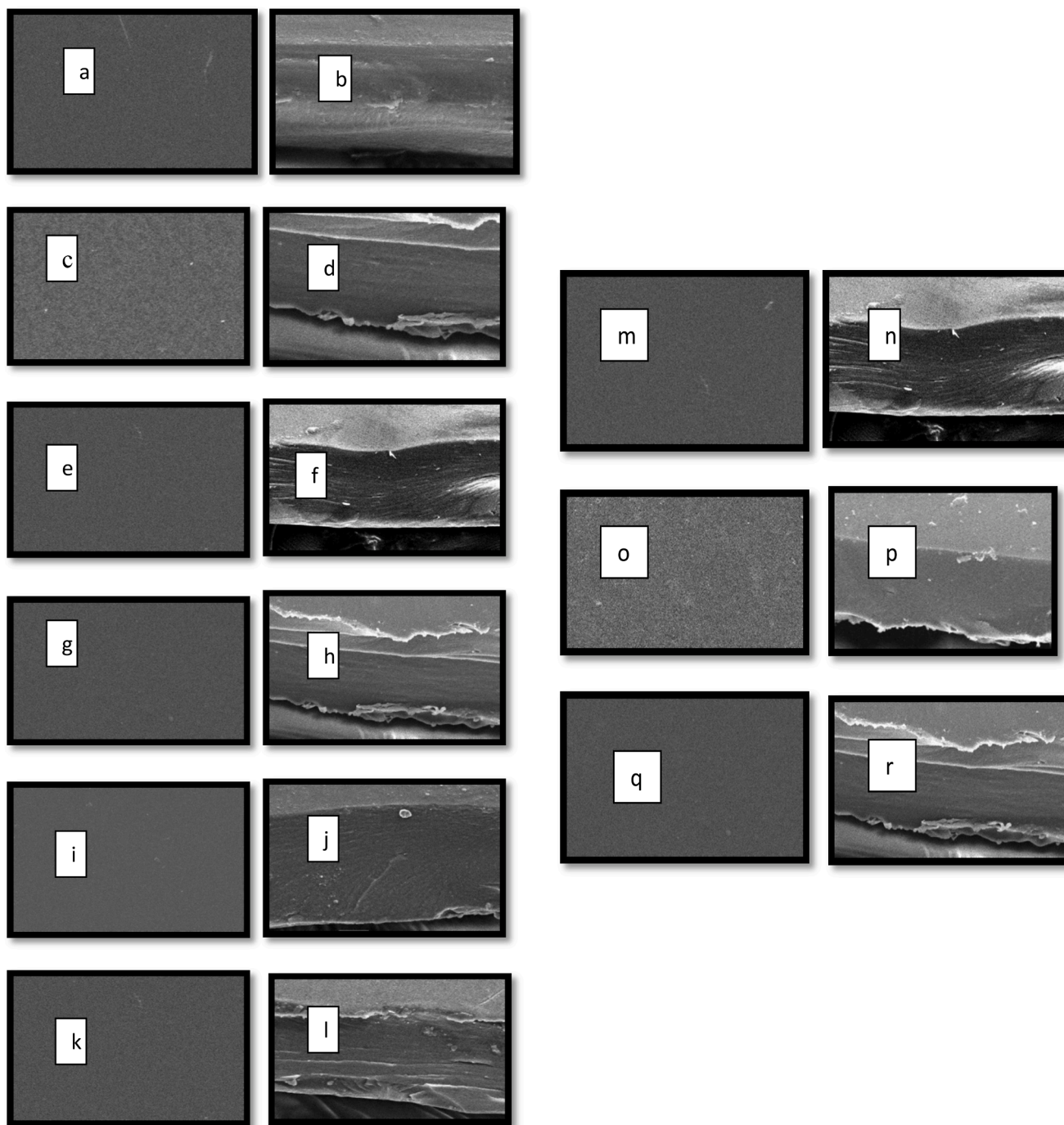


Fig. 3. Scanning electron micrograph (SEM) of the surface (left) and cross section (right) of alginate aloe vera composite films at magnification 1000x (a) and (b) A, (c) and (d) AAV, (e) and (f) TG2, (g) and (h) TG4, (i) and (j) TG6, (k) and (l) TG8, (m) and (n) TG10, (o) and (p) TG12, (q) and (r) TG14.

control film had almost completely degraded with 92% weight loss, whereas the TG14 films showed 70% weight loss. Throughout the degradation period i.e. upto 32 days, the control film appeared to degrade at a faster rate as compared with the films containing TG. As the concentration of TG increases from 2% to 14% the percent weight loss decreases on each time interval when the reading was taken. These results may not necessarily indicate a negative effect of TG on degradation of the film, the reduction in percent weight loss may be due to the retention of water by TG containing films as TG has a high water affinity and swelling capacity by absorbing water. The prior step of drying the films at 105C for 24 h did not affect the structure of the films as there was no colour change or any change in flexibility and physical

appearance of the films. (Even at a slightly higher temperature of 110C for 24 h, no change was observed while determining moisture content). This indicates the absence of temperature induced cross linkage or any other structural changes in the film which could enable it to resist the microbial action responsible for biodegradation.

4. Conclusion

The improved strength of the films by addition of an organic fibre like gum tragacanth, makes them a viable and biodegradable alternative for synthetic plastic films for the packaging of food materials. From the results of this study it can be concluded that composite films based on

Table 2

Effect of tragacanth gum (TG) on colour parameters and opacity (A600/mm) of alginate aloe vera films.

Samples	L*	a*	b*	Opacity
A	99.2 ± 1.28 ^b	0.3 ± 0.003 ^a	0.9 ± 0.043 ^f	0.14 ± 0.008 ^a
AAV	99.7 ± 2.45 ^d	0.2 ± 0.006 ^b	0.7 ± 0.013 ^d	0.29 ± 0.013 ^b
TG2	98.4 ± 1.92 ^c	0.9 ± 0.042 ^g	0.1 ± 0.004 ^b	0.87 ± 0.042 ^d
TG4	98.6 ± 1.18 ^a	0.7 ± 0.029 ^e	0.1 ± 0.002 ^a	1.32 ± 0.051 ^e
TG6	97.2 ± 3.28 ^e	0.6 ± 0.005 ^b	1.9 ± 0.028 ^e	1.96 ± 0.044 ^d
TG8	97.4 ± 4.24 ^f	0.4 ± 0.007 ^c	0.3 ± 0.014 ^d	2.52 ± 0.038 ^e
TG10	97.8 ± 2.29 ^c	0.5 ± 0.022 ^d	0.6 ± 0.001 ^a	2.89 ± 0.142 ^f
TG12	96.3 ± 3.25 ^e	0.7 ± 0.033 ^f	0.1 ± 0.005 ^b	3.34 ± 0.146 ^g
TG14	96.6 ± 2.58 ^d	0.1 ± 0.004 ^a	0.2 ± 0.009 ^c	3.96 ± 0.149 ^g

Values are means ± SD of triplicates.

Values in the same column with different superscript are significantly different (P < 0.05).

alginate aloe vera/tragacanth make them a strong candidate to effectively replace the conventional plastic films being presently utilized in food packaging.

Practical Application

Alginate and aloe vera, the two natural resources were used for the development of biodegradable and edible food packaging films. The addition of an organic fiber, gum tragacanth, resulted in a threefold increase in the mechanical strength of the films and also improved their flexibility. Gum tragacanth acts as an excellent reinforcing agent and places these films in a better position to compete with synthetic plastics which are adding to environmental pollution.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Alina Hadi reports writing assistance was provided by University of Karachi. Alina Hadi reports a relationship with University of Karachi that includes: non-financial support.

References

Aloui, H., Deshmukh, A. R., Khomlaem, C., & Kim, B. S. (2021). Novel composite films based on sodium alginate and gellan extract with enhanced antioxidant, antimicrobial, barrier and mechanical properties. *Food Hydrocolloids*, 113, Article 106508.

Arisemendi, C., Chillo, S., Conte, A., Del Nobile, M. A., Flores, S., & Gerschenson, L. N. (2013). Optimization of physical properties of xanthan gum/tapioca starch edible matrices containing potassium sorbate and evaluation of its antimicrobial effectiveness. *LWT-Food Science and Technology*, 53(1), 290–296.

Bajer, D., Janczak, K., & Bajer, K. (2020). Novel starch/chitosan/alginate composites as promising biopackaging materials. *Journal of Polymers and the Environment*, 28(3), 1021–1039.

Beikzadeh, S., Khezerlou, A., Jafari, S. M., Pilevar, Z., & Mortazavian, A. M. (2020). Seed mucilages as the functional ingredients for biodegradable films and edible coatings in the food industry. *Advances in Colloid and Interface Science*, 280, Article 102164.

Bergonzi, C., d'Ayala, G. G., Elviri, L., Laurienzo, P., Bandiera, A., & Catanzano, O. (2020). Alginate/human elastin-like polypeptide composite films with antioxidant properties for potential wound healing application. *International journal of biological macromolecules*, 164, 586–596.

Chen, J., Wu, A., Yang, M., Ge, Y., Pristijono, P., Li, J., ... Mi, H. (2021). Characterization of sodium alginate-based films incorporated with thymol for fresh-cut apple packaging. *Food Control*, 126, Article 108063.

Cheng, T., Xu, J., Li, Y., Zhao, Y., Bai, Y., Fu, X., ... Mao, X. (2020). Effect of gum ghatti on physicochemical and microstructural properties of biodegradable sodium alginate edible films. *Journal of Food Measurement and Characterization*, 1–12.

Ebrahimi, S., Koocheki, A., Milani, E., & Mohebbi, M. (2016). Interactions between *Lepidium perfoliatum* seed gum–Grass pea (*Lathyrus sativus*) protein isolate in composite biodegradable film. *Food Hydrocolloids*, 54, 302–314.

Gholamipoor, S., Ghanavati, Y., Oromiehie, A., & Mohammadi, M. (2013). Extraction and Characterization of Alginat from *Sargassum angustifolium* collected from northern coasts of Persian Gulf, Bushehr. *International Symposium on Advances in Science and Technology*, Iran.

Gorji, S. G., Gorji, E. G., Mohammadifar, M. A., & Zargaraan, A. (2014). Complexation of sodium caseinate with gum tragacanth: Effect of various species and rheology of coacervates. *International journal of biological macromolecules*, 67, 503–511.

Hadi, A., Nawab, A., Alam, F., & Zehra, K. (2021). Physical, mechanical, optical, barrier, and antioxidant properties of sodium alginate–aloe vera biocomposite film. *Journal of Food Processing and Preservation*, e15444.

Jangchud, A., & Chinnan, M. (1999). Peanut protein film as affected by drying temperature and pH of film forming solution. *Journal of Food Science*, 64(1), 153–157.

Jiang, W., Chen, Y., He, X., Hu, S., Li, S., & Liu, Y. (2018). A study of the tyramine/glucose Maillard reaction: Variables, characterization, cytotoxicity and preliminary application. *Food chemistry*, 239, 377–384.

Karangwa, E., Murekatete, N., de Dieu Habimana, J., Masamba, K., Duhoranimana, E., Muhoza, B., & Zhang, X. (2016). Contribution of crosslinking products in the flavour enhancer processing: The new concept of Maillard peptide in sensory characteristics of Maillard reaction systems. *Journal of food science and technology*, 53(6), 2863–2875.

Khodaei, D., Oltrogge, K., & Hamidi-Esfahani, Z. (2020). Preparation and characterization of blended edible films manufactured using gelatin, tragacanth gum and Persian gum. *LWT*, 117, Article 108617.

Lu, W., Chen, M., Cheng, M., Yan, X., Zhang, R., Kong, R., ... Wang, X. (2021). Development of antioxidant and antimicrobial bioactive films based on Oregon essential oil/mesoporous nano-silica/sodium alginate. *Food packaging and shelf life*, 29, Article 100691.

Martucci, J. F., & Ruseckaite, R. A. (2009). Biodegradation of three-layer laminate films based on gelatin under indoor soil conditions. *Polymer Degradation and Stability*, 94(8), 1307–1313.

Mohamadnia, Z., Zohuriaan-Mehr, M. J., Kabiri, K., & Razavi-Nouri, M. (2008). Tragacanth gum-graft-polyacrylonitrile: Synthesis, characterization and hydrolysis. *Journal of Polymer Research*, 15(3), 173–180.

Mostafavi, F. S., Kadkhodaei, R., Emadzadeh, B., & Koocheki, A. (2016). Preparation and characterization of tragacanth–locust bean gum edible blend films. *Carbohydrate polymers*, 139, 20–27.

Nawab, A., Alam, F., Haq, M. A., Lutfi, Z., & Hasnain, A. (2017). Mango kernel starch-gum composite films: Physical, mechanical and barrier properties. *International Journal of Biological Macromolecules*, 98, 869–876.

Norajit, K., Kim, K. M., & Ryu, G. H. (2010). Comparative studies on the characterization and antioxidant properties of biodegradable alginate films containing ginseng extract. *Journal of Food Engineering*, 98(3), 377–384.

Paula, G. A., Benevides, N. M., Cunha, A. P., de Oliveira, A. V., Pinto, A. M., Morais, J. P. S., & Azeredo, H. M. (2015). Development and characterization of edible films from mixtures of κ-carrageenan, ι-carrageenan, and alginate. *Food Hydrocolloids*, 47, 140–145.

Pirsa, S., Mohtarami, F., & Kalantari, S. (2020). Preparation of biodegradable composite starch/tragacanth gum/nanoclay film and study of its physicochemical and mechanical properties. *Chemical Review and Letters*, 3(3), 98–103.

Qasemi, S., & Ghaemy, M. (2020). Novel superabsorbent biosensor nanohydrogel based on gum tragacanth polysaccharide for optical detection of glucose. *International journal of biological macromolecules*, 151, 901–908.

Rodrigues, D. C., Caceres, C. A., Ribeiro, H. L., de Abreu, R. F., Cunha, A. P., & Azeredo, H. M. (2014). Influence of cassava starch and carnauba wax on physical properties of cashew tree gum-based films. *Food Hydrocolloids*, 38, 147–151.

Saberi, B., Thakur, R., Vuong, Q. V., Chockchaisawasdee, S., Golding, J. B., Scarlett, C. J., & Stathopoulos, C. E. (2016). Optimization of physical and optical properties of biodegradable edible films based on pea starch and guar gum. *Industrial Crops and Products*, 86, 342–352.

Sahraei, R., & Ghaemy, M. (2017). Synthesis of modified gum tragacanth/graphene oxide composite hydrogel for heavy metal ions removal and preparation of silver nanocomposite for antibacterial activity. *Carbohydrate polymers*, 157, 823–833.

Sánchez, J. T., García, A. V., Martínez-Abad, A., Vilaplana, F., Jiménez, A., & Garrigós, M. C. (2020). Physicochemical and functional properties of active fish gelatin-based edible films added with aloe vera gel. *Foods*, 9(9), 1248.

Sarheed, O., Rasool, B. K. A., Abu-Gharbieh, E., & Aziz, U. S. (2015). An investigation and characterization on alginate hydrogel dressing loaded with metronidazole prepared by combined isotropic gelation and freeze-thawing cycles for controlled release. *AAPS PharmSciTech*, 16(3), 601–609.

Shahvalizadeh, R., Ahmadi, R., Davandeh, I., Pezeshki, A., Moslemi, S. A. S., Karimi, S., ... Mohammadi, M. (2021). Antimicrobial bio-nanocomposite films based on gelatin, tragacanth, and zinc oxide nanoparticles–Microstructural, mechanical, thermo-physical, and barrier properties. *Food chemistry*, 354, Article 129492.

Sharma, B., Thakur, S., Mamba, G., Gupta, R. K., Gupta, V. K., & Thakur, V. K. (2021). Titania modified gum tragacanth based hydrogel nanocomposite for water remediation. *Journal of Environmental Chemical Engineering*, 9(1), Article 104608.

Silva, K., Mauro, M., Gonçalves, M., & Rocha, C. (2016). Synergistic interactions of locust bean gum with whey proteins: Effect on physicochemical and microstructural properties of whey protein-based films. *Food Hydrocolloids*, 54, 179–188.

Sui, C., Zhang, W., Ye, F., Liu, X., & Yu, G. (2016). Preparation, physical, and mechanical properties of soy protein isolate/guar gum composite films prepared by solution casting. *Journal of Applied Polymer Science*, 133(18).

Thomas, D., Nath, M. S., Mathew, N., Reshmy, R., Philip, E., & Latha, M. (2020). Alginate film modified with aloe vera gel and cellulose nanocrystals for wound dressing application: Preparation, characterization and in vitro evaluation. *Journal of Drug Delivery Science and Technology*, 59, Article 101894.

Tonyali, B., Cikrikci, S., & Oztop, M. H. (2018). Physicochemical and microstructural characterization of gum tragacanth added whey protein based films. *Food Research International*, 105, 1–9.

- Wang, L., Auty, M. A., & Kerry, J. P. (2010). Physical assessment of composite biodegradable films manufactured using whey protein isolate, gelatin and sodium alginate. *Journal of Food Engineering*, 96(2), 199–207.
- Yang, H., Li, J. G., Wu, N. F., Fan, M. M., Shen, X. L., Chen, M. T., ... Lai, L.-S. (2015). Effect of hsian-tsaο gum (HG) content upon rheological properties of film-forming solutions (FFS) and physical properties of soy protein/hsian-tsaο gum films. *Food Hydrocolloids*, 50, 211–218.
- Zohuriaan, M., & Shokrolahi, F. (2004). Thermal studies on natural and modified gums. *Polymer Testing*, 23(5), 575–579.