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REVIEW ARTICLE

Use of fenugreek seed gum in edible film formation: major drawbacks and applicable methods to overcome

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Abstract Researching on potential biopolymer sources with the aim of developing edible films with better mechanical and barrier properties has become innovative as it would be a key factor to minimize the use of synthetic polymers in food packaging. Therefore, different biopolymers such as galactomannan have been gaining attention recently. Fenugreek seed gum is a rich source of galactomannan which is minimally researched on its applicability in edible film making. The degree of galactose substitution and polymerization are the main factors that determine the functional properties of galactomannan. A strong and cohesive film matrix cannot be produced from fenugreek seed gum as its molecular interaction is weakened due to the high galactose substitution with a high galactose/mannose ratio, 1:1. Structural modifications of galactomannan in fenugreek seed gum will lead to films with the required mechanical properties. Hence, this review summarizes recent scientific studies on the limitations of fenugreek seed gum as a film forming agent and the specific modification techniques that can be applied in order to increase its film forming capability and performance.

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Abbreviations

FSG Fenugreek Seed Gum
G/M Galactose/Mannose

Introduction

Worldwide plastic waste has been reported to reach 6.3×10^{12} kg by 2015 of which 3.8×10^{12} kg are accumulated in landfills or natural environments (Reinold et al. 2020). Since food packaging accounts for around 50% of fossil fuel-derived plastics (Ncube et al. 2020), its contribution to environmental plastic accumulation is significant (Pilevar et al. 2019). Song et al. (2018) reported that the amount of food packaging waste in China had increased from 0.2×10^{9} kg in 2015 to 1.5×10^{9} kg by 2017 of which 75% are plastic containers made from polypropylene and polystyrene. Due to this, research has focused on developing bio-based packaging materials that have minimal impact on the environment (Jeevahan et al. 2020; Mohamed et al. 2020). Recent worldwide consumer demand has been for natural, safe high-quality food products and food packaging which doesn't cause environmental pollution (Wensing et al. 2020). Therefore, edible packaging resulting in zero waste disposal has been getting attention recently. A packaging material that is suitable for human consumption is referred to as an edible packaging. Two main categories of edible packaging exist first one is edible coating which is formed directly on the food surface

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as a thin layer and the other is edible film which is formed separately as a wrapping material for the food products (Jeevahan et al. 2020). Such an edible film should not alter the components or the processing methods of the original food product (Gahruie et al. 2020) while protecting food products from deterioration by controlling water vapor and gas transfer, maintaining their quality and extending shelf life (Lan et al. 2021).

Spinning, extrusion and solvent casting are the methods commonly used in edible film formation. Briefly in spinning, a polymer solution is passed through a spinneret under pressure to form fibrous membranes while extrusion uses elevated temperature and shear force in order to form cohesive film matrices by passing the solution through an extruder (Li et al. 2020). Solvent casting is a common and simple method that is frequently used at the laboratory level for film making of which film forming solutions are manually poured into petri plates or Teflon coated plates followed by drying and detaching (Li et al. 2020) as shown in Fig. 1. However, the casting process is a high energy consuming method since the water in cast films need to be evaporated as well as, leading to increased in drying time. Similarly, the difficulty in thickness controlling of cast films is an issue with the solvent casting method which makes it inapplicable in edible film formation on an industrial scale (Jeevahan et al. 2020).

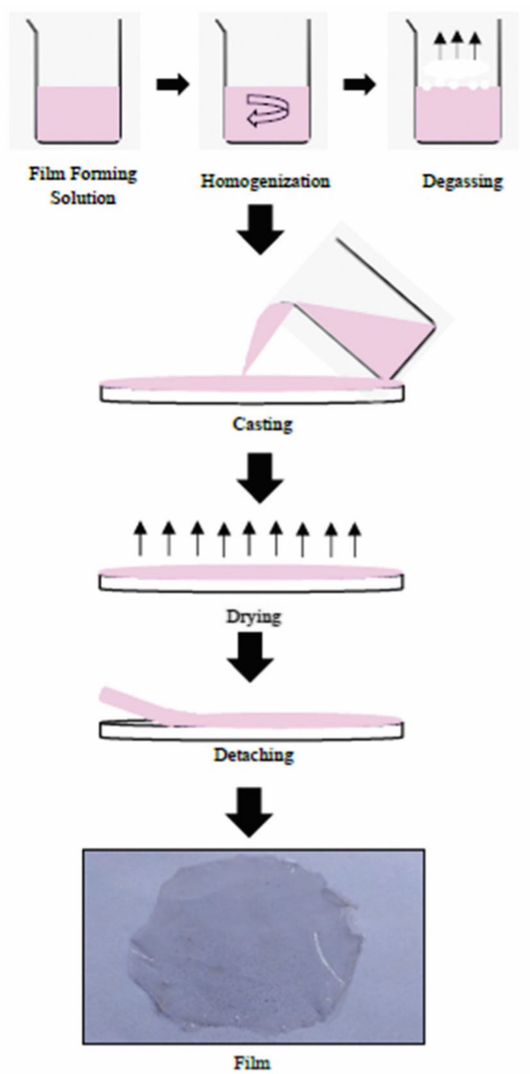

Fig. 1 Film forming process using solvent casting

A packaging film should fulfil requirements such as high mechanical strength, high barrier properties, excellent physical properties and easy production while being low cost. With edible packaging films, several additional requirements need to be considered such as safe for human consumption, health benefits and excellent sensory attributes. Nevertheless, when comparing biopolymer-based films with synthetic polymers, although the biodegradability and edibility are positive factors, both pros and cons can be identified in the sphere of physical, mechanical and barrier properties of bio-based films (Jeevahan et al. 2020; Otoni et al. 2017). Since the edible film formation is at the research level, the satisfactory parameters of these properties in determining the suitability of a film with respect to food products are still not standardized to the best of our knowledge. These characteristics of edible films vary with the biopolymer source used in the manufacture of the edible material namely proteins, lipids and polysaccharides (Mohamed et al. 2020). Even though, protein based edible films have poor moisture barrier properties which limit their applications in food packaging (Cakmak et al. 2020; Wu et al. 2020), their unique structure, mechanical properties and optical properties are better than that of polysaccharide-based films but still not equivalent to synthetic polymers (Eghbal et al. 2016). Although high moisture barrier properties can be achieved from lipid-based films due to the low polarity of lipids, the hydrophobicity of lipids makes the films more brittle and thicker (Gahruie et al. 2020; Hassan et al. 2018). The films made from polysaccharides have good gas barrier properties but not water vapor barrier properties (Mohamed et al. 2020). Among the polysaccharides used for edible film formation such as starch, cellulose and pectin, plant-based gum and mucilage have become a promising area of current research due to having beneficial properties in film making (Beikzadeh et al. 2020).

Gum and mucilage are considered plant hydrocolloids or hydrophilic compounds which form viscous solutions with water and are composed of monosaccharides joined with organic acids. However, there are some slight differences between gum and mucilage as indicated in Fig. 2 (Beikzadeh et al. 2020). Guar (Dhumal et al. 2019), locust bean (Kurt et al. 2017), tara (Chen et al. 2020), fenugreek (Salarbashi et al. 2019), cress (Jouki et al. 2013a), basil

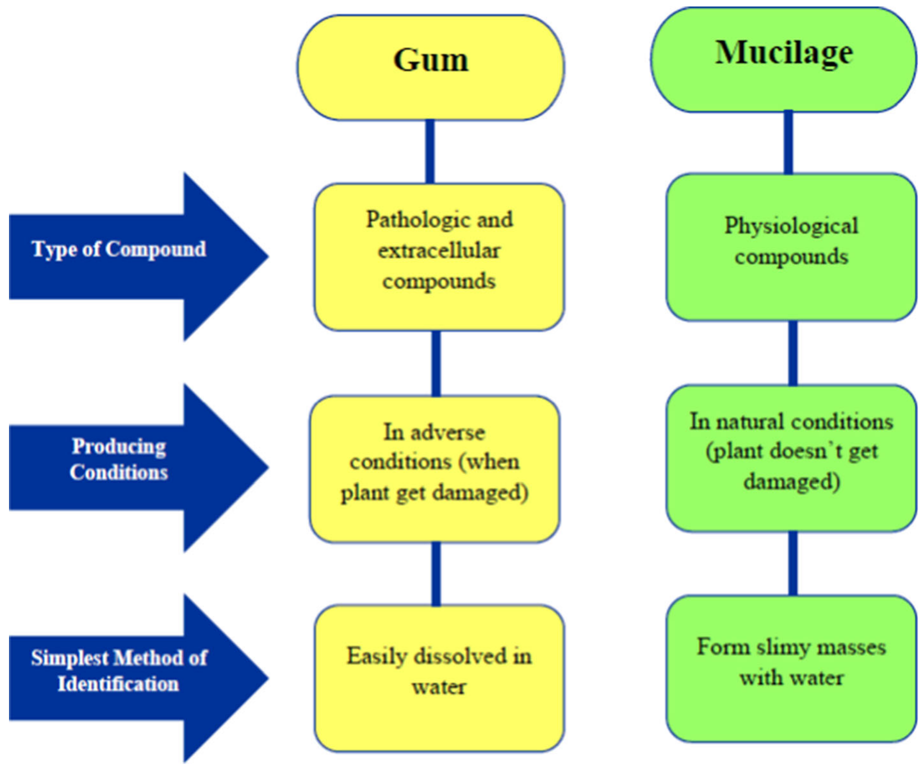

Fig. 2 The main differences between gum and mucilage (Beikzadeh et al. 2020)

(Khazaei et al. 2014), cassia (Cao et al. 2018), chia (Urbizo-Reyes et al. 2020) and quince (Jouki et al. 2013b) seeds are some sources of gum and mucilage which are currently being used and/or being studied for possible edible film making. Guar, Locust bean, Tara and Fenugreek seed gums are considered as the main sources of galactomannan, which is used (Kontogiorgos 2019) as thickening, stabilizing and emulsifying agents in the food industry (Cerqueira et al. 2011). Galactomannan extracted from Guar, Locust bean and Tara seeds has already been identified as a possible compound for edible film formation (Cerqueira et al. 2011). Accordingly, a number of research studies have been conducted throughout the world in order to improve the properties of films made from these galactomannan sources (Antoniou et al. 2015; Harnkarnsujarit 2017; Nuvoli et al. 2020; Saberi et al. 2016; Tahir et al. 2019). However, relatively very limited scientific studies (Nieto 2009; Salarbashi et al. 2019) have reported on fenugreek based edible film manufacturing and their properties. Dearth of technical know-how is the major drawback in utilizing Fenugreek seed gum because, a limited number of researches have been carried out on its galactomannan. A proper understanding of the chemical structure and properties of galactomannan in fenugreek seed gum would facilitate to widen its applicability in edible film manufacture (Brummer et al. 2003). Hence, this review summarizes the challenges with fenugreek seed gum in edible film making and applicable methodological and structural modifications of fenugreek seed gum in order to increase its film forming capability.

Fenugreek seed gum (FSG)

Fenugreek (*Trigonella foenum–graecum* L.); a leguminous crop that belongs to the family Fabaceae is cultivated mainly in India (Dhull et al. 2020). The seeds (Fig. 3a) are slightly bitter in taste and used in spice blends. According to the findings of Naidu et al. (2011) and Wani and Kumar (2018), fenugreek seeds contain 11–12% moisture, 23–26% protein, 6–7% fat, 3–4% total ash, 30–38% soluble dietary fiber and 20–25% insoluble dietary fiber. The soluble fiber in fenugreek seeds (Fig. 3b) is mainly galactomannan (Dhull et al. 2020).

When fenugreek seeds are immersed in water, a transparent gum leaches forming a viscous solution (Salarbashi et al. 2019), which can be extracted by ethanol (Brummer et al. 2003). The average extraction yield of FSG is varied according to the geographical areas where plants are cultivated (Salarbashi et al. 2019). According to Brummer et al. (2003), the extractable yield of FSG from Canadian seed was 22% while Iurian et al. (2017) reported that the extractable yield from Romanian grown fenugreek seed was 15%. The extraction and purification of FSG is

(a)

(b)

Fig. 3 **a** Fenugreek seeds. **b** Gum extracted from fenugreek seeds (Before drying)

challenging because it is attached to the protein matrix of the seed (Brummer et al. 2003). Pronase treatment, phenol solvent treatment etc. are some of the techniques that can be used to purify FSG by removing protein up to 0.5–0.6% and 0.1–0.2% respectively. However, no technique yet has been developed for the complete removal of attached protein (Salarbashi et al. 2019). The commercially available FSG contains 80% polysaccharides and 5% proteins which can be reduced up to 0.6% by using pronase treatment in the FSG manufacturing process (Salarbashi et al. 2019).

Chemical composition of FSG

The FSG contains 91–92% carbohydrate polymers, 10–11% moisture, 4–5% protein, 2–3% fat and 1–2% ash (Liu et al. 2020). The monosaccharide composition of FSG varies with age of the plant, growing conditions and the method of purification (Salarbashi and Tafaghodi 2018). As summarized in Table 1, galactose and mannose are the most abundant monosaccharides in FSG which is structured as galactomannan and is the storage polysaccharide of fenugreek seeds similarly to guar, tara and locust bean gum (Brummer et al. 2003).

Chemical structure of FSG

Galactomannans (Fig. 4) consist of a β-(1–4)-D mannan backbone with α-(1–6) D-galactose molecule substitution at O-6 position of D-mannopyranosyl residues in which the mannose to galactose ratio is dependent on the source (Rodriguez-Canto et al. 2020; Wei et al. 2015). In FSG, each mannose unit in the linear backbone is combined with a galactose unit (Nieto 2009) which means its Galactomannan/ Mannose (G/M) ratio is 1:1. Galactomannans are water soluble heteropolysaccharides, which form highly viscous and stable aqueous solutions (Mikkonen et al. 2007). Therefore, the thickening, binding, gelling, emulsifying and suspending properties of FSG is due to the presence of a substantial amount of galactomannan (Srivastava and Kapoor 2005).

Use of galactomannan in edible film making

The physicochemical properties of galactomannan such as viscosity, water solubility are mainly affected by the degree of branching or the galactose substitution, the G/M ratio. In addition, the degree of polymerization, molecular weight and steric hindrance also affect the properties of galactomannan (Busch et al. 2015; Liu et al. 2020; Pollard and Fischer 2006). Relatively strong interactions between polymer chains can lead to form cohesive and strong film matrices. The presence of more galactose subunits can force the mannan chains apart because of the steric hindrance and weaken the interactions between individual polymer chains. Also, polymerization of unsubstituted mannose units and their block wise distribution can form cooperative interactions between polymer chains (Mikkonen et al. 2007). Therefore, for the formation of cohesive film structures, the low substitution of galactose and the high degree of polymerization which means long chain polymeric structure, is preferred (Mikkonen et al. 2007). As the molecular weight of galactomannan increase so does the viscosity (Mikkonen et al. 2007) which may

Table 1 Monosaccharide composition of crude, purified, and protein free fenugreek gum (Youssef et al. 2009)

Type of FSG	Protein (%)	Rhamnose (%)	Arabinose (%)	Galactose (%)	Glucose (%)	Mannose (%)
Crude FSG	3.74 ± 0.03	0.16 ± 0.00	0.54 ± 0.04	26.22 ± 0.02	0.64 ± 0.01	31.40 ± 0.03
Purified FSG	1.10 ± 0.02	0.00	0.00	33.91 ± 0.01	0.00	41.57 ± 0.01
Protein free FSG	0.16 ± 0.02	0.00	0.00	32.87 ± 0.02	0.00	41.84 ± 0.01

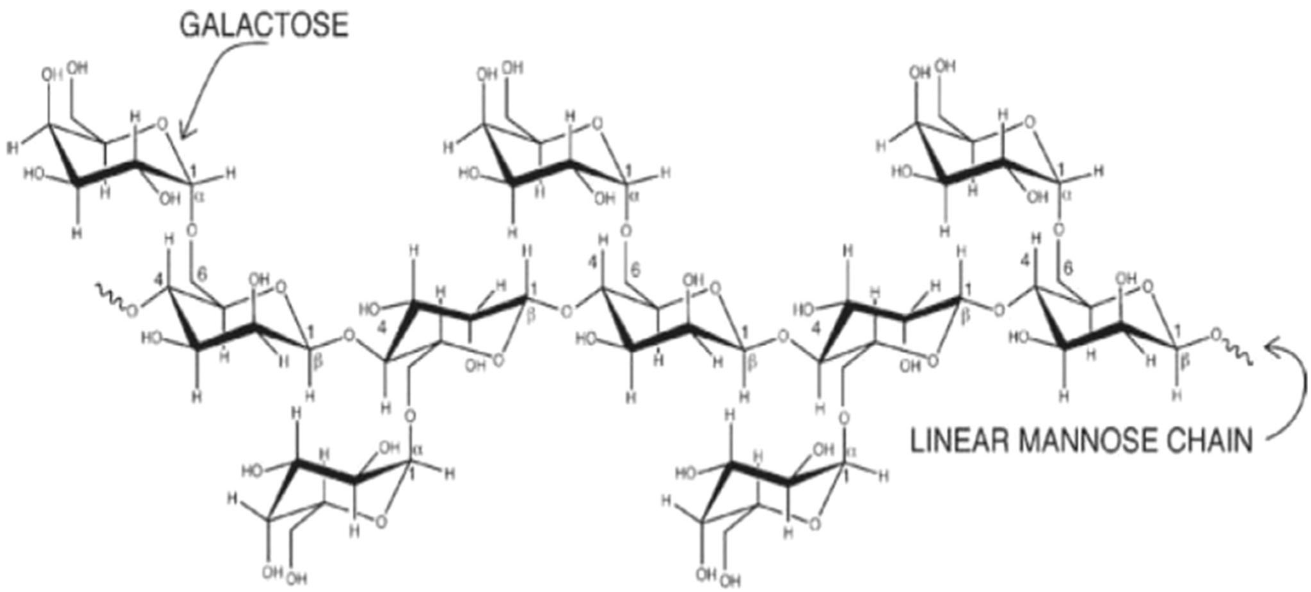

Fig. 4 Chemical structure of fenugreek seed gum (Nieto 2009)

Table 2 G/M ratios and molecular weights of different gums containing galactomannan

Type of the gum	G/M ratio	Molecular weight (g/mol)	Reference
Guar gum	0.55	2.2×10^5	Cerqueira et al. (2011) and Prado et al. (2005)
Locust bean gum	0.25	3.1×10^5	Cerqueira et al. (2011) and Prado et al. (2005)
Fenugreek seed gum	1.00	5.6×10^5	Cerqueira et al. (2011) and Salarbashi et al. (2019)

facilitate film formation. The degree of galactose substitution and the polymerization are varied with the galactomannan source. The G/M ratios and the molecular weights of guar, locust bean and fenugreek seed gums are indicated in Table 2 (Cerqueira et al. 2011; Prado et al. 2005; Wu et al. 2017). Many research studies have been carried out on edible film making from the galactomannan sources of which, the galactose substitution is low. Whereas, guar gum has drawn attention widely for making edible films by incorporating other polymer sources to achieve enhanced film properties. For instance, Saberi et al. (2016) optimized a method to form an edible film from pea starch by incorporating 0.3, 2.5 and 25% w/w guar gum, pea starch and plasticizer (glycerol) respectively. The outcome of the study was that incorporating guar gum into pea starch formed a composite film as an applicable alternative for the food packaging industry with suitable physical and optical properties. To achieve a film with good mechanical properties Sorde and Ananthanarayan (2019) incorporated coconut protein into guar gum, by treating the material with transglutaminase enzyme they improved the mechanical properties such as tensile strength and elongation from 1.76 MPa to 3.79 MPa and 0.56 cm to 1.84 cm respectively. Moreover, as demonstrated by Chu et al. (2020) study, stronger mechanical strength can be achieved from a guar gum based film by positively charging the gum and the resulting film had the tensile strength of 65.41 MPa. Tara gum which is also a galactomannan it can produce a relatively strong film matrix, as its galactose substitution is comparatively low, the G/M ratio is 1:3 (Cerqueira et al. 2011). The mechanical and barrier properties of the formed films are poor resulting in limited commercial applications (Tahir et al. 2019). A comparative study was conducted by Antoniou et al. (2015) through synthesizing tara gum films by incorporating nano chitosan to obtain better mechanical and barrier properties. The tensile strength of this composite film increased by 35.73 MPa while the water vapor permeability and water solubility were reduced by 22.7% and 74.3% respectively. Nuvoli et al. (2020), incorporated Tara gum powder into fish gelatin-based films which resulted in improving the mechanical and thermal properties. However, further investigations should be done on the effect of galactomannan contained in seed gums on the glass transition temperatures of films (Harnkarnsujarit 2017).

The intrinsic viscosity value of galactomannan is dependent on the molecular hydrodynamic volume attributed to the repulsive and attractive forces between chain segments. These interactions can be modified by introducing different electrostatic charges by adding sugars or salts which is also applicable in film making to achieve better properties. According to the findings of Behrouzian et al. (2014), the addition of sucrose up to 20–40% and lactose up to 5–15% could increase the intrinsic viscosity of seed gum solutions containing galactomannan as these sugars can boost the random coil formation of seed gum. However, the addition of salts such as NaCl, $CaCl_2$ has decreased the intrinsic viscosity with the increase of their concentrations which is due to the shielding effect of charges on macromolecular chains with the availability of added ions.

Temperature is another important factor that affects the solubility of galactomannan. With the increase in temperature, the solubility is increased by further dissolving of galactomannan with higher molecular weights or a low level of galactose residues (Liu et al. 2020). It is reported that guar gum can achieve its full viscosity in cold water while locust bean gum is partially soluble in cold water and needs heating to be totally dissolved (Dakia et al. 2008; Liu et al. 2020). Although, Singh et al. (2020) suggested carboxymethylation as a method to increase the water solubility of locust bean gum, the significant reduction in viscosity is the main drawback with this method when applied to film making. The solubility of galactomannan depends on the solubilization time, solute concentration, particle size and the availability of impurities (Rodriguez-Canto et al. 2019). The viscosity of the galactomannan solutions can be increased by reducing the number of galactose subunits up to a certain point and further reduction results in the decrease in viscosity due to the formation of polymers with low solubility (Mikkonen et al. 2007). In the galactomannan structure, where the average unsubstituted chain length is more than six units of mannose, the interactions are dependent on the unsubstituted areas of the backbone. If the average unsubstituted length is below six mannose units, interactions are dependent on the structural features associated with unsubstituted regions. As the interactive properties are influenced by the degree of polymerization and the viscosity of the galactomannan solution, which can be increased by increasing the molar mass, films with better cohesive strength can be achieved by utilizing galactomannan with long chain polymeric structures (Mikkonen et al. 2007). Hence, the study of galactomannan beneficial in making strong and cohesive films is important to make FSG an industrially applicable film forming agent since limited research studies exist in the area.

Use of FSG in edible film making

Identified drawbacks of FSG in edible film making

As reviewed above, the film making capability of galactomannan depends on its degree of galactose substitution and polymerization. Although, galactomannan in FSG has a higher molecular weight, as its G/M ratio is comparatively high, it can form a stronger hindrance to interpolymer chain association (Salarbashi et al. 2019). When the G/M ratio of FSG is 1:1, every mannose unit in the linear backbone is substituted with galactose and the intermolecular hydrogen bonds between polymer chains are weakened. Hence, FSG can't produce cohesive film structures (Nieto 2009). Further, the galactomannan with a higher G/M ratio such as in FSG produces films with lower water solubility and more oxygen permeability (Salarbashi et al. 2019). Though the water solubility of FSG is 82.82%

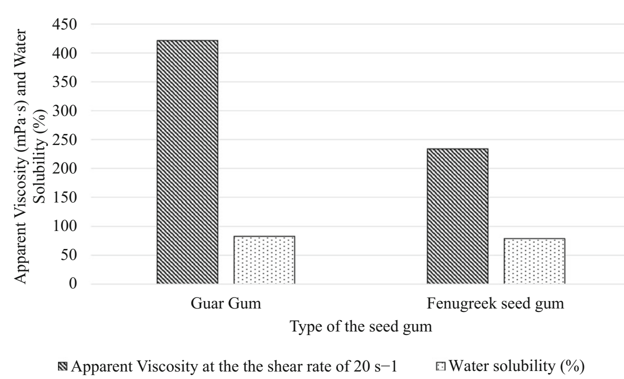

Fig. 5 Viscosity and solubility differences between guar gum and fenugreek seed gum (Haddarah et al. 2014; Liu et al. 2020; Pollard and Fischer 2006)

at 25 °C is higher than that of guar gum (78.75%) at the same temperature, the apparent viscosity of FSG is 233.95 mPa·s at the shear rate of 20 s^{-1} is lower than the guar gum (421.91 mPa·s at the same shear rate) as indicated in Fig. 5 which leads to forming a weaker film matrix. This scenario occurs due to the difference between two particular gums to the extent of galactose substitution and their molecular weights (Haddarah et al. 2014; Liu et al. 2020; Pollard and Fischer 2006).

The slow hydration of FSG due to poor hydrophilic properties causes it to take longer to completely dissolve into a homogeneous solution which also affects film forming (Salarbashi et al. 2019). The unpleasant flavor and color of fenugreek seed gum can limit its industrial applications (Salarbashi et al. 2019) because it negatively affects the consumer acceptability. As the film is consumed along with the food, it is essential to have excellent sensory attributes with none to minimal impact to the product.

Hence, it is needed to identify relevant modification techniques in order to overcome these undesirable effects of FSG in edible film making.

Modification methods of FSG to make applicable in edible film making

Enzymatic treatment

Enzymatic treatments can be used to enhance the functional properties of galactomannan by structural modifications. Endomannanase (EC 3.2.1.78) enzyme cuts the internal linkage between mannose units in the backbone, and α-galactosidase (EC 3.2.1.22) cleaves the galactose subunits in order to decrease both degree of polymerization and degree of substitution. Thereby, the gelation properties of galactomannan can be increased (Mikkonen et al. 2007). By the treatment of α-galactosidase, the galactose substitution of FSG can be reduced to an optimum level which may lead to an increase in the film making capability. Hence, the reduction of the G/M ratio may positively affect making strong film structures with better mechanical and barrier properties. Therefore, scientific studies are needed to identify the optimum G/M ratio and degree of polymerization for the galactomannan in FSG with a view to achieve better film forming properties.

Extrusion

The hydration of FSG is slower than most other plant-based gums as the FSG molecule is not electrostatically charged (Salarbashi et al. 2019). Therefore, scientific studies have focused on modification methods of FSG such as extrusion which can increase the exposure of hydrophilic groups to react with water molecules (Chang et al. 2011; Roberts et al. 2012). Research findings indicate that the hydration capability and water solubility of FSG were increased due to the extrusion process which also affects preparing film forming solutions (Chang et al. 2011; Roberts et al. 2012). In addition, according to Chang et al. (2011) and Pasqualone et al. (2020), extrusion process can remove the unpleasant flavor and color of FSG which facilitates its industrial applications as an edible film. Crude and purified FSG have better emulsification properties than protein free FSG because proteins can impart emulsification ability to the polysaccharides (Hamdani et al. 2019; Salarbashi et al. 2019). Moreover, FSG can form emulsions which are more stable than 11 commercially available gums (Memis et al. 2017). Since FSG can act as an emulsifier, it will be beneficial in developing composite edible films by improving the stability of dispersed biopolymers to form a homogeneous film forming solution. Chang et al. (2011) also reported that the effect of the extrusion process on the emulsification capability of FSG is not considerable. Therefore, extrusion is an applicable technique to overcome several identified drawbacks of FSG in film making.

Incorporation of nanoparticles

Introducing nano SiO_2, TiO_2, Ag^+ particles and nano-clays in film forming has been studied in the recent past in order to overcome some shortcomings to enhanced mechanical and barrier properties with lower moisture sensitivity and higher thermal stability (Antoniou et al. 2015; Lin et al. 2020; Salarbashi et al. 2019). With the aim of increasing both strength and the properties of FSG film, Memis et al. (2017) incorporated nano-clays. Their research findings showed that as the amount of nano-clay increased, the thermal and oxygen barrier properties improved significantly. Incorporating nano clay up to 5% could form a homogeneous, smooth film structure with higher tensile strength but low elongation at breakage (Memis et al. 2017). Strong antibacterial properties against foodborne pathogens (*Listeria monocytogenes*, *Escherichia coli*, *Staphylococcus aureus* and *Bacillus cereus*) have been exhibited by FSG based nanocomposite films which could lead to the development of precise antimicrobial food packaging material (Memis et al. 2017). Therefore, the incorporation of nano clays in forming FSG films can lead to making industrially applicable food packaging film with antimicrobial effects. However, further studies are needed on improving the utilization of FSG in making cohesive, strong film matrices.

Conclusion

FSG is a source of galactomannan which can be applied in edible film making with few modifications. The main identified drawback of FSG film formation is the high G/M ratio which leads to weakened film matrices. Even though, FSG has a high degree of polymerization, due to its high galactose substitution the viscosity of FSG is lower when compared to other gums which have lower molecular weights. Hence, the main requirement is to reduce the galactose substitution in FSG galactomannan which can be achieved by α-galactosidase enzymatic treatment. Therefore, further scientific studies are needed to identify the optimum degree of galactose substitution and polymerization needed for the formation of strong and cohesive films with better mechanical and barrier properties from FSG. Further, extrusion is a process that can improve the hydration ability of FSG galactomannan and remove its unpleasant flavor without affecting its emulsifying property which can facilitate the film formation and its industrial applicability. Additionally, the incorporation of nanoparticles is an attractive method in improving film attributes of FSG galactomannan. However, further studies are essential to identify the applicability of enzymatic treatment, extrusion methodologies and introducing nanoparticles in edible film making from FSG. In addition, it is important to focus on other possible modification techniques to convert FSG galactomannan to an industrially applicable film forming agent.

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Code availability Not applicable.

Declarations

Conflict of interest No potential conflict of interest was reported by the authors.

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