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Rheological properties of tamarind (*Tamarindus indica* L.) seed mucilage obtained by spray-drying as a novel source of hydrocolloid

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ABSTRACT

Tamarind seed mucilage (TSM) was extracted and obtained by spray drying. The power law model well described the rheological behavior of the TSM dispersions with determination coefficients R^2 higher than 0.93. According to power law model, non-Newtonian shear thinning behavior was observed at all concentrations (0.5%, 1%, 1.5% and 2%) and temperatures (25, 30, 40, and 60 °C) studied. Increasing temperature decreased the viscosity and increased the flow behavior index, opposite effect was observed when increasing the concentration. The temperature effect was more pronounced at 2.0% TSM concentration with an activation energy of 20.25 kJ/mol. A clear dependence of viscosity on pH was observed, as pH increased from acidic to alkaline conditions, the viscosity increased. It was found that the rheological properties of TSM were affected by the sucrose and salts and their concentrations as well due to the addition of ions (or sucrose) decreases repulsion and allows molecule expansion promoting a significant reduction in viscosity. These results suggest that TMS could be applied in the production of foods that require additives with thickening capacity.

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1. Introduction

Hydrocolloids as biological macromolecules are widely used in the pharmaceutical, cosmetic, textile, paper and food industries as thickeners, water retention agents, emulsion stabilizers, suspending agents, binders, etc. [1–4]. In recent years, the mucilages are the preferred hydrocolloids since these are cheap, nontoxic, ecofriendly and nonpolluting during production and application, because they are able to bind and immobilize a large amount of water [5–7]. Therefore, the rheological behavior of hydrocolloids is of special importance since they are applied looking to modify the textural attributes of food. Rheological behavior of every biological macromolecule is unique and this information is very useful in a large number of industrial applications and should be carefully taken into account for designing and modelling

purposes [8–10]. Shear-thinning or pseudoplastic non-Newtonian behavior has been reported in gums and mucilages; this property depends on many factors like concentration of the active compound, temperature, degree of dispersion, dissolution, electrical charge, previous thermal and mechanical treatment, presence or absence of other lyophilic colloids, and the presence of electrolytes or non-electrolytes [5,9,11–13]. Also, the chemical structures of the hydrocolloids and its conformation, particle size distribution, and particle shape, as well as the interactions between suspended particles, are known to affect flow behavior [10–14]. Mucilage extracted from the tamarind seed (TSM, up to 72%), is a natural polysaccharide available as a by-product of tamarind pulp industry mucilage [12]. TSM is composed of β -(1,4)-D-glucan backbone 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 glucose, xylose and galactose units are present in the ratio of 2.8:2.25:1.0, as the monomer units [15] and with a molecular weight of 720–880 kDa [12]. The mucilage dispersed in water has the ability of forming viscous solutions, with high thermal and chemical stability, edible, biodegradable, non-carcinogenic,

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biocompatible and nontoxic properties [15]. TSM contains high amounts of many essential amino acids, like isoleucine, leucine, lysine, methionine, phenylalanine and valine [16], making the mucilage affordable as food additive. However, detailed data about the rheological behaviour of TSM spray dried at different environments is not available. This information is valuable to establish possible industrial applications, particularly for food products. TSM has the added benefit of employing a plant-based resource from a by-product of tamarind pulp industry, which is not been exploited yet, and could have a positive impact on the development of producer economies. Therefore, the aim of the present study was to characterize the rheological properties of aqueous dispersions of TSM spray dried obtained from tamarind seeds grown in Mexico, evaluating the effect of some conditions of the medium such as mucilage concentration, temperature, pH, type of salt, and sucrose concentrations.

2. Materials and methods

2.1. Materials

Tamarind dried pods were purchased at a local market of Toluca City, Mexico. The initial moisture content of tamarind seeds was ~2.5 kg water/kg dried solid. Chemical reagents were purchased from Sigma Aldrich S.A. de C.V. (Toluca, Mexico). All the water used in the experiments was deionized.

2.2. Tamarind seed mucilage extraction

Mucilage extraction was performed taking as basis the method proposed by Khounvilay and Sittikijyothin [12] with some modifications. The seeds were extracted manually from mature pods of tamarind, milled and grounded through a 355 μm mesh using a hammermill Pulvex 100 Mini 2HP (Mexico City, Mexico). 20.0 g of milled tamarind seeds were placed in 1.0 L beaker and deionized water was added in a 1:10 wt. ratio. The resulting mixture was stirred in a hot plate stirrer (Thermo scientific SP131325, China) adjusted at level 8 for 10 min to achieve a homogenous mixture. Deionized water was added in a 1:40 wt. ratio in relation with the initial weight of seeds and kept with constant stirring. The mixture was heated and kept at a constant temperature of 80 °C for 60 min. The mixture was put aside at 20 °C for 24 h to assure the release of the mucilage, and then was centrifuged with a Hermle Z323 K highspeed centrifuge (Hermle, Labortechnik, Germany) for 8 min at 524g. The supernatant represents the mucilage fraction, which was decanted and stored at ~4 °C for subsequent analysis.

2.3. Spray drying of tamarind seed mucilage

The extracted mucilage was feed at a rate of 40 mL/min to a Nichols/Niro spray-drier (Turbo Spray PLA, NY, USA) operated with an inlet temperature of 135 \pm 5 °C, outlet temperature at 80 \pm 5 °C and injecting compressed air at 4.0 bar. The spray-dried mucilage was stored in desiccators containing P₂O₅ to prevent increases in absorbed moisture.

2.4. Proximal analysis

Moisture, lipid and ash content of tamarind seed mucilage was determined according to the AOAC standard methods, 925.10, 920.85 and 923.03, respectively [17]. The total protein content of the mucilage was determined by Kjeldahl procedure (N \times 6.25) as described in AOAC official method 981.10. Total carbohydrate content was evaluated by difference.

2.5. Rheological measurements

Rheological characterization was carried out using steady shear tests using a Physica MCR 300 (Physica Meßtechnik GmbH, Stuttgart, Germany) modular compact rheometer, with a cone-plate geometry, with cone diameter of 50 mm in diameter and cone angle of 2° in all the cases. The viscosity of mucilage dispersions were carefully poured in the measuring system, and left to rest for 5 min for structure recovery and temperature equilibration. The apparent viscosity (η_{app}) – shear rate ($\dot{\gamma}$) behaviour of the mucilage dispersions was determined by applying an increasing shear rate from 0.1 to 100 s⁻¹. Three replicates of each sample were made. The flow behavior index (n) and consistency index (k) values were computed by fitting to the Ostwald-de-Waele or Power Law model (Eq. (1)):

$$\tau = k\dot{\gamma}^n \quad (1)$$

where τ is the shear stress (Pa), $\dot{\gamma}$ is the shear rate (s⁻¹), k is the consistency coefficient (Pa s ^{n}) and n is the flow behavior index (dimensionless).

2.6. Evaluation of the temperature dependence of mucilage dispersions

TSM dispersions were prepared with deionized water at concentrations of 0.5%, 1.0%, 1.5% and 2% (w/w), under slow stirring at room temperature. Then the dispersion was kept at 4 °C for 24 h to complete hydration prior to assessment. Prepared samples were loaded into the plate and maintained for 5 min at measurement temperatures of 25, 30, 40 and 60 °C. A solvent trap was used in order to minimize the solvent loss due to evaporation. Optical microscopy images were obtained in a microscopy MOTIC BA-400 Xiamen, China, at resolution of 100x for concentrations of 0.5%, 1.0%, 1.5% and 2% (w/w) of TSM at room temperature. Three replicates of each sample were made.

The temperature dependency of consistency coefficient (indicator of the viscous nature of the sample) was assessed by fitting to the Arrhenius model (Eq. (2)) as was suggested by Sengul et al. [18]:

$$k = k_0 e^{(E_a/RT)} \quad (2)$$

where k_0 is the proportionality constant (or consistency coefficient at a reference temperature, Pa s ^{n}), E_a the activation energy (J/mol), R the universal law gas constant (8.314 J/mol K), and T the absolute temperature (K).

2.7. Determination of flow properties at different pH

Dispersions of TSM at 1.0% (w/w) were used to measure the flow properties with different pH values of 4.0, 7.0 and 10.0 (adjusted using 0.1 mol/L of NaOH and HCl) at shear rates from 0.1 to 100 s⁻¹ and constant temperature of 25 °C. Optical microscopy images were obtained in a microscopy MOTIC BA-400 Xiamen, China, at resolution of 100 \times for this dispersions of TSM at room temperature. Three replicates of each sample were done.

2.8. Flow properties at different salts concentrations

Dispersions of TSM at 1.0% (w/w) were prepared. Monovalent (NaCl and KCl) and divalent (CaCl₂) salts were added to dispersions to give final concentrations of 0.01 M, 0.02 M and 0.03 M, respectively. Viscosity measurements were performed at a shear rate from 0.1 to 100 s⁻¹ and keeping constant temperature at 25 °C. Three replicates of each sample were made.

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