MICRO- AND NANOENGINEERED GUM-BASED BIOMATERIALS FOR DRUG DELIVERY AND BIOMEDICAL APPLICATIONS

Edited by
Sougata Jana
Subrata Jana

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CHAPTER

14

Tamarind gum as a wall material in the microencapsulation of drugs and natural products

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14.1 Introduction

14.1.1 Components of tamarind tree, tamarind pulp, and seed

Tamarind (*Tamarindus indica* L.) is native to the dry savannas of eastern tropical Africa, probably from the island of Madagascar. It was introduced to India by Arab traders, where it adapted perfectly to the agro-ecological conditions of the country's coastal plains. The tamarind was domesticated in the East Indies (Southeast Asia) and the islands of the Pacific. It was introduced to the New World by the Spanish and Portuguese in the 18th century (Rao and Mathew, 2012; Sharma and Bhardwaj, 1997). The main tamarind-producing areas in Mexico are located on the coastal plain of the Pacific Ocean coast and to a lesser extent on the coasts of the Gulf of Mexico. It is exploited commercially for the production of fruit in Colima, Guerrero, Oaxaca, Michoacán, Jalisco, and Veracruz.

Mexican tamarind is a slow-growing and large tree; in optimal conditions of development, the trees reach 24–30 m in height. The tamarind is a tree with evergreen foliage (perennifolio); however, in very dry climates it behaves as a subdeciduous, as it can lose its foliage for a short period of time during hot months. It is a long-lived tree, since it can live more than 200 years.

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The trunk is short, thick, and straight; the bark has cracks along the trunk and main branches, and presents different shades ranging from ash gray to dark brown (Orozco, 2001).

The leaves are pale to dark green, alternate and parapinnate; they are 7.5–15 cm long and have 10–20 pairs of leaflets. The flowers are hermaphroditic, pale yellow with red or orange veins and measure 2–2.5 cm in diameter, formed in small clusters 5–10 cm long and with 8–14 flowers per cluster (Parrotta, 1990; Viveros-García et al., 2012). The fruit is an indehiscent pod (remains closed when ripe), protruding and oblong, cinnamon brown or greyish brown, slightly curved and flattened. It is 7–20 cm long and 1–3 cm wide. The fruits are formed in abundance along the branches or in the terminal parts of them. When the fruits ripen, their skin is brittle and can break easily. The pulp is the edible part of the fruit, brown in color, firm, viscous, granular, with a pleasant bittersweet flavor and high content of sugar and acid. It has 1–12 seeds of a bright brown color, flat, oval and united by fibers that are located in the pulp of the fruit. In a kilogram there are from 2000 to 2500 seeds (Bhattacharya et al., 1993; Viveros-García et al., 2012).

The pulp of the tamarind fruit has been a very important culinary ingredient in Mexico for a long time. On average, 65.95% moisture, 2.35% protein, 0.48% fat, 26.2% sugars, 0.45% sucrose, 2.7% cellulose, and 1.87% other compounds can be found on average in ripe tamarind pulp. About 60 volatile compounds have also been found in tamarind pulp. There is also a variety of tamarind with a sweeter flavor with a reddish hue. This sweet taste can be caused due to a lower presence in the amount of polyprotonated acids (Shankaracharya, 1998; Viveros-García et al., 2012).

On the other hand, tamarind seed represents on average 35% of all fruit; this is made up of two main parts: the testa, representing 30% of the seed, and the endosperm or kernel, representing the remaining 70%. Analyzing the composition of the seed, it was possible to find proteins in 18%, fats in 7%, carbohydrates in 69%, fiber in 3%, and other compounds with 3% (Bhattacharya et al., 1993; Kumar and Bhattacharya, 2008; Tsuda et al., 1994). The protein contained in the endosperm is abundant in lysine, glutamic acid, aspartic acid, glycine, leucine, and potassium, but deficient in amino acids that contain sulfur groups (Bhattacharya et al., 1994; Kumar and Bhattacharya, 2008).

14.1.2 Technologies of tamarind seed gum extraction

Tamarind seed gum is a polysaccharide which main compound is xyloglucan. It was discovered during World War II when the Forest Research Institute, Dehra Dun, was searching for new sizing materials; nowadays it has different applications including in the food, pharmaceutical, confectionary, cosmetic, and textile industries. This compound was commercially isolated in 1941 by Daurala Sugar Works, India, when it was proposed as a pectin substitute. Tamarind seed gum was commercially produced in 1943 and the extraction method in laboratories was first devised in 1945 (Rao and Mathew, 2012; Sharma and Bhardwaj, 1997).

Most common extraction technologies involve some of the following steps: the first step is washing the crude tamarind seeds with water and subsequent heating to make the testa (seed coating) brittle and friable. The seeds are then decorticated to leave the heavier crushed endosperm, which is finally ground to yield tamarind kernel powder. After this, it is boiled with about 30–40 times its weight of water for about 30–40 min under agitation and allowed to sit

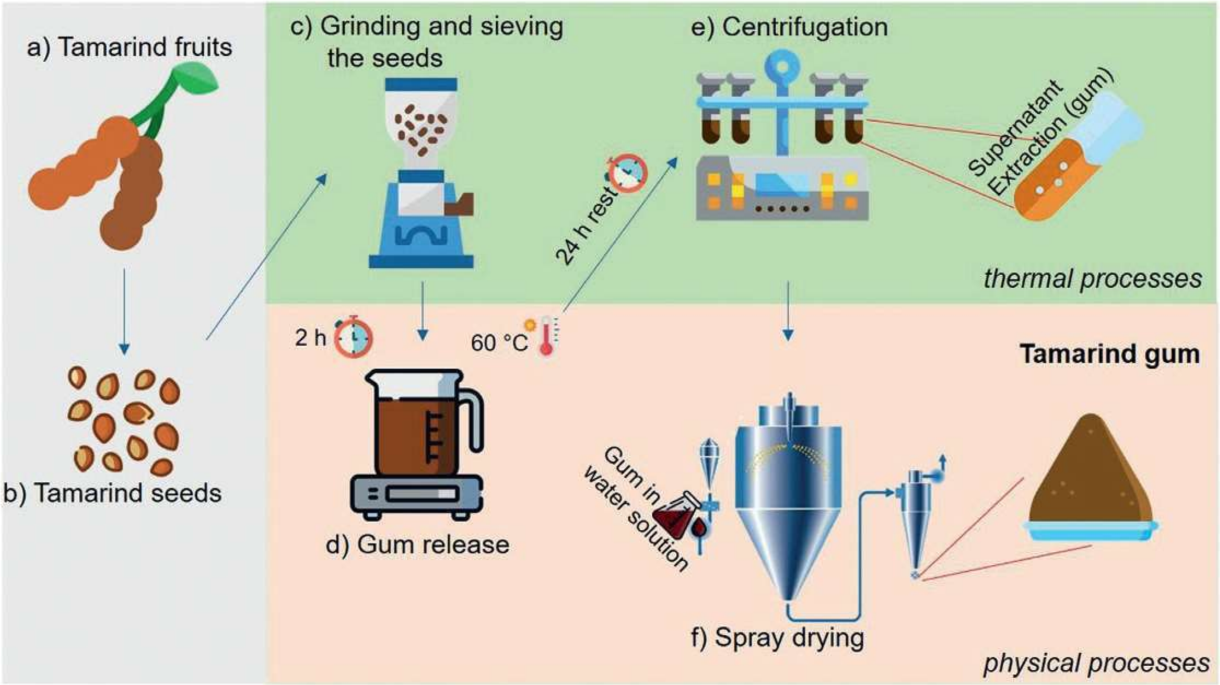

FIG. 14.1 Eco-green-based technology of tamarind gum extraction. Method based on Alpizar-Reyes et al. (2017a): (A and B) The seeds are extracted manually from mature pods of tamarind, then (C) milled and grounded through a 355 μm mesh. (D) 20 g of milled tamarind seeds are dispersed in 1 L of bidistilled water and stirred for 2 h at 60 °C, then the dispersion is kept at 20 °C for 24 h for the gum release, (E) and centrifuged for 8 min at 524 × *g*. The supernatant represents the gum, (F) which is fed at 40 mL/min to a spray-drier with an inlet temperature of 135 ± 5 °C, outlet temperature at 80 ± 5 °C and injecting compressed air at 4 bar. Tamarind gum extracted by an eco-green-based technology uses thermal and physical processes, avoiding chemical pollution.

overnight in a settling tank in order to precipitate and settle out fibers and for the most part of the proteins. The next step is crucial: the noneco-green gum is precipitated with solvents as ethanol and some salts and dried to a constant weight using some of the drying technologies available (Fig. 14.1) (Nishinari et al., 2009; White and Rao, 1953; Yamatoya and Shirakawa, 2003).

Through the methodologies used for tamarind gum extraction, different chemical reagents have been used at commercial and laboratory scale. The original method proposed by Ghose and Krishna (1942) was pouring tamarind kernel powder into cold water at a ratio of 1:10, after that, it was added to boiling water at a ratio of 1:30 or 1:40 and maintained at boiling temperature for 20–30 min; it was then strained through cloth and sulfur dioxide was added to bleach the gum. The mix was left overnight before centrifugation and drum drying. It was observed that sulfur dioxide caused the original gelling properties of the polysaccharide to be lost; very tough gels were formed instead if this treatment was not carried out. Later, it was suggested to add an organic acid to the boiling water in order to obtain the desired degradation degree of the gum that enabled soft gels. The yield obtained with this process was higher than 50% of the tamarind kernel powder. Precipitation with ethanol is an alternative to drum drying; using this technique, the polysaccharide is usually filtered mechanically, dried in an air oven, and finally milled to homogenize particle size. Another method utilized for extraction and purification of the tamarind seed gum consists of dilution, dialysis, and precipitation. From this process a fine white powder with less than 1% protein content is obtained (Rao et al., 1973).

A method patented by Jones et al. in the United States consists of defatting of the tamarind kernel powder using C-6 or C-8 aromatic hydrocarbons or C-1, C-2, or above halogenated lower hydrocarbons or C-1 or C-5 mono or dihydroxy alcohols. The defatted powder is then recovered by filtration or centrifugation and dried. After that, flow properties of the powder are enhanced with salicaceous materials, and the powder is grounded to reduce its size below 100 μm and further classified into three groups: fine fraction rich in protein, moderately fine fraction rich in polysaccharides, and coarser fraction rich in mechanical properties. Finally, the tamarind seed gum can be isolated from the moderately fine fraction (Gupta et al., 2010).

Solvent-induced precipitation was proposed to extract tamarind gum with the next steps: first, the seeds were heated in sand (seed: sand ratio of 1:4) to remove the testa, then the seeds were crushed and soaked for 24 h, after that the dispersion was boiled for 1 h and left to sit for 2 h. The seeds were squeezed and the gum separated was mixed with equal quantity of acetone to precipitate it. Finally, the mucilage was dried at 50 °C, powdered, and passed through sieve number 80. The yield obtained with this procedure was 78% (w/w) from tamarind seeds (Phani Kumar et al., 2011).

Another alternative to solvent employment is to isolate the tamarind seed polysaccharide using a common method. Firstly, a reflux system was used with water as a solvent at 70 °C for about 6 h. The extract was then pressed in cheese cloth bag, cooled to 4 °C, and mixed with alcohol 2:1 (v/v) to achieve tamarind seed polysaccharide precipitation. It was stirred continuously for 15 min and then left to stand for 2 h to allow the formation of coagulates which were further filtered, washed with alcohol, and pressed. Finally, the pressed product was dried at 35–45 °C in a hot air oven, grounded, and sieved through sieve number 20 (Bansal et al., 2013).

There has been considerable progress in the development and usage of various extraction and purification techniques for tamarind gum; their selection is crucial for this polysaccharide's physicochemical and techno-functional properties. Additionally, the environmental impact of complex extraction techniques opened a new window on implementing new eco-green extraction technologies with more minor ecological effects.

Some environmentally friendly methodologies reported for the extraction of tamarind gum are described as follows. Alpizar-Reyes et al. (2017a, b) obtained tamarind gum by manually extracting the tamarind seeds, and milling and grinding them through a mesh. The powdered seeds were added to bidistilled water to achieve a weight ratio of 1:10, and the aqueous dispersion was heated and stirred for 10 min. Additional bidistilled water was then added to reach a weight ratio 1:40 and heated at 80 °C for 60 min. The dispersion was left to stand, centrifuged, and finally, the supernatant was spray-dried (Fig. 14.2) yielding ~29% (w/w) of tamarind gum.

Other solvent-free methods have been also tested, i.e., methods that obtained yields for tamarind gum of ~27%–32% using two distinct methods. The first method consisted of tamarind seeds that were washed and dried at 100 °C, and grinded using a blender. The resultant powder was mixed with water and poured into boiling water, stored overnight, and centrifuged. The supernatant phase was precipitated with aqueous ethanol (95% *v*/v) and then, dried at 50 °C for 4 h in an oven. The second method consisted of the use of

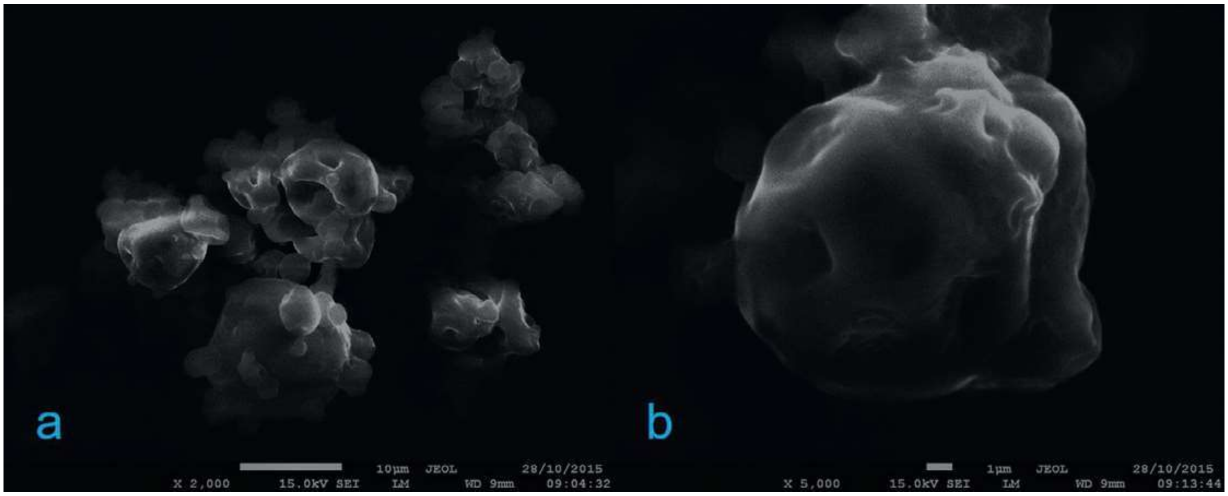

FIG. 14.2 Scanning electron microscopy for tamarind gum (A) at 2000 magnifications and (B) at 5000 magnifications.

defatted tamarind seed powder followed by the same procedure previously described (Chawananorasest et al., 2016).

Different techniques for tamarind gum extraction include that reported by Limsangouan et al. (2019), who applied subcritical water and compared some characteristics from the obtained tamarind gum with that extracted by the "conventional method." The latter consisted of defatting the tamarind kernel powder with hexane at the ratio of 1:5 (w/v) and extracting tamarind gum with boiling distilled water for 30 min. Once the hydrocolloid dispersion was cooled to ambient temperature, it was treated with protease enzyme. The mixture was then centrifuged at 5000 rpm for 15 min at 25 °C, and the supernatant was mixed at the ratio 1:2 (v/v) with aqueous ethanol (95% v/v) for 30 min to induce the formation of a gel-like phase, which was filtered through muslin cloth and dried in a hot air oven at 60 °C for 8 h. The resultant material was grinded and sieved with a common 50-mesh. On the other hand, extraction with subcritical water consisted of mixing the filtered defatted powder with distilled water (5%, w/v), pouring into a high-pressure resistant vessel, and heated until reaching temperatures of 100, 125, 150, 175, and 200 °C. The extracted solution was treated with protease enzyme and precipitated as the conventional method. The extracted gum exhibited higher yields (52%–62%), greater color intensity and water solubility index, but lower molecular weight, holding strength, viscosity, water absorption index, antioxidant capacity, and total phenolic content than that obtained with the conventional extraction method.

14.2 Tamarind gum characterization

14.2.1 Chemical structure and composition

The chemical structure and composition from different extraction techniques of tamarind gum have been studied by many researchers. The chemical structure of tamarind gum (Fig. 14.3) consists of a cellulose-like main chain of a branched polysaccharide with molecular weight of 700–880 kDa (Khounvilay and Sittikijyothin, 2012), composed of a β-(1,4)-D-glucan

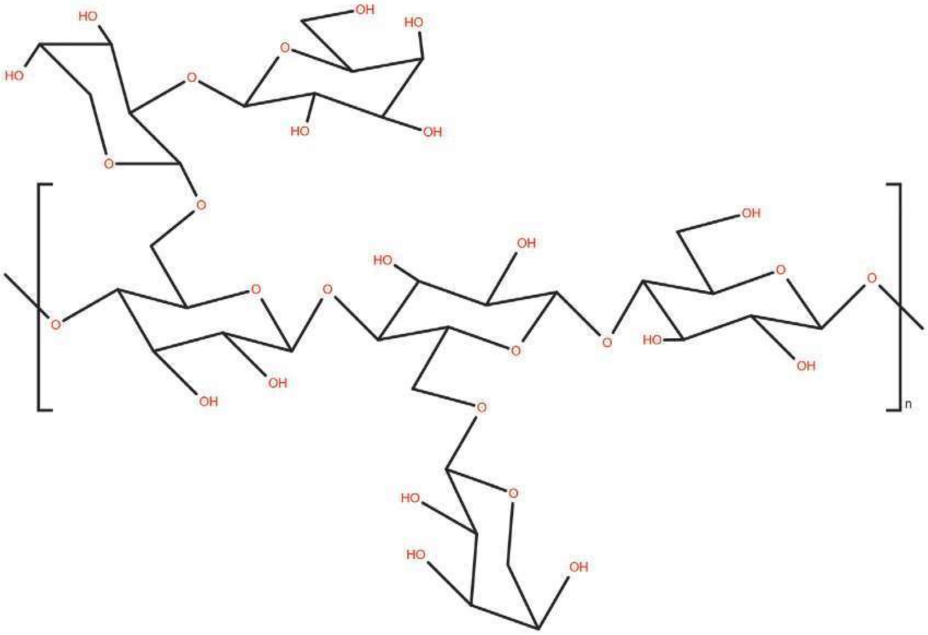

FIG. 14.3 Chemical structure of tamarind gum β-(1,4)-D-glucan backbone chain, partially substituted with side chains of α-(1,4)-D-xylopyranose and (1,6) linked [β-D-galactopyranosyl-(1,2)-α-D-xylopyranosyl] to glucose residues.

backbone chain, partially substituted with side chains of α-(1,4)-D-xylopyranose and (1,6) linked [β-D-galactopyranosyl-(1,2)-α-D-xylopyranosyl] to glucose residues, where tamarind gum is conformed from monomer units that essentially contain three types of sugars, glucose, galactose, and xylose, with a molar ratio of 2.8:2.25:1.0. A chain length of tamarind gum from 300 to 3000 glucose units was reported (Fry, 1989). Moreover, xylose units are shown to be more hydrophilic than glucose units. Due to the presence of hydrophilic and hydrophobic units, tamarind gum shows good solubility in water, even though individual macromolecules do not fully hydrate, resulting in the formation of aggregated species in water even at very dilute solutions. Tamarind gum can be chemically or enzymatically modified to assess improved rheological properties, i.e., gel formation or extra-high viscosity, where this modification has a direct correlation between the structure of a polysaccharide and its surface and functional properties (Kulkarni et al., 2017; Pardeshi et al., 2018).

The yield of the extraction of tamarind gum prior to be used as a wall material in the microencapsulation of drugs and natural products exerts a great influence on the economic aspects, where a higher performance with a lower environmental impact is always desired. In this sense, in Table 14.1, it can be seen that eco-green-based technology (solvent free) has the greatest economic advantages compared to solvent application technologies. Solvent-free processes offer greater competitiveness and a window of economic opportunity for future applications of tamarind gum as a micro-engineered gum-based biomaterial for drug delivery and for biomedical applications.

The chemical composition of tamarind gum (Table 14.2) is dominated by the carbohydrate content (70%–90%) because gums are polysaccharides by nature (Prajapati et al., 2013), in

TABLE 14.1 Yield for tamarind gum with different extraction methods.

Extraction method	(%) reported	References
Eco-green-based technology (solvent free)	29.83 ± 1.32	Alpizar-Reyes et al. (2017a)
Low solvent application (ethanol precipitation)	27.93 ± 0.23	Crispín-Isidro et al. (2019)
Medium solvent application (ethanol precipitation + hexane distillation)	22.12 ± 0.11	
High solvent application (ethanol precipitation + hexane distillation + NaOH)	18.93 ± 0.35	

Summary of the results supported by the references cited.

TABLE 14.2 Summary of the chemical composition in dry basis quantified for tamarind gum with different extraction methods used.

	Extraction method	(%) reported	References
Carbohydrates	Eco-green-based technology (solvent free)	80.25	Alpizar-Reyes et al. (2017b)
		79.76 ± 0.72	Alpizar-Reyes et al. (2017a)
		82.17 ± 0.21	Alpizar-Reyes et al. (2018)
	Milled and grounded seeds (analysis not in dry basis)	80.66	Khounvilay and Sittikijyothin (2012)
	Strong employment of chemicals (ethanol precipitation + hexane distillation + NaOH)	85.74 ± 0.75	Chandra Mohan et al. (2018)
	Low solvent application (ethanol precipitation)	79.24 ± 0.68	Crispín-Isidro et al. (2019)
	Medium solvent application (ethanol precipitation + hexane distillation)	81.29 ± 0.09	
	High solvent application (ethanol precipitation + hexane distillation + NaOH)	89.56 ± 1.48	
Glucose:xylose: galactose ratio	Milled and grounded seeds (analysis not in dry basis)	2.61:1.43:1.00	Khounvilay and Sittikijyothin (2012)
Proteins	Eco-green-based technology (solvent free)	14.24	Alpizar-Reyes et al. (2017b)
		14.78 ± 0.45	Alpizar-Reyes et al. (2017a)
		12.96 ± 0.09	Alpizar-Reyes et al. (2018)
	Milled and grounded seeds (analysis not in dry basis)	13.51	Khounvilay and Sittikijyothin (2012)

Continued

TABLE 14.2 Summary of the chemical composition in dry basis quantified for tamarind gum with different extraction methods used—cont'd

	Extraction method	(%) reported	References
	Strong employment of chemicals (ethanol precipitation + hexane distillation + NaOH)	0.58 ± 0.08	Chandra Mohan et al. (2018)
	Low solvent application (ethanol precipitation)	12.11 ± 0.14	Crispín-Isidro et al. (2019)
	Medium solvent application (ethanol precipitation + hexane distillation)	16.53 ± 0.19	
	High solvent application (ethanol precipitation + hexane distillation + NaOH)	9.00 ± 0.12	
Fats	Eco-green-based technology (solvent free)	4.96	Alpizar-Reyes et al. (2017b)
		4.76 ± 0.36	Alpizar-Reyes et al. (2017a)
		4.28 ± 0.17	Alpizar-Reyes et al. (2018)
	Milled and grounded seeds (analysis not in dry basis)	5.76	Khounvilay and Sittikijyothin (2012)
	Low solvent application (ethanol precipitation)	0.10 ± 0.01	Crispín-Isidro et al. (2019)
	Medium solvent application (ethanol precipitation + hexane distillation)	0.08 ± 0.01	
	High solvent application (ethanol precipitation + hexane distillation + NaOH)	7.20 ± 0.31	
Ashes	Eco-green-based technology (solvent free)	0.55	Alpizar-Reyes et al. (2017b)
		0.70 ± 0.12	Alpizar-Reyes et al. (2017a)
		0.59 ± 0.07	Alpizar-Reyes et al. (2018)
	Milled and grounded seeds (analysis not in dry basis)	0.07	Khounvilay and Sittikijyothin (2012)
	Strong employment of chemicals (ethanol precipitation + hexane distillation + NaOH)	3.68 ± 0.11	Chandra Mohan et al. (2018)
	Low solvent application (ethanol precipitation)	0.72 ± 0.00	Crispín-Isidro et al. (2019)
	Medium solvent application (ethanol precipitation + hexane distillation)	0.94 ± 0.01	
	High solvent application (ethanol precipitation + hexane distillation + NaOH)	0.38 ± 0.07	

Summary of the results supported by the references cited.

agreement to (Crispín-Isidro et al., 2019), the purity of tamarind gum polysaccharide is increased when the carbohydrate content is bigger. In this regard, the amount and type of solvents used for the extraction and purification of tamarind gum have been shown to influence the amount of carbohydrate content in the polysaccharide, especially those employed for removing fats and proteins associated to the polysaccharide backbone in tamarind gum. According to the monosaccharide content, tamarind gum exhibits a ratio of glucose, xylose, and galactose of 2.61:1.43:1.00, allowing it to hold water molecules in relation with monosaccharide composition and structure of gums. The variation in the availability of hydrophilic sites that hold water molecules in the polysaccharide chain may be the cause of their different moisture contents (Hamdani et al., 2019).

Researchers have reported a wide range of protein content of tamarind gum with different extraction techniques between 0.58 and 16.53% (Table 14.2). Hydrophobic proteins in tamarind gum are often responsible for the emulsifying ability main factor that allows tamarind gum to act as a wall material in the microencapsulation of drugs and natural products. Hydrophobic proteins adsorb onto the surface of oil droplets while the hydrophilic carbohydrate moiety inhibits the flocculation and coalescence of molecules through electrostatic and stearic repulsions (Dickinson, 1994).

Solvent-free technologies reported (Table 14.2) higher protein content of 12.96%–14.78% (Alpizar-Reyes et al., 2017a, b, 2018, 2020) and 13.51% (Khounvilay and Sittikijyothin, 2012) for tamarind gum extracted with hot water without any further solvent. Solvent application lowered the protein content, to 12.11% for low solvent usage and 0.58%–9.00% for the strong employment of chemicals (Chandra Mohan et al., 2018; Crispín-Isidro et al., 2019). The poor protein content exhibited is attributed to the extent purification process to which it was subjected (defatted + soluble protein alkaline extraction). It is known that gums precipitation with ethanol can induce the coprecipitation of other materials, such as protein, organic acids, certain salts, and other similar substances (Naod and Tsige, 2012).

Significant differences have been reported in the fat content in tamarind gum, from traces up to 7.20% (Table 14.2), which may be due to the differences in geographical origin, variety, and growing conditions, and in the extraction and purification processes. Fat content of tamarind gum may include saturated as well as unsaturated fatty acids. The fat content present in free solvent (nondefatted) technologies for tamarind gum has been reported as 4.76% (Alpizar-Reyes et al., 2017a) and 5.76% (Khounvilay and Sittikijyothin, 2012). The application of defatting technologies to tamarind gum has significantly reduced the content of fats to values near to traces (0.08%) (Chandra Mohan et al., 2018; Crispín-Isidro et al., 2019).

Ash content of gums has been found varying between 0.07% and 0.94% (Table 14.2). The presence of minerals like calcium, magnesium, manganese, zinc, and lead have been reported in plant gums, so it can be said that plant gums contain various essential nutrients (Fathi et al., 2016; Hamdani et al., 2019; Rezaei et al., 2016).

14.2.2 Infrared (FTIR) evaluation

One of the most successful approaches for studying and understanding the functional groups of various gums, such as tamarind gum, is to employ Fourier Transform Infrared (FTIR) spectroscopy. When new processes to extract tamarind gum are involved, the role of FTIR techniques to analyze the composition of these developments is useful to monitor the specific functional groups. In general, the use of FTIR analysis in the field of gums application is crucial to support the justification of the changes in their properties and performance in various drugs applications.

FTIR spectrums for tamarind gum commonly show two notable regions corresponding to wavenumbers of 3600–2700 cm^{-1} (mainly corresponding to lipids section) and 1800–800 cm^{-1} (consistent to proteins and carbohydrates wavenumber). Independently of the purification process involved on tamarind gum extraction, FTIR spectrums shows a wide band centered at 3310 cm^{-1} attributed to O—H stretching vibration and a sharp band at 2919 cm^{-1} due to aliphatic C—H stretching vibration of the lipid fraction (Alpizar-Reyes et al., 2017a; Chandra Mohan et al., 2018; Crispín-Isidro et al., 2019). Thus, it is evident that the processing conditions are not able to remove the small fat fractions present on tamarind gum.

For the second region, the band for —C═O stretching characteristic amide I band from proteins was observed at 1610 cm^{-1} of the acetylated units (—$CONH_2$ groups); and at 1525 cm^{-1} a strong band is associated to —$NH_3{}^+$ groups for the amide III region (Rezaei et al., 2016) and band at 1280 cm^{-1} is attributed the stretching of the C—O bond (Alpizar-Reyes et al., 2017a). Peaks at 1370, 1150, 1037, 1071, 944, and 987 cm^{-1} are characteristics of xyloglucan (Alpizar-Reyes et al., 2017a), which is an important component of tamarind gum. A band at 1370 cm^{-1} denotes the CH_2 bending of xyloglucan, the soft peak at 1150 cm^{-1} corresponds to O—C—O asymmetric stretching, while peaks at 1037 and 1071 cm^{-1} are the result of C—O and C—C stretching of the xyloglucan ring. A band at 944 cm^{-1} corresponds to the ring vibration of xyloglucan and finally, an 897 cm^{-1} band is due to C—H stretching characteristic of glucose and xylose β-anomeric links (Munir et al., 2016).

For protein structures pertaining to the exploration of eco-green-based technology (solvent free), low solvent application (ethanol precipitation), medium solvent application (ethanol precipitation + hexane distillation), and high solvent application (ethanol precipitation + hexane distillation + NaOH), Gaussian peaks were assigned to the corresponding structure based on their center (Crispín-Isidro et al., 2019). Changes in the secondary structures of the proteins for medium and high solvent application indicated that they were denatured probably due to the thermal and alkaline conditions used to purify tamarind gum. Crispín-Isidro et al. (2019) studied different extraction techniques to purify tamarind gum to a higher degree, but they concluded that the chemical structure of tamarind gum was not altered by the purification methods involved.

When strong chemicals were employed (ethanol precipitation + hexane distillation + NaOH), meaning the employment of multistage alkali wash of tamarind gum, the protein molecules in tamarind seed gum were completely removed (Chandra Mohan et al., 2018) (confirmed by absence of characteristic bands corresponding to primary and secondary amine). Finally, it is possible to establish that eco-green extraction techniques seem to fit better to the actual market requirements.

14.2.3 NMR spectrum

NMR spectroscopy has been used to determine molecular identity in polysaccharide materials, with a particular focus on the monosaccharide ratio that compounds these biopolymers. The ratios of the main monosaccharides were studied by Rodrigues et al. (2018) at 600 MHz spectra by the ^{1}H and ^{13}C NMR and xylose, glucose, and galactose were reported as the main carbohydrate residues of the tamarind gum. The authors reported that an area under glucose (at δ 3.44), xylose (sum of the signals at δ 4.95 and δ 5.56), and galactose (difference between the signal at δ 4.56 and at δ 3.44) resulted in a ratio of glucose: xylose: galactose of 3:2.58:1.63. The ^{13}C NMR was also used to evaluate the glucose: xylose: galactose ratio, which resulted in a glucose: xylose: galactose of 3:2.77:1.46. These studies are in concordance with the results generally reported to be in a molar ratio of glucose:xylose:galactose xyloglucan as 3:2:1 (Gidley et al., 1991; Hamdani et al., 2019; Rodrigues et al., 2018).

14.2.4 Thermal stability

The thermal stability of tamarind gum was investigated for different extraction methods by thermo gravimetric differential scanning calorimetry (TGA-DSC) analysis. Thermo gravimetric analysis (TGA) and differential scanning calorimetry (DSC) curves of tamarind seed remain the same for all tamarind gums despite the extraction technologies applied. In general, the TGA plot for tamarind gum shows two mass loss events. The first mass change from 1.0% to 5.5% was observable at temperatures higher than 75 °C, which can be attributed to the loss of moisture correspondent to free water in the gum particles (Alpizar-Reyes et al., 2017a; Bergstřm et al., 2012; Chandra Mohan et al., 2018; Crispín-Isidro et al., 2019). This transition has been associated to the hydrophilic nature of the functional groups of each polysaccharide. Major weight loss from 50% to 65% for tamarind gum occurred between 175 and 480 °C, which was confirmed by TGA with peak decomposition temperature at 322 °C, and was commonly attributed to the polysaccharide thermal decomposition (Chandra Mohan et al., 2018). TGA results of tamarind gum proved its thermal stability until 170 °C, thus it is useful for any process for drug delivery under this decomposition temperature.

On the other hand, the DSC technique is commonly used for studying thermal transitions that occurred during heating in the presence of an inert atmosphere. Alpizar-Reyes et al. (2017a) reported DSC plots that showed two main regions; the first region was positioned from 65 to 175 °C and, with a peak of 98 °C, indicating the evaporation of free water desorbed from the polysaccharide matrix, thus all the gums were thermally stable independently of the purification process to which they were subjected. The second peak, located at 310 °C, related to an exothermic event due to the polysaccharide decomposition.

14.2.5 X-ray diffraction (XRD) studies

XRD is a technique used to identify whether the nature of the materials is crystalline or amorphous. It will define the quantification of gum materials; therefore, XRD is a versatile tool to examine the nature of tamarind gum. The diffraction curve of tamarind gum belongs to that typically exhibited by an amorphous material indicated by the absence of sharp peaks, with an amorphous halo with a broad band centered at $2\theta = 20$ degree (Alpizar-Reyes et al., 2017a; Kaur et al., 2012a, b; Madgulkar et al., 2016; Premalatha et al., 2017). As a result, it is easy to establish that the extraction technique does not change the general conformation and structure of the gum, and only exerts influence on protein and fat content.

14.3 Functional properties of tamarind gum

Some of the functional properties of tamarind gum, such as solubility, water-holding capacity, oil-holding capacity, emulsion ability, emulsion stability, surface tension, and electrophoretic mobility and rheology, at specific conditions, will be described in the following sections. The study of the functional properties of tamarind gum is important to evaluate the gum's potential technology applications as a drug carrier and control release system, to design and to operate processing equipment, and to control its storage (Kumar and Bhattacharya, 2008).

14.3.1 Water solubility, water-holding capacity, swelling ability, and oil-holding capacity

Tamarind gum is water soluble, and the steric hindrance of its side chain inhibits the aggregation to cellulose-like chains. It forms a homogeneous solution on heating with water while stirring. The chemical modification of this gum, such as carboxymethylation, tends to increase its solubility in cold water (Goyal et al., 2007; Mali et al., 2019). In another study, Alpizar-Reyes et al. (2017a) evaluated the water solubility of tamarind seed gum depending on the temperature, and they found that water solubility of this gum increased from 8% to 21.8% on increasing the temperature from 25 to 65 °C.

Water-holding capacity (WHC) represents the amount of water held and absorbed by the hydrated sample after an external force is applied, and provides information about the stability, yield, and sensory characteristics of a gum. Alpizar-Reyes et al. (2017a) evaluated the WHC of tamarind gum as a function of temperature and found that WHC increased as the temperature increased, going from 0.18 g/g at 25 °C to 1.07 g/g at 65 °C.

Furthermore, tamarind gum swelling studies using water found that tamarind gum swelled 1.6 times its weight, while carboxymethylated tamarind gum (CTG) swelled 2 times with respect to the *v*olume of the dry gum (Mali et al., 2019). In addition, the enhancement of substitution degree in the carboxymethylation of gum had a favorable effect on swelling of tamarind gum (Goyal et al., 2007). The rapid swelling ability of tamarind gum suggests its use in hydrophilic matrix tablets and its application in controlled drug delivery (Phani Kumar et al., 2011).

The oil-holding capacity (OHC) of a gum is the absorption of oil through the lateral nonpolar sites within protein molecules. A gum with high OHC may be able to retain oil-based flavoring and enhance mouthfeel in food applications; likewise, it may retain oil-based drugs. Alpizar-Reyes et al. (2017a) evaluated the OHC of tamarind gum as a function of temperature and found that OHC increased as the temperature increased, ranging from 0.068 g/g at 25 °C to 0.133 g/g at 65 °C.

14.3.2 Emulsifying ability and emulsifying stability

Emulsifying ability (EA) measures the ability of an emulsifying agent to form emulsions while the emulsion stability (ES) to heating measures the breakdown of the emulsion when it is heated, where the proteins adsorbed to the surface of the oil droplets unfold and expose nonpolar amino acid, which leads to hydrophobic attraction between droplets and flocculation occurs.

EA measured considering the fraction in weight of tamarind gum and volume of oil of 0.2:1 tamarind gum:oil ratio, the EA obtained was 78.3%, while using 1:1 tamarind gum/oil ratio the EA was 90% (Alpizar-Reyes et al., 2017b). This behavior was attributed to the increment of the gum, and thus to the increment of the branched structure among the surface active to absorb oil molecules that reduced surface tension. Furthermore, the authors evaluated the ES based on mass of tamarind gum/volume of oil ratio and found that decreasing the gum fraction with respect to the oil increased the ES—that is, using 1:1 tamarind gum/oil ratio, the ES was 82.2%, while using 0.2:1 tamarind gum/oil ratio, the ES was 91.1%.

Moreover, the EA of two different formulations of oil/water emulsions to prepare microcapsules was evaluated (Alpizar-Reyes et al., 2020). Emulsion 1 was composed of tamarind gum/sesame oil ratio 1:1 in dry basis, 10% (w/w) of total solid content, and dispersed phase volume fraction $\varphi_{O/W} = 0.05$. Emulsion 2 was composed of tamarind gum/sesame oil ratio 1:2 in dry basis, 15% (w/w) of total solid content, and $\varphi_{O/W} = 0.1$. EA was greater for emulsion 2 (90.28%) than emulsion 1 (86.26%); conversely, ES was greater for emulsion 1 (82.31%) than emulsion 2 (80.33%). Both emulsions showed good stability to heating; however, the reduction on the surface tension for tamarind seed gum used for stabilizing emulsion 2 was attributed to the reduction in its long-term stability.

Tamarind gum has demonstrated good emulsifying properties and was applied in various emulsions studies; for example, Bhattacharya et al. (1993), Kumar and Bhattacharya (2008), and Tsuda et al. (1994) described the formation of stable castor oil-in-water emulsions using tamarind seed gum at 2% *w*/*v* as stabilizer agent, exhibiting droplet sizes from 1 to 10 μm.

On the other hand, the fabrication of oil-in-water emulsions stabilized with tamarind gum aqueous dispersions (2%, w/w) at different levels of purification was reported (Crispín-Isidro et al., 2019). For emulsions with low purity in tamarind gum, the initial area-volume mean diameter ($d_{3,2}$) was 1.35 ± 0.15 μm; meanwhile, emulsions where medium purity tamarind gum was used displayed droplet sizes of 4.82 ± 0.10 μm, and for emulsions of high purity, tamarind gum of 9.45 ± 0.10 μm. Thus, low-purity gum achieved the smallest droplet size and the highest emulsion stability. Then, the purification of tamarind gum had a significant effect not only on the final chemical composition but also on the emulsifying properties of this polysaccharide.

14.3.3 Surface and interfacial tension

Tamarind gum has surfactant properties since this produces changes in the surface tension of the water. Phani Kumar et al. (2011) determined the surface tension of tamarind gum (0.1%, w/v) by the drop count method, using a stalagmometer, and the obtained result was 83.26 dynes/cm.

The use of tamarind gum with three different levels of purification (low, medium, and high purity), in agreement to its carbohydrate content, was studied for stabilizing canola oil/water emulsions, and the time evolution of the dynamic interfacial tension (σ) was also reported (Crispín-Isidro et al., 2019). All tamarind gum aqueous dispersions exhibited the same trend with an initial sharp decrease of σ during the first 200 s, followed by a slow progressive drop in σ at longer times, until reaching asymptotic values, where σ did not change by more than

0.5 mN/m in 30 min. The main results showed that tamarind gum with the lowest purity decreased the interfacial tension faster than gums with higher purity, due to its relatively high content of surface-active compounds, such as protein and polyphenols, which may contribute to its adsorption at the interface.

14.3.4 Electrophoretic mobility (ζ-potential)

ζ-potential values are related to the stability of the colloidal systems. If the particles in a suspension have a large negative or positive ζ-potential, they tend to repel each other and no tendency for the aggregation of particles can be observed. In contrast, if the particles have a low ζ-potential value, aggregation and flocculation occur (Crispín-Isidro et al., 2019). González-Martínez et al. (2017) reported a ζ-potential value of −12 mV at pH 10.0 in a solution of crude tamarind seed gum.

In addition, tamarind gum at different purity levels (low, medium, and high purity), shows negative ζ-potential values ranging from −11.00 ± 0.19 mV for low-purity tamarind gum to −5.72 ± 0.20 mV for the highest-purity tamarind gum (Crispín-Isidro et al., 2019). The decrease in the ζ-potential values as the purity in the tamarind gum increased was related to the elimination of charged impurities throughout the purification, that contribute to the electrophoretic mobility in the aqueous dispersion.

14.3.5 Rheology

There is a great variety of studies that evaluate the rheological behavior of tamarind gum. Kumar and Bhattacharya (2008) evaluated the rheology of tamarind kernel powder at different concentrations (2%, 4%, 6%, 8%, and 10%, w/w) and these behave like non-Newtonian, shear-thinning fluids with low yield stress values at low concentration and vice versa (Kumar and Bhattacharya, 2008). The Herschel–Burkley model adequately fits the shear stress–shear rate data.

In another study, tamarind seed gum solutions at 20 °C, in a range of concentrations from 0.67% to 5.70% exhibited shear-thinning flow behavior at high shear rate and Newtonian region at low shear rate. At higher concentrations, pronounced shear-thinning was shown and at lower concentrations, the viscosity did not show dependence on shear rate. Tamarind seed gum showed a typical random-coil polymer behavior. Dilute and semidilute regions were observed with slopes of 2.2 and 4.3, respectively. When the specific viscosity at zero shear rate (η_{sp0}) was plotted against the coil overlap parameter (C[η]), it was found the critical concentration $C^*[\eta] = 4.23$, that is, about 0.90% (w/w), which agrees with the results of the majority of coiled hydrocolloids. In addition, a viscoelastic study was carried out and the Cox-Merz rule was applied and well-adjusted at 2.30 and 2.75% (w/w) of tamarind gum (Khounvilay and Sittikijyothin, 2012).

Tamarind gum dispersions reported by Alpizar-Reyes et al. (2018) had non-Newtonian shear-thinning behavior described by the Power law model. In this study, the effects of tamarind gum concentration, temperature, pH, and salt addition were evaluated on apparent viscosity. It was determined that as tamarind gum concentration increased (from 0.5% to 2.0%, w/w), the viscosity and pseudoplasticity of gum dispersions increased; as temperature increased (from 25 to 60 °C), the viscosity and pseudoplasticity of gum dispersions decreased;

as pH increased (from 4 to 10), the apparent viscosity of the gum dispersions increased; and finally, it was observed that the addition of salts (NaCl, KCl, and $CaCl_2$) modified the apparent viscosity differently.The rheological behavior of the tamarind seed gum showed that this fluid is resilient against pH, temperature, salt, and sugar concentrations, therefore, it can be applied as a food additive (Alpizar-Reyes et al., 2018).

A similar rheology study was carried out by Shao et al. (2019), who found that tamarind seed gum aqueous solutions from 0.5% to 4% (w/v) exhibited non-Newtonian shear-thinning behavior described by the Williamson model. Apparent viscosities of tamarind seed gum solutions decreased drastically in an alkaline solution of $pH > 10$; however, these were only slightly affected by $pH < 10$. The apparent viscosity of the tamarind seed gum solution at 2% (w/v) decreased slightly with increasing temperature (5–85 °C) at three different shear rates. On the other hand, dynamic oscillatory analysis of tamarind seed gum was evaluated at concentrations from 2% to 10% (w/v). For 2% (w/v) of tamarind seed gum solution, the storage modulus was lower than the loss modulus ($G' < G''$) throughout the frequency range, indicating a viscous-like behavior. As the concentration increased 4%, 8%, and 10% (w/v), the G' and G'' increased, and a crossover occurred at a frequency of 80, 25, and 7 rad/s, respectively. Thus, by increasing the concentration of tamarind seed gum from 2% to 10% (w/v), there was a change in the behavior of the gel from viscous to elastic, and at 10% a weak-gel was formed (Shao et al., 2019).

Crispín-Isidro et al. (2019) studied the flow behavior of tamarind gum solutions with different purity levels at 1.5% and 2.0% (w/w). The apparent viscosity was described by a plateau region (Newtonian behavior) at low shear rate values and a shear-thinning behavior (non-Newtonian) at higher shear rates, and these curves had a better fit to the Ellis model. The low shear viscosity values of the tamarind gum solutions tended to be higher when the concentration and the purification of the gum were higher (Crispín-Isidro et al., 2019).

The rheology of chemically modified tamarind gum has also been evaluated. Carboxymethylated tamarind gum (CMTG) increased the viscosity of the gum since CMTG disrupts the organization and exposes the polysaccharide network to hydration, which results in higher viscosity (Goyal et al., 2007). CMTG can be used as a matrix former and release retardant in the development of novel drug delivery systems (Mali et al., 2019).

In addition, tamarind seed gum has been thiolated by esterification of its hydroxyl groups with thioglycolic acid in order to improve its mucoadhesivity and cohesive properties. Kaur et al. (2012a, b) compared carbopol-based metronidazole gels using thiolated and nonthiolated tamarind seed gum gels, and found higher mucoadhesion to chicken ileum when the thiolated tamarind gum gel was used. The gels containing thiolated tamarind seed gum had the lowest hardness and adhesiveness but the highest cohesiveness.

Tamarind xyloglucans are neutral polysaccharides of low viscosity and low molecular weight compared to other polysaccharides. Tamarind xyloglucans have been used as a gelling agent since these can function as pectin; therefore, they represent an alternative of gelling raw material for the pharmaceutical and food industries (Mishra and Malhotra, 2009).

14.3.6 Functional properties of binary mixtures using tamarind gum

The functional properties of tamarind gum change when it is mixed with other compounds such as starches, gums, proteins, etc., with which it forms binary mixtures.

The addition of tamarind seed gum to starch causes an increase in its viscosity. Pongsawatmanit et al. (2006) evaluated the viscosity of 5% tapioca starch/tamarind xyloglucan mixtures, at different mixing ratios, and found that peak and final viscosities increased with increasing tamarind xyloglucan content. Furthermore, a mechanical system of 5% (w/w) tapioca starch/tamarind xyloglucan mixtures changed from gel behavior to concentrated solution and showed higher loss tangent (G''/G') with increasing tamarind xyloglucan concentration.

Mung bean starch gel increased its peak of viscosity (Liu and Xu, 2019) from 4372 to 10,285 cP adding 10% (w/w) of tamarind gum. The addition of tamarind gum at high concentration to starch generated firm and spring gels, and reduced the gel syneresis. Xie et al. (2020) mixed tamarind gum with three types of corn starches (normal, waxy, and high amylose corn starch), using different amylose-amylopectin ratios, and demonstrated that tamarind gum retards the gelatinization of starch granules mainly affecting amylopectin. Binary mixtures of tamarind gum with normal, and with waxy corn starch formed weak gels, while tamarind gum with high amylose corn starch increased their elastic properties.

On the other hand, tamarind gum has been mixed with other gums. Zhang et al. (2008) mixed tamarind gum (2%, w/w) and sodium alginate (0.6%, w/w), and formed beads using calcium chloride; they evaluated the swelling behavior of this mixture varying pH ($3-11$) and found that in the pH range 3–7 there was no destruction of hydrogel; however, increasing the pH above 7 produced the disintegration of alginate chains and an increase in the swelling degree. In later studies, Nayak and Pal (2011) applied tamarind gum–alginate composite beads for controlled delivery of diclofenac sodium for prolonged period with good results, and they found that swelling and degradation of the beads were affected by the change in pH.

In the optimization of spray-drying tamarind gum, soya protein isolate was added, which produced an increase in yield, a reduction of the gum hygroscopicity, and an increase in the solubility of the powder, since the protein is an efficient carrier agent (Muzaffar and Kumar, 2015). Also using proteins, Jana et al. (2016) used gelatin/carboxymethylated tamarind gum (CMTG) mixtures to control the delivery of aceclofenac and they found that gelatin alone had difficulty to retain a significant amount of the drug, while mixing gelatin with CMTG improved the drug entrapment efficiency above 90%.

Tamarind gum has also been mixed with other substances to modify its functional properties in certain applications; for example, Yadav et al. (2017) studied composite films using polyvinyl alcohol (PVA) and carboxymethyl tamarind gum, and found an increase in firmness of PVA films when CMTG was added, and this was dependent on the gum concentration. Increasing CMTG content in the film also increased the elastic component due to the improvement in the reinforcement effect. The mechanical characteristics of the films improved on adding CMTG, due to an increase of intermolecular hydrogen bonding, an enhanced crystallinity of polymers, and low molecular rearrangement under stress. The mechanical stability of films is important during handling and storage.

Likewise, Sharma et al. (2014) prepared ion gels using tamarind gum and synthetic and bio-based ionic liquids by a heating/cooling process and these gels exhibited thixotropic behavior, which involved the recovery of gel structures after 10 consecutive cycles. The ion gels had superior quality in viscosity, viscoelasticity, and thixotropic behavior compared with the hydrated tamarind gum (Rao and Mathew, 2012; Sharma and Bhardwaj, 1997).

14.4 Tamarind gum-based colloidal systems in food and pharmaceutical applications

As previously mentioned, tamarind gum is extracted from the kernel of tamarind seed, which is a residue from the tamarind pulp industry, mainly composed of a galactoxyloglucan polysaccharide. Tamarind gum is considered as a promising biopolymer due to its applications as a stabilizer, thickener, viscosity enhancer, emulsifier, gelling, drug carrier, release retardant, and binder agent in the food and pharmaceutical industries. When native tamarind gum is dispersed in aqueous media, it displays excellent ability to swell in the aqueous medium and form a mucilaginous solution, which exhibits rheological properties like a pseudoplastic fluid, besides possessing a hydrophilic character that allows it to display gel-forming, biomucoadhesive characteristics, and a stiffer conformation with the large volume of occupancy, making it suitable as a potential excipient in the preparation of hydrogels, oleogels, emulsions, and controlled released systems. Other distinguishing properties of tamarind gum are related to its high swelling index and high thermal stability, making it a suitable excipient for drug delivery systems.

In the food industry, tamarind gum applications include being used as a thickener agent in sauces, fruit pulp beverages, low-fat milk, and cocoa because of its smooth flow and lack of sticky texture, as gelling and water retaining in jellies and pudding by providing an elastic gel behavior to concentrated sugar solutions, and as a suppressor of the aging of starch by conferring heat stability and mechanical strength, improving the texture of starch in bakery products, custard cream, flour paste, stew, and noodles (Ferrero, 2017; Yamatoya et al., 2020). As tamarind gum exhibits good emulsion stability in acidic conditions, Kim et al. (2006) reported that tamarind gum provides benefits in dressing applications by adjusting the texture of dressings in combination with xanthan gum due to its thermoreversible physical gel-like properties. Tamarind gum was also used in frozen desserts, displaying an overrun and suppressing the ice crystal growth and sugar crystallization after storage, associated to its water-retention capabilities, indicating that tamarind gum holds more free water in freezing mixes around the ice crystals and prevents their growth; in addition, tamarind gum-sugar gel becomes harder and more elastic after freeze-thaw, suggesting that freeze-thaw processes make the gel-like network stronger (Yamatoya et al., 2020; Yamatoya and Shirakawa, 2003).

Therefore, in this section, the relationship between physicochemical and functional properties for colloidal applications of tamarind gum and its functionalized derivatives will be described.

14.4.1 Tamarind gum hydrogels

Hydrogels are defined as three-dimensional (3D) cross-linked polymer networks with high capability to uptake a large amount of water and even biological fluids resembling biological tissues (Ahmad et al., 2019; Ahmed, 2015). These 3D networks are connected to each other through cross-linking polymers (either physically or chemically) to render the network insoluble and immersed in an aqueous solution (De et al., 2002; Maharana et al., 2017). The spaces available within the formed 3D network allow the immobilization of aqueous or organic solvents. The cross-linking reaction can be achieved by using cross-linker compounds, chemical modification, grafting, or high energy radiation (gamma or UV rays), being more stable systems than those carried out by chemical processes (Ali and Ahmed, 2018). In addition, the biopolymer network may result in hydrophilic or hydrophobic materials, where hydrophilic components are related to the induction of swelling in the hydrogel structure, whereas hydrophobic components tend to control the swelling rate of the gel as well as the mechanical properties (Zhang et al., 2008).

Tamarind gum functionalization

Tamarind gum does not form gel-like structures when it is placed alone in an aqueous solution; instead, it dissolves in water, yielding high-viscous dispersions, where gelation is hindered by steric hindrance of (1→2)-β-galacto-xylose branches, remaining preferably in sol state. However, when tamarind gum is in the presence of sugars like sucrose, ethanol, polyphenolic compounds, and iodine, their interactions induce the gelled-phase formation (Yamatoya et al., 2020; Yuguchi et al., 2001). Gelation of tamarind gum solution mixed with sugar or alcohol is considered to involve cross-linking of tamarind gum molecule aggregation domains due to a dehydrating action by these additives. It is also reported that these tamarind gum gels can become sols by heating and revert to gel again by cooling, indicating that the transition between sol and gel is thermoreversible, exhibiting high elasticity and low water release, even when single tamarind gum does not form gelled systems by heat treatment (Yamatoya et al., 2020).

Functionalization of tamarind gum by different chemical modifications has been demonstrated to improve the physicochemical properties of native gum. The first modification is tamarind gum degalactosylation, which induces the formation of gelled structures by cross-linking occurred between the degalactosylated tamarind gum molecules, forming arrangements in lateral aggregates that lead to flat plate shapes, which increase as the loss of (1→2)-β-galacto-xylose occurs, resulting in an opaque thermoreversible gelation process (Shigenobu et al., 1999).

Carboxymethylation of tamarind gum (CMTG) consists in the attachment of pendant carboxylic acid groups (–COOH) to the native tamarind gum structure via Williamson' etherification, using monochloroacetic acid and sodium hydroxide reaction at high temperature (Khalil et al., 1990). This reaction leads to a nonspecific degradation via β elimination and/or peeling reaction initiated at decreasing sugar units in the native gum structure due to the highly alkaline pH environment; this derivatization process provokes the disruption in the organization of the macromolecule, which exposes the polysaccharide network to hydration, causing the decrease in the molecular weight of the derivatized gum but increasing the viscosity of the biopolymer aqueous dispersion (Manchanda et al., 2014; Nayak and Pal, 2018; Olusola et al., 2014). This type of modification is one of the most common due to its simplicity, lower costs, and great variety of applications for the resultant CMTG. This CMTG displays higher hydrophilicity and solubility in an aqueous medium than its respective native gum, due to the presence of carboxymethyl groups in the tamarind gum structure; it displays higher viscosity due to its higher swelling capability, as well as higher resistant toward enzymatic attack and therefore lower degradability in aqueous environments, making it a good excipient material in hydrophilic drug delivery systems (Manchanda et al., 2014; Nayak and Pal, 2018).

CMTG is an anionic polysaccharide able to interact with cationic moieties to form a gel structure. Alpizar-Reyes et al. (2017a), Kaur et al. (2012b), Madgulkar et al. (2016), and Premalatha et al. (2017) demonstrated that CMTG can interact with divalent cations like Ca^{2+} and form ionically gelled nanoparticles, such as those formed with alginate and calcium chloride, where the gelled particle sizes were directly dependent on the CMTG and Ca^{2+} cross-linker concentrations. Alpizar-Reyes et al. (2017a), Kaur et al. (2012b), Madgulkar et al. (2016), and Premalatha et al. (2017) showed that CMTG is able to interact with macromolecules like chitosan by the formation of polyelectrolyte complex linkage between free -NH_3^+ of chitosan and -COO^- of CMTG, respectively. In this work, functional properties of hydrogels, like the swelling index and drug carrier, were dependent on the polymer charge and concentration, degree of ionization, cross-linking density, hydrophilicity and hydrophobicity, as well as the pH of the aqueous medium, where higher entrapment efficiencies and better swelling equilibrium were achieved at higher CMTG contents, and closely related to higher viscosity systems. Similar results were observed by Jana et al. (2016), who synthesized gelatin-CMTG hydrogel composites using glutaraldehyde as a cross-linker agent. This gelatin-CMTG hydrogel achieved higher drug entrapment efficiency than that reached by the cross-linked gelatin alone, and the release profile was dependent upon the extended cross-linking and the amount of CMTG. In a similar biopolymer system, Shaw et al. (2015) reported that CMTG exhibited high compatibility to the gelatin phase, resulting in better mechanical and mucoadhesive properties of the hydrogels, which was attributed to the presence of the free carboxylic groups in CMTG, inducing the formation of pH-sensitive swelling and drug release hydrogels, demonstrating the potential applications of this CMTG. The same trend was observed by Meenakshi and Ahuja (2015) when CMTG-polyvinyl alcohol (PVA) cryogels were synthesized using the freeze-thaw method as a physical cross-linking strategy. In this work, lower CMTG concentrations and high PVA contents led to lower release of metronidazole as a core material, achieving an increase in metronidazole release as the CMTG content increased.

Another modification applied to tamarind gum is thio-functionalization (TTG), where tamarind gum polysaccharide is modified by its esterification with thioglycolic acid and confirmed by the S–H stretch in Fourier-transformed infra-red spectra (Manchanda et al., 2014), aiming the improvement of the mucoadhesion of natural polysaccharides. In this sense, Kaur et al. (2012a, b) reported that thio-functionalization in tamarind gum is achieved by the esterification of the hydroxyl groups of galactoxylan moieties with the carboxyl groups of the thioglycolic acid, resulting in a white and water-soluble powder with a higher degree of crystallinity. TTG hydrogel showed an increase of 6.85-fold greater mucoadhesive strength and better surface roughness than tamarind gum hydrogel and commercial formulations.

On the other hand, grafting of tamarind gum (GTG) was reported by Shailaja et al. (2012); this modification overcame some disadvantages, such as uncontrolled rate of hydration, drop viscosity on storage, and susceptibility to microbial degradation, that tamarind gum exhibits. GTG is formed by bonding synthetic monomers molecules onto the polymer chain, combining the functional properties of copolymers and polymers molecules; they are biodegradable to some extent and stable to shearing due to the attachment of flexible synthetic polymers onto the more rigid polysaccharide backbone. Generally, this type of modification is carried out by conventional methods like redox, or nonconventional ones like microwave irradiation, γ-ray irradiation, or electron beams, where the resultant grafted biopolymers exhibit excellent capabilities for controlled drugs release (Ghosh et al., 2010; Rani et al., 2012). The main molecules used for grafting of tamarind gum are polyacrylamide (Sen and Pat, 2009), methyl methacrylate (Shailaja et al., 2012), ethyl acrylate (Del Real et al., 2015), and acrylonitrile (Singh et al., 2009), where the functional properties of tamarind gum and its hydrogels, such as water retention capacities and gelling tendency, were modified after grafting, producing better natural products with less side effects and minimum loss of the initial properties of the substrate used.

Sulfonation in tamarind gum was assessed by swelling the tamarind gum polysaccharide in a dimethylformamide and sulfur-trioxide-pyridine complex; alkylamination, an oxidation of galactosyl hydroxyl methyl groups to formyl groups, by using galactose oxidase catalase; and its cross-linking with epichlorohydrin, which exhibited superior wicking and swelling behavior, as well as better retarding effect in the drug release than native tamarind gum (Kumar et al., 2018; Nayak and Pal, 2018).

Applications of hydrogels from tamarind gum, native or functionalized, are focused mainly on pharmaceutical approaches such as: the production of thickened ophthalmic solutions used as a vehicle for sustained release of drugs due to the mucoadhesive properties that extend the retention time onto the surface of the eye; acyclovir drug loaded in nanoparticles with greater in vivo bioavailability than commercial products where acyclovir is used in a suspension product; and the sustained release of hydrophilic drugs like acetaminophen, caffeine, theophylline, and salicylic acid, or nonpolar compounds like indomethacin, exhibiting a zero-order release rate with the capability to control their release by modifying the diluent or the type of binders (Kumar et al., 2018).

Tamarind gum has also been functionalized by the incorporation of carbon nanotubes (CNTs). Choudhary et al. (2018) incorporated CNTs, hydroxyl functionalized CNTs (OH-CNTs), and carboxyl functionalized CNTs (COOH-CNTs) in tamarind gum hydrogels. Their results showed that CNTs induced modifications in the microstructure by altering the intra- and intermolecular interactions in the hydrogels. These changes provoked modifications in the physicochemical properties of the tamarind gum hydrogels and also in the differential drug release patterns of tigecycline as a model drug. Even when all the tamarind gum-CNTs hydrogels were easily spreadable, differences in the mechanical properties, microarchitecture, topography, and electric impedance were observed, and were dependent on the type of CNTs used.

Hybrid tamarind gum composites

Biopolymer hydrogels are materials that display technological features like environment-responsive, self-healing, self-assembled conductive, and shape memory, and are considered as supramolecular materials (Mahinroosta et al., 2018). In this sense, crude and CMTG functionalized tamarind gums have been used in combination with other macromolecules like proteins and polysaccharides in order not only to improve the controlled and sustained release of drug, but also to release it in specific target sites.

Hybrid functionalized tamarind gum composites formed by a combined process of chemical cross-linking and freeze-drying were reported by Jana et al. (2016), where gelatin and CMTG hybrid composites were formed and tested for controlled delivery of aceclofenac, a nonsteroidal antiinflammatory drug widely used for symptomatic relief of rheumatoid arthritis, osteoarthritis, and ankylosing spondylitis. It was found that the structural arrangement obtained from the composites led to a suppression in the drug core release when the biopolymer particles were subjected to in vitro acidic media, extending the release at neutral pH values in phosphate buffer solution (pH 6.8). This behavior was attributed to the performance of polymer chain relaxation/swelling and diffusion mass transfer mechanisms, as well as the functional properties of CMTG for modulating the release of drugs like aceclofenac at specific triggering agents and aiding to the reduction of gastrointestinal side effects, costs, and patient suffering associated with the frequent dosing of this drug (Jana et al., 2016).

Synthesis of polyvinyl alcohol (PVA) and CMTG hybrid composites was reported (Yadav et al., 2017); these composites were applied for active films loading ciprofloxacin hydrochloride as a drug model. It is noteworthy that despite PVA being a water-soluble biopolymer used in drug delivery systems and wound dressing applications due to its good physicochemical properties, this polymer is lacking in terms of cell-specific bioactivities. Therefore, the addition of CMTG to PVA promoted the formation of films with better mechanical, thermal, and biological properties than the forming biopolymers, where the improvement in these characteristics was dependent on the concentration of CMTG and was associated with the presence of intermolecular hydrogen bonding, with low molecular rearrangement under stress conditions, and enhanced crystallinity. The evaluation of antibacterial activity showed that films based on PVA-CMTG composites loaded with ciprofloxacin hydrochloride did not exhibit significant differences when CMTG content varied in the formulation. However, the evaluation of human epidermal keratinocyte cell (HaCaT) proliferation, as an accepted cellular candidate for probing epidermal biology in vitro, showed that low contents of CMTG in the composite film supported to a greater extent the cell proliferation than films where higher CMTG contents were tested. The explanation for this behavior was related to the use of CMTG as a "chemical cue" where the cell proliferation was dependent on its concentration, joined to the combined effect of surface properties of the biopolymer film and the ligand distribution, showing the good cytocompatibility of the PVA-CMTG film.

Tamarind gum grafting copolymerization

Graft copolymerization in natural polysaccharides is an important resource for developing advanced materials with improved functional properties, allowing them to be used in agricultural materials and their by-products to be used as substitutes for unsustainable synthetic polymeric materials.

Graft copolymers are defined as a long sequence of one polymer (backbone polymer) with one or more branches (grafts) of another (chemically different) polymer. The process of graft copolymer synthesis in natural biopolymers starts with a preformed polymer, as the polysaccharide, then an external agent is used to create free radical sites on the polymer backbone; after this, the monomer is added up through the chain propagation step, leading to the formation of grafted chains. The various methods of graft copolymer synthesis mainly differ in the types of generation of the free radical sites on this preformed polymer (Ghosh et al., 2010; Rani et al., 2012).

An example of this type of tamarind gum (TG) grafting is reported by Singh et al. (2009), where acrylonitrile was grafted on to TG polysaccharide using persulfate/ascorbic acid redox initiator to synthesize the poly(acrylonitrile)-grafted-TG (PAN-g-TG). The main results indicated that grafted materials displayed different water/saline retention, gel forming ability, and enhanced shelf life of the grafted gum solutions with great potential for its commercial utilization in pharmaceutical and industrial approaches.

Polyacrylamide-grafted-tamarind gum (PAM-g-TG) synthesized by conventional redox grafting, microwave-initiated grafting, and microwave-assisted grafting showed that grafted copolymers where a maximum percentage of grafting was achieved exhibited enhanced intrinsic properties, associated to the longer chains of PAM grafted onto the backbone of TG, leading to the increase in the hydrodynamic volume of the biopolymer molecule in a solution and therefore an increase in the intrinsic viscosity and molecular weight (Ghosh et al., 2010).

Ghosh et al. (2010) reported the functional properties of PAM-g-TG in terms of its flocculation characteristics in a kaolin suspension system, where this performance was improved when graft TG copolymers were obtained.

Nandi et al. (2019) synthesized a similar PAM-g-TG system, by a free radical method assisted with microwave with ceric (IV) ammonium nitrate (CAN) as a free radical initiator, where the flocculating potential in a peroral paracetamol suspension was evaluated. It is noteworthy that pharmaceutical dosage forms include flocculated suspensions, which are characterized by the formation of loosely packed cake which can easily be redispersed by gentle shaking. The use of natural hydrophilic polysaccharides, like TG, has demonstrated their capability for acting as suspending agents, joined to their protective and coating roles, which may prevent or induce the cake formation. In this study, the PAM-g-TG exhibited improved thermal stability not only for the biopolymer material, but also for the loaded drug, and showed that flocculation efficiency in paracetamol suspension was enhanced in the grafting as the TG content increased. This behavior was associated to the PAM functionalization by the TG grafting and the incorporation of numerous side branches in the main polymeric backbone, resulting in a comb-like structure (Nandi et al., 2019).

Similar results were reported by Sen and Pat (2009), where PAM-g-CMTG was used as a flocculant agent in a kaolin suspension with better functional properties than those found when CMTG was used for the same purpose. Furthermore, Pal et al. (2012) stated that PAM grafted polysaccharides usually find applications as efficient flocculants agents in environmental or pharmaceutical approaches when are used at low doses, as well as their controlled biodegradable, shear-resistant, inexpensive, and ecofriendly characteristics. Nonetheless, these materials display some drawbacks and limitations such as their low surface area, small hydrodynamic radius, and complicated diffusion processes. Pal et al. (2012) overcame these drawbacks by developing a high-performance nanocomposite based on silica nanoparticle-incorporated PAM-g-CMTG. The functional properties that were evaluated indicated that these nanocomposites exhibited an enhanced adsorption of methylene blue dye and better properties as a flocculant agent in comparison with PAM-g-CMTG. This was attributed to the enhancement of hydrodynamic volume and hydrodynamic radius and their direct correlation to this type of functional property, where surface-active properties determine the efficacy of the materials.

Del Real et al. (2015) reported the graft copolymerization of ethyl acrylate (EA) onto TG (EA-g-TG). Their results showed that free radical polymerization mechanism allowed the grating reaction, which was confirmed by FTIR and NMR ^{1}H spectroscopies. Techno-functional properties of EA-g-TG showed higher thermal stability when compared with TG polysaccharide. Fresh grafted copolymer was only soluble in water, and became insoluble in water and organic solvents after drying. Mechanical properties were increased for tensile strain and also exhibited high biodegradability under anerosion conditions in the presence of the bacterium strain *Alicyliphilus* sp. *BQ1.*, making this copolymer adequate for usage in disposable products (Del Real et al., 2015).

A tamarind gum-chitosan (TG-chitosan) copolymer was proposed for use in producing magnetic microspheres. According to Zhang et al. (2007), the composite magnetic microspheres were synthesized utilizing the suspension cross-linking technique in the Fe_3O_4 magnetic carrier technology. The composite magnetic microspheres displayed particles sizes ranging from 230 to 460 μm, with sufficient magnetic field intensity to excite all the dipole moments of magnetic carrier. This TG-chitosan copolymer represents a promising magnetic support to be employed in magnetic carrier technology with good magnetic quality, and as expected, swelling properties changed as aresponse to the pH of the swelling medium, this porperty is useful in potential modulation systems in biomedical fields.

Moreover, synthesis of interpenetrating networks (IPNs), defined as a polymeric network of two or more polymers that form a rigid composite network structure by cross-linking of at least one polymer in the presence of another, has demonstrated improvements in the mechanical strength, loading capacity, and sustained and controlled drug release, and provides space for drug encapsulation in a three-dimensional structure by combination of individual properties of polymers. In this sense, Mali et al. (2017a, b) reported the synthesis of pH-dependent site-specific IPNs of aceclofenac using CMTG and chitosan. In this study, chitosan was used as a base polymer, cross-linked with glutaraldehyde to form a network, and CMTG was the second polymer entangled in cross-linked chains of chitosan. The intercalation of CMTG in the chitosan backbone was carried out due to formation of a polyelectrolyte complex between free $–NH_3^+$ of chitosan and $–COO^-$ of CMTG. The CMTG-chitosan IPNs exhibited enhanced aceclofenac entrapment efficiency as the cross-linker content increased, whereas CMTG concentration displayed a significant effect due to the formation of a thick surface that diminished the loss of the core material by means of the high viscosity of CMTG. Drug delivery in the IPNs showed their pH dependence on swelling properties, suggesting that these composites are suitable for oral site-specific delivery of drugs in order to avoid exposure of drugs to an erratic gastric environment and drug release in the intestine.

14.4.2 Tamarind gum in polymer complexation

The uses of tamarind gum have been explored as a microsphere drug carrier, as other natural polymers; this polysaccharide is cheap, biodegradable, and safe for pharmaceutical formulations. Microsphere systems of naturally derived polymers have been prepared by several techniques, including coacervation phase separation (Farooq et al., 2014).

In aqueous solutions, a great diversity of interactions occurs when proteins and polysaccharides interact. These interactions are defined by and depend strongly on environment conditions, such as pH, temperature, and hydrocolloids ratio (Espinosa-Andrews et al., 2013). Depending upon the composition of the formulations, drastic changes in the structural properties of the protein-polysaccharide may occur. When the aqueous solution of the proteins and the polysaccharides are mixed together, there is a possibility of formation of a liquid water in-water emulsion, complex coacervates (where both the polymers appear in a single concentrated phase), and soluble complexes due to the formation of self-organized structures, where the formation of any arrangement is governed by the thermodynamic compatibility among the proteins and the polysaccharides (Shaw et al., 2017). In this sense, complex coacervates can be classified as soluble or insoluble according to their electrostatic repulsive or attractive forces (González-Martínez et al., 2017; Kaushik et al., 2015).

Complex coacervation between tamarind gum and whey protein isolate (WPI) was first reported by González-Martínez et al. (2017), where the formation of the complex coacervation was attained at a pH value where both hydrocolloids reached their electrical equivalence and an insoluble complex was obtained that was completely neutral. This study states that a maximally electrostatic interaction between the TG polysaccharide and the WPI protein was achieved at a mass ratio of 0.3:1.0 respectively, at pH 3.68. The resultant coacervate phase that was separated was spray-dried and characterized, exhibiting higher crystallinity than the TG polysaccharide. In addition, the complex coacervate exhibited better thermal stability against denaturation of the biopolymers.

Shaw et al. (2015, 2017) reported the use of gelatin-tamarind gum (gelatin-TG) and gelatin-carboxymethyl tamarind gum (gelatin-CMTG) phase-separated hydrogels, formed due to inter- and intra-polymeric interactions, in order to develop vehicles for controlled release of drugs like ciprofloxacin (fluoroquinolone antibiotic), as well as a film supporter for proliferation of human keratinocytes in tissue engineering areas. This polymer complexation led to better mechanical properties, controlled release of drug under triggering stimulus like pH changes, and the improvement of cell proliferation. In addition, both complexed biopolymer hydrogels showed good mucoadhesive properties, due to the presence of the free carboxylic groups in TG and CMTG, and pH-sensitive swelling and drug release behavior (Shaw et al., 2015, 2017).

14.4.3 Tamarind gum in Bigel and Emulgel systems

Gel-based formulations have been explored and used as controlled delivery systems, and can be classified on hydrogels, which consist of a colloidal network of a hydrophilic polymer that traps water molecules (McKee et al., 2014), or oleogels, which may consist of either amphiphilic or hydrophobic crystals (sorbitan monostearate, sorbitan monopalmitate, stearic acid, and stearyl alcohol) that form a network that immobilizes oils (Patel et al., 2014). Formulations where hydrogels and oleogels are mixed to form a new type of gelled structure are regarded as bigels. Since hydrogels are polar and oleogels are apolar, bigels may be regarded as emulsions having both internal and external immobilized phases (Kodela et al., 2017). In this sense, the immobilization of the external phase prevents the motion of the internal phase, and hence the occurrence of coagulation of the internal phase is avoided; as the internal phase is also immobilized, the leaching of the internal phase is minimized (Satapathy et al., 2015). Moreover, bigels exhibit inherent thermodynamic stability compared to the emulsions, even though both bigels and emulsions are biphasic formulations. It is noteworthy that if the external phase of the bigels is externally cross-linked, it will result in the formation of a permanent bigel (Paul et al., 2018).

Paul et al. (2018) have described the suitability of hydrogel-in-oleogel and oleogel-in-hydrogel bigels, by using an oleogel prepared with stearic acid and rice bran oil, and a hydrogel based on tamarind gum with a hydroethanolic solution, for drug delivery systems.

Bigel systems exhibited significant differences in the structural arrangement, while oleogel showed the presence of a hyperbranched fibrous structure formed by the stearic acid and tamarind gum hydrogel that indicated the presence of numerous small droplet-like structures. In the bigels, both distinct phases were observed: a near-globular phase present in a continuous phase and a branched fibrous structure, attributed to the network formed by the stearic acid molecules and irregularly shaped black globular bodies assigned to the presence of hydrophilic tamarind gum gel. Clear differences between the microarchitecture conformation of the oleogel, hydrogel-in-oleogel type of bigel, an oleogel-in-hydrogel type of bigel, and a hydrogel were assessed. Among the physicochemical properties for bigel systems, a reduction in the electrical impedance was observed and was dependent on the hydrogel proportion used in the bigel. In terms of their use for carrying and controlled release of moxifloxacin as a model drug, the formulations exhibited diffusion-mediated drug release, and improved significantly in a composition-dependent manner as the tamarind gum hydrogel proportion increased, maintaining the antimicrobial activity in the gelled matrix (Paul et al., 2018).

On the other hand, emulgels or emulsion gels consist in semisolid multiphase systems with both emulsion and gel properties, in which the liquid phase is immobilized in a structured/gel phase (Farjami and Madadlou, 2019). Gelation of an emulsion can be achieved by two methods: gelation of the continuous phase and aggregation of the emulsion droplets, and it can be formed based on the use of proteins, carbohydrates, or a mixture of these macromolecules (Nasirpour-Tabrizi et al., 2020). In this regard, Rawooth et al. (2020) developed tamarind gum and rice bran oil (RBO)-based emulgels for carrying and delivering ciprofloxacin as a model drug. Their findings showed that these systems were of biphasic nature, with the presence of two types of globular structures associated to the aqueous phase, apparently trapped in a continuous hydrophobic matrix from xyloglucan moieties and those corresponding to the RBO droplets. Moreover, the RBO content in emulgel display an effect on the reduction of hydrogen bonding between the components; despite the in vitro diffusion of ciprofloxacin being decreased with the RBO increase, the corneal permeation was improved with the increase in the RBO content. Therefore, emulgel systems had excellent potential for obtaining controlled and sustained delivery systems for therapies in ocular drug delivery.

14.5 Tamarind gum in industrial applications

14.5.1 Food applications

Tamarind gum has interesting physicochemical properties that give it the possibility of being applied as a food additive. Some recent publications on food applications of tamarind gum are as follows. In baking, tamarind gum was used to optimize the batter characteristics for good performance during leavening and to obtain a suitable final texture of gluten-free rice bread. Tamarind gum was applied at 1% and 2% of concentration, and the optimized formula was obtained using 1% of tamarind gum, 100 g water, 5 min of mixing time, and 60 min of fermentation time (Hong and Kweon, 2020). In another study, the addition of 0.2%, 0.4%, and 0.8% of tamarind gum to gluten-free cakes was probed and the sample with 0.4%

tamarind gum had the better results since increased the dietary fiber, improved the sensory characteristics, and extended the shelf life of the cakes compared to cakes made with 100% rice flour and wheat flour (Wu et al., 2020).

In food packaging, tamarind gum has been used in the production of edible films. The xyloglucan extracted from tamarind seeds was used for elaborating films for packaging cut-up "Sunrise Solo" papaya, which showed good physical characteristics. The film with better results was prepared using 4.5% of xyloglucans and 1.5% of glycerol. To conclude this, the study involved the analysis of the moisture, tensile strength, elongation at break, and mass loss when it was applied (Santos et al., 2019). In another study, tamarind xyloglucan films were added to sesame seed oil (0–20 wt% based on xyloglucans) for preparing emulsion films by different methods. The emulsion films were analyzed by droplet size, permeability, and tensile properties; in addition, these could have antimicrobial and/or antioxidant characteristics that may be useful when applied in foods (Rodrigues et al., 2018).

On the other hand, tamarind gum was used as a wall material in microencapsulation of sesame seed oil using ratios of 1:1 and 1:2, respectively. Both types of microcapsules were compared, and the 1:1 tamarind gum-sesame seed oil ratio had a smaller droplet size, higher thermal stability, higher encapsulation efficiency (91%), and higher oil oxidation stability after 6 weeks than the 1:2 ratio microcapsules. Thus, tamarind gum can be used as a wall material for protection of edible oils against oxidation, increasing their shelf life (Alpizar-Reyes et al., 2020).

Tamarind gum was mixed with starches (potato, rice, mung bean, and lotus root), as binary mixtures, in order to improve their physicochemical, textural, and rheological characteristics that allow them to withstand freezing thawing and gelatinization processes, and to delay their retrogradation. In this work, tamarind gum significantly increased the viscosity of mung bean starch gels and improved the texture properties and mouthfeel of lotus root starch gels, which is useful in baked foods (Liu and Xu, 2019). On the other hand, tapioca starch was mixed with tamarind xyloglucan in different ratios (10:0, 9:1, 8:2, 7:3, and 6:4, respectively) using a total concentration of 5% of the mixtures. The addition of tamarind xyloglucan to tapioca starch increased the viscosity and improved the thermal stability during the freeze-thaw process of gelatinized mixtures regarding tapioca starch (Pongsawatmanit et al., 2006).

14.5.2 Pharmaceutical applications

Tamarind gum has many applications in drug formulation, and an important issue in this field is the drug release. Some recent publications of pharmaceutical applications of tamarind gum are as follows.

Tamarind gum (crude and modified) was used as a binder in the tablets formulation of diclofenac sodium by freeze drying, with the aim of reducing oral dissolution times and of improving the drug release. Chemically modified tamarind gum tablets had better results since they enhanced the dissolution rate of the drug and achieved the complete release of the drug (Huanbutta et al., 2019). In another study, CMTG and crude tamarind gum were used to formulate Thai cordial tablets. Tamarind gum was carboxymethylated at different substitution degrees. The tablets were evaluated by their swelling and erosion behavior and were fractured for knowing their breakdown times. CMTG tablets had higher hardness and faster disintegration than crude tamarind gum tablets (Huanbutta and Sittikijyothin, 2017).

Tamarind gum has been used alone or mixed with other biopolymers, to allow controlled oral release of drugs, and some examples are shown below. Tamarind gum was blended with gellan gum through Ca^{2+}-ion cross-linked ionically gelation technique for forming beads of metformin HCl for oral drug delivery. In this work, an optimization was performed where the factors were gellan gum to tamarind gum ratio and $CaCl_2$ concentration, and the response variables were the drug encapsulation efficiency and the cumulative drug release after 10 h. The optimized beads showed good mucoadhesivity and hypoglycemic activity in alloxan-induced diabetic rats due to proper metformin release (Nayak et al., 2014). In a similar optimization study, tamarind gum was blended with alginate by ionotropic gelation with $CaCl_2$ to form composite beads of diclofenac sodium. FTIR and NMR analyses were carried out in order to evaluate biopolymer-drug compatibility. The swelling, degradation, and drug release of the composite beads were influenced by pH changes. Finally, these composite beads allowed the controlled release of the drug for a prolonged time (Nayak and Pal, 2011).

In dental applications, tamarind gum (1%, w/w) was used to formulate oral disintegration tablets of tea powders for oral care. The tablets were analyzed regarding the swelling degree, hardness, friability, disintegration time, adhesiveness, and antimicrobial activity. Tamarind gum tablets of tea powders had a low disintegration time, high mucoadhesivity, and antimicrobial activity against *S. mutans* (Kiniwa et al., 2019). Likewise, tamarind seed gum was used to formulate a thermoreversible gel with lidocaine hydrochloride in order to be applied as local anesthesia into periodontal pocket. The mucoadhesive property of this gel allowed the retention in the site of application and the immediate action of the drug with a release time up to 2 h, which allows a dental procedure to be carried out without pain. This gel is natural, of low cost, and biodegradable, and is an alternative to injected anesthesia (Pandit et al., 2016). In addition, tamarind gum was used to prepare buccal patches of metronidazole. The patches formulation was optimized using as factors: tamarind gum, epichlorohydrin (cross-linker), and propylene glycol (plasticizer), and as response variables: ex vivo drug permeation, mucoadhesiveness strength, folding endurance, and buccal residence time. It was determined that tamarind gum can be used in buccal patches due to its mucoadhesiveness and its drug release mechanism by controlled dissolution (Jana et al., 2010).

The drugs have other routes of application than oral—for example, tamarind gum tablets were used for colon delivery of propranolol HCl to treat blood pressure. These tablets were analyzed by in vitro release studies at the following conditions: 0.1 N of HCl for 1.5 h, then pH 6.8 phosphate buffer for 2 h and pH 7.4 phosphate buffer until complete drug release, and the release profiles were fitted to different pharmacokinetic mathematical models. The prolonged drug release time of tamarind gum tablets, compared to other biopolymers used in this study, had good compression characteristics; tamarind gum is also a cost-effective material (Newton et al., 2015). On the other hand, tamarind gum was used in the formulation of FITC-dextrans microparticles by spray-drying to be applied as a drug in the nasal cavity, which favored its transport to the brain. The microparticles were analyzed by size, morphology, and mucoadhesiveness using laser diffraction, scanning electron microscopy, and a texture analysis, respectively. Through this work, it was found that 10 μm-sized tamarind gum microparticles achieved better deposition in the nasal cavity than smaller particles (Yarragudi et al., 2017).

In ocular applications, tamarind gum and rice bran oil (0%, 5%, 10%, 15%, and 20%) were used to formulate emulgels for ocular delivery of the antibiotic ciprofloxacin HCl. The effect of different rice bran oil concentrations was evaluated in the application of these emulgels. In vitro release study of the drug showed lower release when the emulgels contained rice bran oil; in contrast, ex vivo corneal permeation showed higher drug release as rice bran oil concentration increased in the emulgel (Rawooth et al., 2020). Tamarind xyloglucan nanoaggregates were loaded with tropicamide for ophthalmic delivery. In this study, an optimal formula was found using 0.45% (w/v) of tamarind xyloglucan and 0.55% (w/v) of poloxamer-407 which had higher corneal permeation of the drug than a commercial aqueous formulation. These nanoaggregates had high mucoadhesiveness due to the tamarind gum, and did not irritate the eye (Dilbaghi et al., 2013). Likewise, carboxymethylated tamarind gum nanoparticles were loaded with tropicamide via ionotropic gelation for ocular delivery. An optimization study revealed that carboxymethylated tamarind gum and $CaCl_2$ concentrations had a synergistic effect on particle size and encapsulation efficiency; in addition, these nanoparticles showed ex vivo corneal permeation due to their mucoadhesiveness (Kaur et al., 2012a, b). In another work, tamarind gum and hyaluronic acid were mixed in different ratios, for use as potential excipients of eye drops and to detect a possible synergistic effect with respect to the polymers separately. The mixtures were analyzed using NMR to evaluate the interpolymeric interactions. Tamarind gum-hyaluronic acid (3:2) mixtures formed stable supramolecular aggregates that retained water, had high mucoadhesiveness and low viscosity, stabilized the tear film, and achieved the maximum residence time of the drug in the precorneal area of rabbit eyes (Uccello-Barretta et al., 2010).

Another route of drug application is topical. Tamarind gum was used to produce nanofiber patches using polyvinyl alcohol and fabricated by electrohydrodynamic atomization, and then the patches were loaded with clindamycin (1%–3%) to be applied as wound-dressing materials. These nanofiber patches had skin adherence, were translucent, and had ventilation properties to be applied topically. The diameter of nanofibers was affected by the voltage applied during their formation. The nanofiber patches were analyzed by scanning electronic microscopy, differential scanning calorimetry, and X-ray diffraction, and the antimicrobial activity of clindamycin patches was probed on *S. aureus* (Sangnim et al., 2018). In a similar study, carboxymethylated tamarind gum was added to polyvinyl alcohol to synthesize composite films of ciprofloxacin with mechanical, thermal, and antibiotic properties for skin diseases. Through FTIR spectroscopy, the presence of hydrogen bonding was revealed between the components of the films. These ciprofloxacin films had antimicrobial activity against *E. coli*; on the other hand, these films achieved cell proliferation using human keratinocytes so these can be applied in skin tissue engineering (Yadav et al., 2017). Tamarind gum was also mixed with glycerin and propylene glycol in order to prepare clindamycin transdermal patches which allowed a controlled release of the drug. The patches were evaluated by tensile strength, drug release, and antimicrobial activity against *S. aureus*. The incorporation of different glycerin-propylene glycol ratios to tamarind gum patches affected the properties of the transdermal patches, the 4:6 ratio being the one that had the best results in terms of drug release and antimicrobial activity (Sureewan et al., 2014).

Drug hydrogels are also applied in a topical way. Tamarind gum was mixed with gelatin gum to form a hydrogel that was added with three different types of carbon nanotubes and filled with salicylic acid. Through field emission SEM, these hydrogels were observed as agglomerates where the carbon nanotubes were confined within a dispersed phase of tamarind gum. The carbon nanotubes had interactions with the hydrogel which had large crystallite size, and a mechanical study revealed that these hydrogels had better resistance to the breakdown than the control. The drug was released by the diffusion method and these hydrogels were cytocompatible with human keratinocytes; thus, they can be applied in wound healing and tissue engineering (Maharana et al., 2017).

Carboxymethylated tamarind gum was chemically cross-linked with gelatin to obtain a biocomposite hydrogel that efficiently retained aceclofenac, a drug with an antiinflammatory effect. The drug was compatible with the interpenetrating network of the biocomposite hydrogel. The rate of drug release mainly depended on the degree of cross-linking, the tamarind gum concentration, and the pH of the medium. Finally, the biocomposite hydrogel loaded with aceclofenac had a prolonged antiinflammatory effect on rats (Jana et al., 2016). In a similar study, CTG was cross-linked with citric acid to form ester cross-links between them in order to prepare hydrogel films loaded with a model drug (moxifloxacin hydrochloride). The formation of ester cross-links was analyzed using ATR-FTIR, solid-state ^{13}C NMR study, and differential scanning calorimetry. These hydrogel films achieved high drug retention, had a controlled release of drug, and can be applied topically (Kaur et al., 2010; Mali et al., 2017b).

Tamarind gum was used to prepare ion gels using both synthetic ionic liquids (1-butyl-3-methylimidazolium chloride and 1-butyl-3-methylimidazolium bromide) and bio-based ionic liquids (choline acrylate, choline caproate, and choline caprylate) by heating/cooling processes. These ion gels had viscoelastic behavior with thixotropic nature, and managed to adhere to human finger muscles and skin; thus, ion gels can have applications as sensors and actuators, and even gels with bio-based ionic liquids could have biomedical applications (Sharma et al., 2014).

On the other hand, tamarind xyloglucan has been used to enhance skin regeneration. Xyloglucans were extracted using cold water and a copper complex precipitation, and these were applied to human skin keratinocytes and fibroblast in vitro. After being analyzed, it was determined that tamarind xyloglucans promote the reepithelization and remodeling of the skin through cell proliferation and migration (Nie and Deters, 2013).

14.5.3 Other fields of tamarind gum applications

Tamarind gum, crude and CMTG, was used as a wall material in citronella oil microencapsulation in order to evaluate the oil release rate. Three different formulations of microcapsules were prepared varying the gum-oil ratio (1.25, 1.14, and 0.87). The microcapsules were characterized by SEM, encapsulation efficiency, and the oil release rate was evaluated. CMTG microcapsules allowed slower oil release than those of crude tamarind gum. Citronella oil is used in perfumery and is a natural repellent of insects, so tamarind gum microcapsules guarantee the release of the active compound for its proper functioning (Khounvilay et al., 2019).

In agricultural matter, CMTG was mixed with sodium-acrylate in order to create superabsorbent hydrogels for conditioning soils. The hydrogels' characterization was performed by FTIR spectroscopy, thermal analysis, SEM, and swelling studies. The soils mixed with these superabsorbent hydrogels (0.1%–0.3%) augmented the moisture absorption up to 35%, the porosity up to 7%, and the water retention capacity by planting chickpea seeds, compared to untreated soil. The superabsorbent hydrogels work by conditioning soils because they are excellent water retainers and nutrient carriers; they are also degradable (Khushbu and Kumar, 2019).

In environmental matter, tamarind gum was used to create an amphiphilic graft copolymer with methyl methacrylate by the method of atom transfer radical polymerization (ATRP) using the mixture CuBr/bpy as a catalyst, with the aim to remove toxic dyes. Copolymer characterization was performed by FTIR spectroscopy, ^{1}H NMR spectral analysis, gel permeation chromatography, TGA, DLS, field emission-SEM, and energy dispersive X-ray spectroscopy. Furthermore, adsorption and desorption studies were carried out. The copolymer had excellent sorption capacity of methylene blue and Congo red dyes, and the selective adsorption of the dyes was highly influenced by pH variations which had an effect on the electrostatic and H-bonding interactions between copolymer and dyes (Pal et al., 2012).

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