Carbohydrate Polymers 91 (2013) 394–402

Contents lists available at SciVerse ScienceDirect

Carbohydrate Polymers

journal homepage: www.elsevier.com/locate/carbpol

Rheological and physical properties of spray-dried mucilage obtained from *Hylocereus undatus* cladodes

E.E. García-Cruz[a], J. Rodríguez-Ramírez[a], L.L. Méndez Lagunas[a,*], L. Medina-Torres[b]

[a] Instituto Politécnico Nacional, Centro Interdisciplinario de Investigación para el Desarrollo Integral Regional, Unidad Oaxaca, Hornos No. 1003, 71230 Oaxaca, Mexico
[b] Facultad de Química "E", Universidad Autónoma de México (UNAM), 04510 México, D.F, Mexico

ARTICLE INFO

Article history:
Received 1 March 2012
Received in revised form 10 July 2012
Accepted 13 August 2012
Available online 22 August 2012

Keywords:
Pitahaya mucilage
Rheological properties
Cross model
Physical properties
Optimization

ABSTRACT

This study examines the rheological behavior of reconstituted spray-dried mucilage isolated from the cladodes of pitahaya (*Hylocereus undatus*), the effects of concentration and its relationship with physical properties were analyzed in reconstituted solutions. Drying process optimization was carried out through the surface response method, utilizing a factorial 2^3 design with three central points, in order to evaluate yield and rheological properties. The reconstituted mucilage exhibited non-Newtonian shear-thinning behavior, which adequately fit the Cross model ($R^2 > 0.95$). This dynamic response suggests a random coil configuration. The steady-shear viscosity and dynamic response are suitably correlated through the Cox–Merz rule, confirming the mucilage's stability of flow. Analysis of the physical properties of the mucilage (Tg, DTP, and particle morphology) explains the shear-thinning behavior.

1. Introduction

The selection of new sources for biopolymers requires an understanding of functional properties such as rheological and physicochemical characteristics (Chin, Chan, Yusof, Chuah, & Talib, 2009). Study of polysaccharide flow properties has contributed to identification of potential applications, development of new products, and methods for evaluating quality and stability during storage. Furthermore, identification of rheological properties has made it possible to characterize the structure of a given biopolymers and understand the changes that it undergoes as a result of preservation processes (Morris, Cutler, Ross-Murphy, Rees, & Price, 1981; Nindo, Tang, Powers, & Takhar, 2007). Mucilages and gums are heteropolysaccharides with rheological properties that are of great interest for a number of applications. The hydrocolloid characteristics of these biopolymers are the primary motivation for their utilization in the foodstuffs, environmental, cosmetic and construction industries (Sáenz, Sepúlveda, & Matsuhiro, 2004). Biological sources of mucilage that have been studied include flaxseed gum, *Alyssum homolocarpum* seed, nopal cactus (*Opuntia ficus indica*) (Koocheki, Mortazavi, Shahidi, Razavi, & Taherian, 2009; León-Martínez, Rodríguez-Ramírez, Medina-Torres, Méndez Lagunas, & Bernad-Bernad, 2011; Medina-Torres, Brito-De La Fuente, Torrestiana Sánchez, & Katthain, 2000; Wang, Li, Wang, Li, & Adhikari, 2010). The demand for hydrocolloids with a specific functionality has increased recently in the foodstuffs industry (Koocheki et al., 2009); to satisfy this demand, research on new sources of polysaccharides, such as *Hylocereus undatus*, is needed. The rheological properties of dragon fruit (*Hylocereus* spp.) purees have been studied (Liaotrakoon et al., 2011); however, the properties of mucilage isolated from the stalks of the plant have not been investigated.

Shear-thinning or pseudoplastic non-Newtonian behavior has been reported in gums and mucilages; this property depends on factors such as pH, ionic strength, and dissolved solids concentration (Koocheki et al., 2009; León-Martínez et al., 2011; Medina-Torres et al., 2000). The chemical structure of the polysaccharide and its conformation, particle size distribution, and particle shape, as well as the interactions between suspended particles, are known to affect flow behavior (Chiou & Langrish, 2007; Nindo et al., 2007).

The behavior of non-Newtonian fluids can be represented through various rheological models, including the Cross model, which describes shear-thinning behavior and includes the Newtonian zone (Chin et al., 2009; Medina-Torres et al., 2000).

The properties of viscoelastic materials, such as certain polymers, are determined via small-amplitude oscillatory tests. The elastic modulus (G'), viscous modulus (G''), and complex viscosity (η^*) provide information on the structure and molecular configuration of materials, whereas the loss tangent ($\tan\delta$) denotes the relative effects the components G' and G'' exert on viscoelastic

* Corresponding author. Tel.: +951 51 7 06 10x82777.
E-mail addresses: lmendezla@ipn.mx, mendezll@hotmail.com (L.L. Méndez Lagunas).

0144-8617/$ – see front matter © 2012 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.carbpol.2012.08.048

behavior. The dynamic and shear flow characteristics of polymer solutions are correlated using the Cox–Merz rule (Gunasekaran & Mehmet, 2000).

Mucilages have a short shelf life. In order to extend this shelf life, methods such as spray-drying are employed; the resulting powders can then be reconstituted with water at the time of use. Various authors have investigated the mechanical flow properties of spray-dried products reconstituted in aqueous media, such as mucilages, gums, fruit juices, and purées (León-Martínez, Méndez-Lagunas, & Rodríguez, 2010; León-Martínez et al., 2011). The conditions of the spray-drying process affect the functional properties and yield of these powders (Gharsallaoui, Roudaut, Chambin, Voilley, & Saurel, 2007); nevertheless the effects of drying parameters on the properties of pitahaya mucilage are unknown.

Additionally, research has been directed at the optimization of this preservation process for sugar-rich foods, through utilization of glass transition temperatures (Truong, Bhandari, & Howes, 2005) and rheological property point values (Oomah & Mazza, 2001); however, rheological model parameter fit values have not yet been utilized in the optimization of this process. The use of these values makes it possible to study the Newtonian, pseudoplastic zone of mucilage's flow curve, which is not possible through the point values method. Therefore, the objective of the present study is to characterize the rheological properties of reconstituted spray-dried mucilage isolated from the cladodes of the pitahaya plant (*H. undatus*); evaluating the effects of spray-drying conditions and concentration. An analysis of the relationship between the rheological behavior and physical properties (glass transition temperature, particle size distribution, powder morphology) of the biopolymer is also presented, along with the fit of the experimental data to a rheological model, and the use of these parameters to optimize the conditions of the spray-drying process.

2. Methodology

2.1. Mucilage extraction

Cladodes of *H. undatus* were collected from an experimental field located at 96°44′ longitude west and 17°04′ latitude north in Mexico. 12-month-old stalks with a moisture content of 83.4% (wb) were carefully washed in potable water and scrubbed with a plastic-bristled brush to remove the spines. A steel knife and a vegetable peeler were used to slice the stalks lengthwise to produce sheets with a length of 12.5 ± 2.5 cm and a width of 2 ± 0.3 mm. Mucilage was extracted by heating a 1:4 mixture of vegetal matter and deionized water at 80 °C for 1 h. Solid matter was separated via decantation, and the aqueous extract with a concentration of 1° Brix was filtered through a 100 screen. The mucilage was stored in refrigeration at 5 °C during 12 h to avoid enzymatic changes.

2.2. Spray-drying

A co-current pilot-scale spray dryer (Niro, Copenhagen, Denmark) equipped with a rotary atomizer (TS-Minor, M02/A) was used for all samples. Distilled water at room temperature (25 °C) was used in the initial feeding for 5 min. Room temperature mucilage, at 1° Brix, was fed into the dryer with the help of a peristaltic pump (Watson-Marlow 505S/RL). A 2^3 factorial design including three replications at the central point carried out in random order (Table 1) was used to evaluate the effects of the following variables on yield of powders and rheological properties of reconstituted mucilage: inlet air temperature, Ti (130–170 °C); feed flow, Fe (2.2–2.8 $\mathrm{Lh^{-1}}$); and atomization speed, As (16,000–22,000 rpm). The spray dryer outlet temperature was measured.

2.3. Physical properties of pitahaya mucilage powders

2.3.1. Moisture content

Moisture content was determined using the gravimetric technique described in the A.O.A.C. (1984); moisture loss was expressed in percentage dry base (db).

2.3.2. Yield

Dry powder yield was determined with Eq. (1):

$$y = \frac{P_\mathrm{p} - X_\mathrm{wb}P_\mathrm{p}}{V_\mathrm{m}S_\mathrm{T}} \times 100 \tag{1}$$

where y is yield (%); P_p is powder weight (g); X_wb is the moisture content of the powders in wet base (wb); V_m is the volume of mucilage fed into the dryer (L); S_T is the total solids (g dry matter/L), determined with Eq. (2) in accordance with the A.O.A.C. (1984) rule.

$$S_\mathrm{T} = \left(\frac{P_2 - P_1}{M}\right) \times 1000 \tag{2}$$

where P_2 is the mass of the capsule with dry residue (g); P_1 is the capsule mass (g); and M is the sample volume ($\mathrm{cm^3}$).

2.3.3. Activity water

Water activity (aw) was determined using a Novasina (Switzerland) instrument at 25 °C.

2.3.4. Glass transition temperature

The glass transition temperature (Tg) of the T7 sample dried (Table 1) was determined using a differential scanning calorimeter (DSC) model 822E (Mettler Toledo). The equipment was calibrated using an empty capsule and a sapphire standard. Approximately 2.5 mg of mucilage powder were placed in an aluminum capsule and exposed to temperatures of 20–100 °C in 1 °C/min intervals, in a nitrogen atmosphere with a flow of 30 ml/min. Tg was considered to be the median temperature of the thermogram.

2.4. Particle morphology of powders

Particle shape and surface morphology of powders were evaluated using a JEOL JSM5900-LV scanning electron microscope (SEM). The mucilage powders were fixed onto an aluminum slide using electrically conductive tape (Bal-Tec, Fürstentum Liechtenstein, Germany) and coated with gold at 10 mbar for 90 s (Polaron SC-7610, Fisson Instruments, CA, USA). The SEM was operated at 5 kV at two levels of magnification of 1000× and 3500× in secondary electron mode.

2.5. Rheological properties of reconstituted mucilage

2.5.1. Preparation of reconstituted solutions

The mucilage powders were reconstituted with deionized water for measurement of rheological properties; solutions were prepared at concentrations of 3% and 6% (w/v), utilizing magnetic agitation during 2 h at room temperature (25 °C). Solutions at 6% (w/v) were used to determine particle size distribution.

2.5.2. Mechanical flow behaviors

A stress-controlled rheometer (Model AR-G2, TA Instruments) with concentric cylinder geometry (21.96 mm outer cylinder diameter, 20.38 mm inner cylinder diameter, 59.50 mm height, and 500 μm gap) was used to determine rheological behavior. A constant temperature of 25 °C was controlled with a circulating bath (Cole Parmer Polystat, U.S., and a Peltier ARG2). Viscous behavior was analyzed in steady shear flow in the shear rate range of 1–600 $\mathrm{s^{-1}}$.

Table 1
Factorial design of drying and physical properties of pitahaya mucilage powders.

Treatment	Ti (°C)	As (rpm)	Fe (Lh^{-1})	Yield (%)	MC (db)	To (°C)
T1	130	16,000	2.2	80.25 ± 3.33	7.95 ± 0.52	70.5 ± 2.47
T2	170	16,000	2.2	57.23 ± 5.72	1.79 ± 0.11	88.0 ± 1.41
T3	130	22,000	2.2	75.42 ± 2.81	7.76 ± 0.81	58.5 ± 1.77
T4	170	22,000	2.2	64.12 ± 0.39	5.10 ± 0.03	75.5 ± 0.35
T5	130	16,000	2.8	71.05 ± 1.14	3.40 ± 0.06	71.0 ± 2.12
T6	170	16,000	2.8	64.46 ± 0.04	5.12 ± 0.18	63.0 ± 8.49
T7	130	22,000	2.8	85.54 ± 0.87	9.57 ± 0.20	56.0 ± 0.71
T8	170	22,000	2.8	70.40 ± 5.71	3.05 ± 0.01	78.5 ± 1.06
C1[a]	150	19,000	2.5	55.15 ± 9.80	5.48 ± 0.63	73.5 ± 1.06
C2[a]	150	19,000	2.5	71.98 ± 4.36	6.17 ± 0.60	74.0 ± 0.71
C3[a]	150	19,000	2.5	75.23 ± 1.74	6.39 ± 0.35	73.5 ± 1.06

[a] Central point.

Viscoelastic properties, storage modulus G', loss modulus G'', complex modulus G^*, and loss tangent $\tan\delta$ were determined through small amplitude oscillatory flow at frequencies ranging from 1 to 100 rad/s. Prior to dynamic experimentation, a strain sweep test was performed at constant frequencies of 1, 10, and 100 rad/s in order to fix the linear viscoelastic zone.

Experimental data in simple shear flow for reconstituted mucilage solutions was fit to the Cross model (Eq. (3)):

$$\frac{\eta - \eta_\infty}{\eta_0 - \eta_\infty} = \frac{1}{1 + (\lambda\dot{\gamma})^m} \quad (3)$$

where η is the shear viscosity (Pa s), $\dot{\gamma}$ is the shear rate (s^{-1}), λ is a relaxation time (s), m is the dimensionless index flow, and η_∞ and η_0 are the limit viscosities at high and low shear rates, respectively (Kirkwood & Ward, 2008).

2.6. Reconstituted mucilage particle size distribution

Powder particle size and distribution were determined using a laser diffraction particle analyzer Mastersizer 2000 (Malvern Instruments Ltd., UK). The equipment parameters were adjusted with a refractive index of 1.34 and an absorbance of 0.1.

2.7. Statistical analysis

Spray-drying tests were carried out in duplicate, and the resulting data was analyzed using variance analysis (ANOVA). Rheological and physical properties, with the exception of glass transition temperature, were evaluated in duplicate. The average and standard deviation of the values are presented below.

3. Results and discussion

3.1. Effects of drying conditions on the physical properties of the powders

The yield and dry base moisture content (MC) of the mucilage powders, as well as the output air dryer temperature (To) used for each treatment are presented in Table 1. The greatest yield was obtained with the T7 treatment, which combined a low inlet air temperature (Ti) with a high atomization speed (As) and feed flow (Fe). This may be attributable to the fact that low temperatures reduce the particle stickiness (Obón, Castellar, Alacid, & Fernández-López, 2009). In almost all treatments, an increase in yield was found with a decrease in Ti and an increase in As. This tendency has been found in other studies dealing with the recovery of powders produced by spray-dried sugar-rich products (Chegini & Ghobadian, 2007; León-Martínez et al., 2010; Liaotrakoon et al., 2011). High temperatures promote changes in the structure and thermoplastic state of the material, increasing deposits on the dryer wall (Chegini & Ghobadian, 2007; Goula & Adamopoulos, 2005). In mucilaginous materials with high sugar content, thermoplastic behavior is also affected by temperature, resulting in low yields (Gharsallaoui et al., 2007). The favorable effect of atomization speed on yield has been explained as a result of the reduction in particle size, which in turn increases contact surface area (Abadio, Domingues, Borges, & Oliveira, 2004); this is addressed below in the analysis of particle distribution size. The small size of the particles increases the transference of heat and mass. Inlet air temperature was the most statistically significant factor ($\alpha = 0.05$) affecting obtained yield (Table 2). The same effect has been reported in *Opuntia ficus indica* (*Ofi*) mucilage and in orange juice (Chegini & Ghobadian, 2007; León-Martínez et al., 2010). However, the present study produced a greater powder yield than that reported for *Ofi* mucilage (León-Martínez et al., 2010). The solids content present in the aqueous extract of *Ofi* mucilage was 2° Brix, which is greater than that found here for pitahaya mucilage extract. Oomah and Mazza (2001) found that low total solids content in the extract of flaxseed gum facilitates dryer feed due to low viscosity; it was also found to decrease stickiness, which in turn increases the yield of obtained powders. The behavior of T1 treatment could attribute to uncontrolled factors such as relative humidity of drying air, fluctuations in the compressed air flow rate and the drying gas flow rate (León-Martínez et al., 2010).

The moisture content (db) of the mucilage powders was less than 10% (Table 1), ensuring the stability of the product. The T2 treatment resulted in the lowest moisture content; this treatment combined high inlet air temperature with low atomization speed and atomization flow. Ti and As were found to affect the final moisture content ($\alpha = 0.05$) (Table 2). The greater the difference between the temperature of the drying medium (air) and the particles, the more quickly moisture is eliminated (Obón et al., 2009; Rodríguez-Hernández, González-García, Grajales-Lagunés, & Ruiz-Cabrera, 2005). Furthermore, a decrease in feed flow produces smaller-sized droplets, which, combined with a decrease in atomization speed, increase the time of contact with the hot air, thus reducing moisture content (Abadio et al., 2004; Goula & Adamopoulos, 2005). These results are similar to those obtained

Table 2
Variance analysis of the effects of process variables on mucilage powders properties.

Process variables	Sum of squares		
	Yield	Moisture content	Outlet temperature
Ti	784.84*	46.44*	600.25*
As	126.34	13.07*	144.00
Ti × As	2.51	5.61*	225.00
Fe	51.98	0.53	144.00
Ti × Fe	39.63	4.05*	100.00
As × Fe	84.46	0.24	156.25
Ti × As × Fe	102.92	34.39*	240.25*

* Significant term at $\alpha = 0.05$.

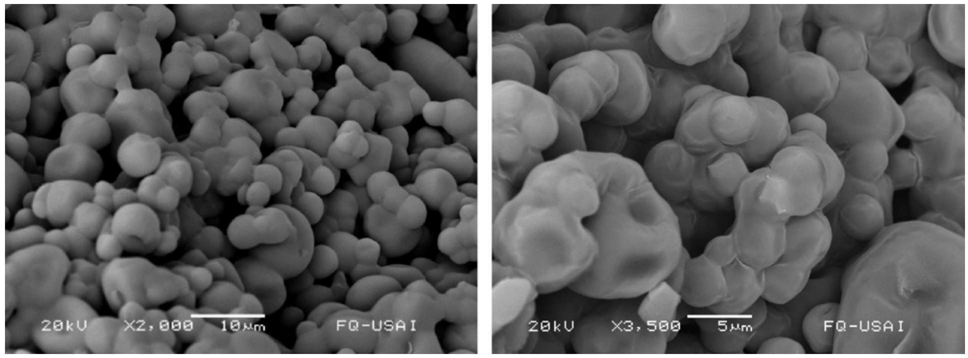

Fig. 1. SEM images of particle morphology of powders pitahaya mucilage, $a_w = 0.2$. Samples exposed to treatments T3 (A) and T1 (B).

for other mucilages and juices (Chegini & Ghobadian, 2007; Goula & Adamopoulos, 2005, 2010; León-Martínez et al., 2010; Obón et al., 2009; Rodríguez-Hernández et al., 2005). Outlet air temperature is also known to be a significant factor (Chegini & Ghobadian, 2007; León-Martínez et al., 2010). The lowest moisture content (T2) was obtained with the highest outlet air temperature of all of the treatments. Thus, the operating conditions, the time particles remain in the dryer, and drying speed have an influence on characteristics of the dry product such as moisture content and glassy state, as evaluated by the glass transition temperature (Tg). This parameter has been associated with dry product yield and storage conditions, and may explain stickiness (Chiou & Langrish, 2007).

3.2. Glass transition temperature (Tg)

The powder sample exposed to the T7 treatment had a mean glass transition temperature (Tg) of 48 °C. The low-molecular-weight sugars and acids present in mucilage are in an amorphous state during spray drying, due to the low time of exposure (Kasapis, 2005). However, the Tg value found in this study indicates that once the mucilage has dried and cooled, it exhibits a glassy state at room temperature (25 °C). An increase of approximately 20 °C above Tg results in unfavorable changes in the dry product; it may become rubbery and sticky, and suffer decreases in viscosity and increases in molecular mobility (Bhandari, Datta, & Howes, 1997; Busso, Schebor, Zamora, & Chirife, 2007).

The Tg of *H. undatus* is similar to that reported for *Ofi* mucilage (León-Martínez et al., 2010); in contrast, Tg values for xanthan gum, pectin, k-carrageenan, alginate, and starch, with moisture values below 30%, lie between 45 and 80 °C (Kasapis, 2005).

In addition, the low moisture content of pitahaya mucilage powders (<10%), decreases its hygroscopicity and ensures stability, as the plastification effect of water is reduced.

3.3. Particle morphology of powders

Fig. 1 shows the particle morphology of mucilage powders obtained with treatments T1 and T3 ($a_w = 0.2$). Image analysis (1500× and 3000×) reveals spherical particles with a skin-like particle structure, or polymeric appearance. The spherical nature of the particles suggests that the droplets dried symmetrically. This morphological characteristic is exhibited by organic materials and foodstuffs containing glucose-like or glucose-based carbohydrates, such as instant beverages, coffee, dairy products including yogurt, eggs, fruit juices, and spray-dried *Ofi* mucilage (Sáenz, Tapia, C, & Robert, 2009; Walton & Mumford, 1999). Mucilage powder particles formed a small proportion of agglomerates that seem to be linked together, with an observable hollow internal structure, similar to the cavities found in the crust of *Opuntia stricta* juice particles, as reported by Obón et al. (2009). Agglomeration may be attributable to electrostatic effects and Van der Waals forces. The formation of hollow or porous particles may be due to the desorption of air that had been absorbed by the droplets during the drying process (Walton & Mumford, 1999), and could negatively affect rehydration of the product, as a result of the air incorporated into the particles (Abadio et al., 2004). Agglomeration of the particles depends on the properties of the material of study, and may be reduced by modifying drying conditions. Flow properties are affected by particle shape and morphology, as well as by the degree of agglomeration; in the present case, pitahaya mucilage exhibits an increased resistance to flow with respect to *Opuntia* mucilage; this can be attributed to increased particle sphericity, and a minimal formation of agglomerates. Walton and Mumford (1999) suggest that spherical particles flow freely, as they lack surface roughness and do not form agglomerates.

3.4. Rheological behavior in simple shear flow

Flow curves of samples T3, T4 and T8 (Fig. 2) were selected, as they represent curves with extreme or intermediate viscosity values. Newtonian behavior was found at low shear rates, followed by a thinning (pseudoplastic) behavior in simple shear flow for mucilage reconstituted at 3% and 6% (w/v). The decrease in viscosity with an increase in shear rate may be attributable to the alignment of high-molecular-weight chains positioned randomly in the structure of the material. These chains interact with each other, and with the aqueous medium. The destruction of the structure is reversible, establishing a structural formation-destruction dynamic

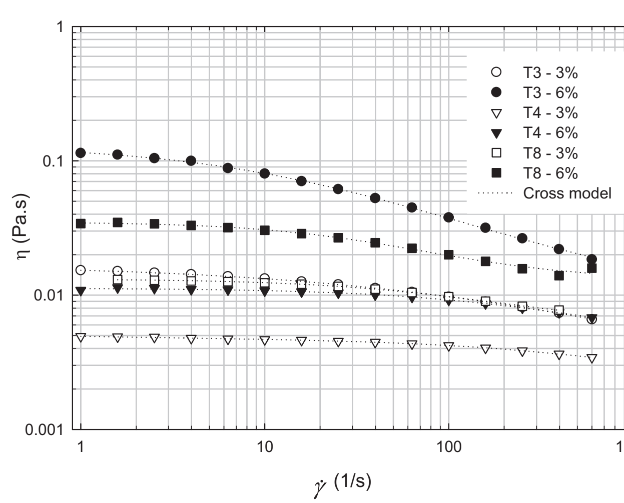

Fig. 2. Flow curves for reconstituted pitahaya mucilage at concentrations of 3 and 6% (w/v).

due to flow. The high-molecular-weight components of mucilage form aggregates. Electrostatic and Van Der Waals forces are weakened and aggregates in a solution disassociate when a deformation is applied; chains align in the direction of the resulting flow. This thinning behavior has been reported for several mucilages, gums, and modified starch (Koocheki et al., 2009; León-Martínez et al., 2011; Li et al., 2009; Liaotrakoon et al., 2011; Medina-Torres et al., 2000; Simas-Tosin et al., 2010; Sopade et al., 2008). In addition, reconstituted mucilage has a water content greater than 80%, which increases the lubricating effect between particles. The molecules of a biopolymer in dispersion undergo conformation rearrangement as a result of deformation (Chuah, Lin, Ling, Choong, & Fakhru'l-Razi, 2008). Other factors that influence the mechanical properties of flow for biopolymers are concentration, molecular weight, degree of branching, electrostatic charges, and temperature (Barnes, 2000; Li et al., 2009). An increase in the concentration of the solutions from 3% to 6% (w/v) increases the solids content and number of molecules that intensify intermolecular interactions, thus increasing viscosity (Li et al., 2009; Sopade et al., 2008). At high concentrations, the molecules of polysaccharides entangle and disentangle after a period of time; this network is stabilized by the intermolecular associations of hydrogen bonding and non-covalent interactions (Van derWaals interactions, electrostatic forces). Viscous response in this study depended on concentration in the studied shear rate range (from 1 to 600 s^{-1}) (Fig. 2). This suggests a behavior different from that reported by Barnes (2000), who reported a dependency only on concentration at high shear rates, due to the fact that the entanglements present in the configuration of the polymeric chain disappear and the chains slide over each other.

3.4.1. Cross model fit

The Cross model has been used to represent macromolecular solutions of polysaccharides in random coil configuration, such as galactomannans and other non-gelling polysaccharides including dextrans, λ-carrageenans, and cellulose derivatives (Morris et al., 1981). Random coil configuration has also been reported for *Ofi* mucilage (Medina-Torres et al., 2000). In the present work, steady-shear viscosity data for pitahaya mucilage solutions were fit to the Cross model ($R^2 > 0.95$); the values of the model parameters are shown in Table 3. In this equation, larger η_0 values indicate higher low-shear-rate viscosity. The parameter λ indicates the characteristic time of the material; that is, the time that the polymer chains in the mucilage solution is aligned, due to the increase in deformation rate (shear rate).

Higher values of λ indicate that the change from the Newtonian zone to the pseudoplastic zone requires a greater amount of time. The magnitude of *m* is related to the flow index; more Newtonian liquids have *m* values tending to zero, whereas more shear-thinning liquids have *m* values tending to unity. Values of η_0 and λ increase with greater concentration, whereas *m* values and the degree of pseudoplasticity decrease. The results of this study pertaining to the effects of concentration on the pseudoplasticity of mucilage agree with those reported for other mucilages and gums (Koocheki et al., 2009; León-Martínez et al., 2011; Medina-Torres et al., 2000).

3.4.2. Weissenberg effect

The recovery of the elastic properties of reconstituted mucilage is manifested through the Weissenberg effect with the increase of the first normal stress difference (N_1) in simple shear flow. Elastic properties are significant for industrial applications; the production of products containing *Ofi* nopal mucilage, such as shampoo and syrups, is based on these properties (Ramírez, 1972). The Weissenberg effect was determined for the T7 treatment, as can be seen in Fig. 3. The results of present work indicate a slight slope change in the N_1 curve for the rehydrated samples at 6%

Table 3
Cross model parameters for reconstituted mucilage at concentrations 3 and 6% (w/v).

Treatment	Model parameters			
	η_0 (Pa s)	λ (s)	*m*	R^2
3% (w/v)				
T1	0.0184	0.0078	0.6644	0.99
T2	0.0221	0.0182	0.3251	0.97
T3	0.0166	0.0094	0.4680	0.99
T4	0.0048	0.0028	0.5873	0.99
T5	0.0062	0.0021	0.9151	0.98
T6	0.0192	0.0226	0.5523	0.99
T7	0.0143	0.0072	0.5542	0.99
T8	0.0131	0.0058	0.8019	0.99
C1	0.0068	0.0018	0.6741	0.97
C2	0.0057	0.0010	0.5786	0.99
6% (w/v)				
T1	0.0574	0.0149	0.5640	0.99
T2	0.0739	0.0340	0.4728	0.99
T3	0.1320	0.0600	0.6449	0.99
T4	0.0113	0.0035	0.7788	0.99
T5	0.0114	0.0012	0.4577	0.96
T6	0.0623	0.0165	0.4941	0.98
T7	0.0598	0.0194	0.5227	0.98
T8	0.0366	0.0119	0.7497	0.99
C1	0.0239	0.0242	0.5844	0.99
C2	0.0250	0.0221	0.5085	0.99

(w/v) concentration, demonstrating a greater interaction between macromolecules and the medium, which is to say, a "pseudosolid-like" behavior (Calderas, Sánchez-Solís, Maciel, & Manero, 2009; González-Aguirre et al., 2009; Manero, Bautista, Soltero, & Puig, 2002; Medina-Torres, Calderas, Gallegos-Infante, González-Laredo, & Rocha-Guzmán, 2009).

3.5. Rheological behavior in oscillatory shear flow

The linear zone was determined in order to evaluate viscoelastic properties at a deformation of 40%. The response of the sample prepared with treatment T7, reconstituted at 3% and 6% (w/v), to oscillatory shear as a function of frequency and concentration is shown in Fig. 4. The viscous component G'' predominated over the elastic component G' ($G'' > G'$); both depend on frequency. Similar behavior was found for all treatments (data not shown). This behavior suggests that the material does not have gel-forming tendencies, which is in concordance with the results found by Goycoolea & Cardenas (2003). The mechanical response shown in Fig. 4 is characteristic of an entangled network of disordered polymer coils (Morris et al., 1981). These results confirm what was

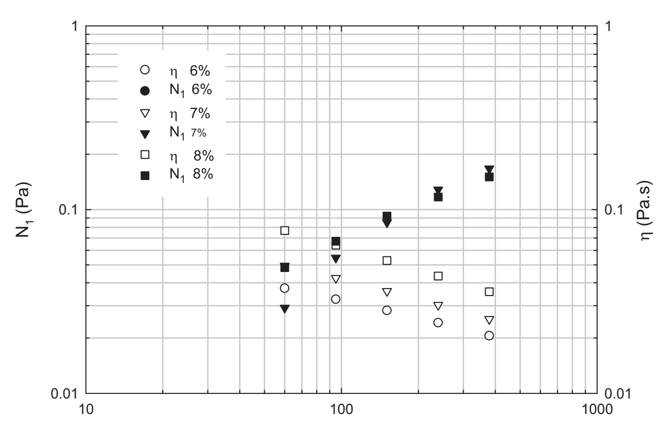

Fig. 3. First normal stress difference and viscosity as a function of shear rate of sample T7 reconstituted.

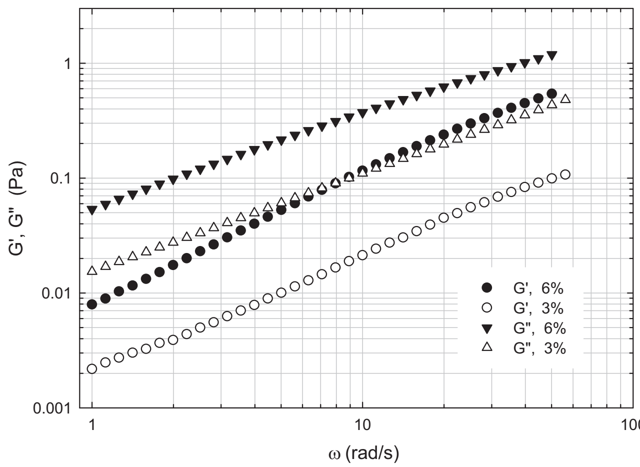

Fig. 4. Mechanical spectra of aqueous solutions of pitahaya mucilage at concentrations of 3% and 6% (w/v) of treatment T7. ○● Dynamic elastic modulus, G′; ▼△ dynamic viscous modulus, G″.

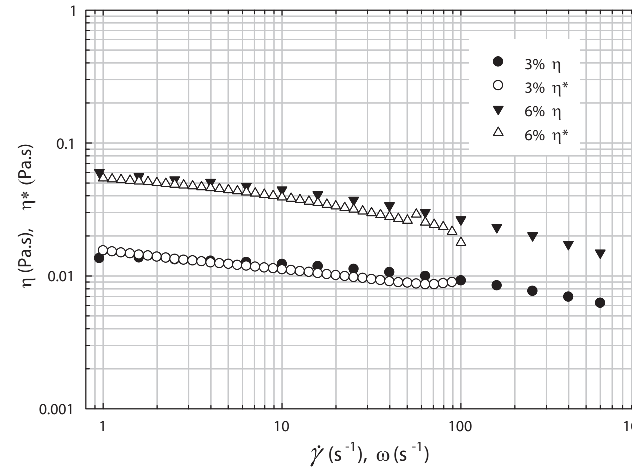

Fig. 5. Cox–Merz rule applied to pitahaya mucilage solutions at 3% and 6% (w/v) of sample T7.

suggested by the deformation value (40%) found during the strain test. This viscoelastic behavior has also been found for *Ofi* gum and mucilage (León-Martínez et al., 2010; Medina-Torres et al., 2000). An increase in concentration increases the magnitude of viscoelastic properties, due to the presence of more points of union between polymer chains; this, in turn, contributes to a greater number of intermolecular impacts (Simas-Tosin et al., 2010). The results of present work indicate a slight slope change in the G' curve for the rehydrated samples at 6% concentration, demonstrating a greater interaction between macromolecules and the medium, which is to say, a pseudosolid-like behavior.

The complex modulus G^* and loss tangent $\tan\delta$ as a function of frequency for all treatments (figure not shown), presented a inverse relationship between $\tan\delta$ and the G^* modulus suggests a greater contribution on the part of the elastic element (T1 and T2) to viscoelastic properties. Tan δ, which is a useful measurement of energy dissipation, was less at lower frequencies, indicating a tendency towards a more pseudosolid-like behavior (Barnes, 2000). These results indicate that differences in viscoelastic behavior depend on the drying treatment, and that it is possible to control the characteristics of the dried materials in keeping with their desired application through manipulation of spray-drying conditions (Gharsallaoui et al., 2007).

3.6. *Effects of the drying conditions on rheological properties*

The effects of Ti on the Cross model viscosity parameters, low shear rate limit viscosity (η_0) and flow index (m) (figure not shown) demonstrate that the variable Ti principally affected the viscous response of the reconstituted hydrocolloid. Mucilage dried at 130 °C displayed a slightly lower viscosity η_0 and higher m values than mucilage dried at 170 °C. This behavior indicates that the samples dried at 130 °C are more pseudoplastic in nature. This can be attributed to the fact that exposure to lower temperatures results in a lesser amount of thermal degradation. Higher temperatures cause pectin-like substances and long-chain carbohydrates to hydrolyze and degrade, resulting in small chains that affect the colloidal properties of the solution, leading to a decreased viscosity (Abu-Jdayil, Banat, Jumah, Al-Asheh, & Hammad, 2004). The effects of Ti on viscosity may be related to the moisture content of the powders. If Ti is low, residual moisture is greater, which contributes to the plastification effect of the mucilage molecules. As has an inverse effect on the viscous response of reconstituted mucilage (data not shown). The inverse effect of Ti and As on the viscosity of reconstituted spray-dried *Ofi* mucilage has been reported by León-Martínez et al. (2011).

The effects of drying conditions on viscoelastic properties were similar to their effects on viscous response (data not shown). In general, the values of the elastic (G') and viscous (G'') components were lower in samples tried at a Ti of 170 °C. An inverse effect was found between As and the magnitudes of G' and G''. The effects of Ti and As were principally observed in the storage modulus G'. This proves that the elastic properties, and thus structure, of the biopolymer are the most affected during the drying process.

3.7. *Comparison of apparent viscosity with complex dynamic viscosity*

Complex (linear) viscosity obtained during the dynamic test (η^*) and apparent (non-linear) viscosity obtained during the shear flow test (η) for mucilage in the T7 sample at concentrations of 3% and 6% (w/v) are presented in Fig. 5 as functions of frequency and deformation velocity ($\omega = \dot{\gamma}$), respectively. The viscosity values were correlated using the Cox–Merz rule, which establishes that η^* as a function of frequency (ω) is almost identical to η as a function of shear rate ($\dot{\gamma}$) (Cox & Merz, 1958). No deviations between η^* and η are observable in Fig. 5. This indicates that similar molecular rearrangements may occur in the two flow patterns over the applied shear rate or frequency range (Gunasekaran & Mehmet, 2000; Xu, Willfor, Holmlund, & Holmbom, 2009). The Cox–Merz rule is valid for synthetic and biopolymer dispersions (da Silva & Rao, 1992), dextran solutions, starch solutions, polysaccharide solutions, locust bean gum, and low methoxil pectin (Morris et al., 1981). Although León-Martínez et al. (2011) reported a failure of *Ofi* mucilage to adhere to the Cox–Merz rule, in that η^* was greater than η, the present work demonstrates that the rule is appropriate for pitahaya mucilage.

3.8. *Spray-drying optimization*

Surface response methodology was applied in order to optimize spray-drying conditions based on drying yield (Y1) and the viscosity of the reconstituted mucilage. The following Cross model parameters were used: low shear rate limit viscosity, η_0 (Y2), characteristic time, k (Y3), and flow index, m (Y4). The desirability function approach is a method that assigns a point value (between 0 and 1) in order to group responses and select the factors that maximize the point value. A value of 1 represents a completely desirable

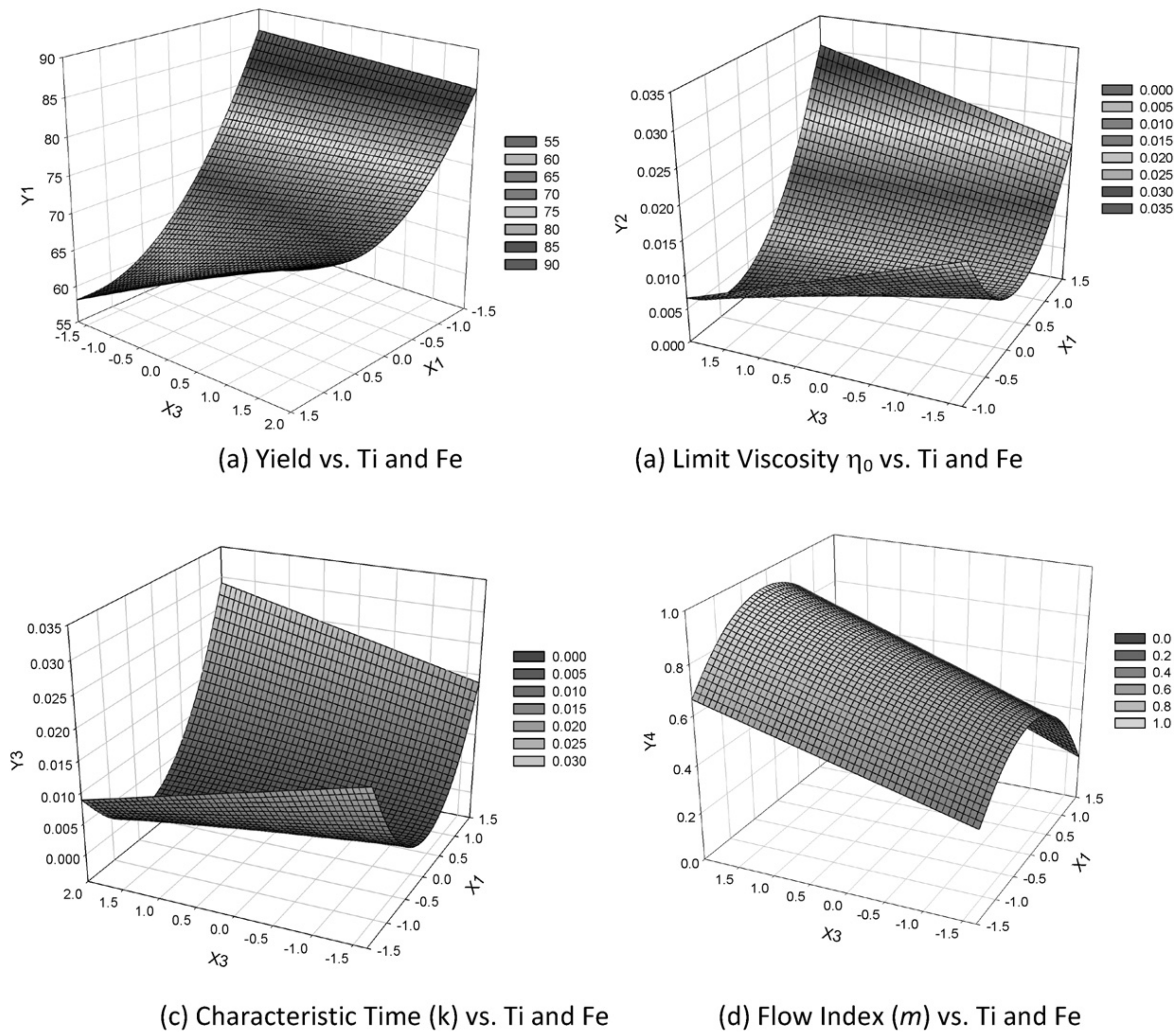

Fig. 6. Surface response of the variables, response vs. Ti (X1) and Fe (X3): (a) yield (Y1); (b) low shear rate limit viscosity, η_0 (Y2); (c) characteristic time, k (Y3); (d) flow index, m (Y4).

value, indicating in this case a 100% drying yield. The independent variables were denoted as X1 (Ti), X2 (As), and X3 (Fe).

A non-linear multiple regression analysis was performed on the experimental data; the estimated coefficients are presented in Table 4. ANOVA indicates a lack of fit of the second order model coefficients to the experimental data.

Fig. 6 shows the surface response graphics for the variables Yield (Y1), η_0 (Y2), k (Y3) and m (Y4) as a function of the independent variables Ti (X1) and Fe (X3), with As (X2) remaining constant.

Table 4
Coefficients estimated of the Second Order Regression Model for optimizing the drying.

Variable	Yield	$\eta_0 \times 10^3$	$k \times 10^3$	M
Constant	67.45	6.03	1.80	0.738
Ti	−7.00	0.00	2.87	−0.042
As	2.81	−2.13	−3.18	−0.006
Fe	1.80	−1.15	0.07	0.097
Ti^2	3.60	8.30	7.69	−0.130
Ti × As	0.39	−3.71	−4.87	0.134
Ti × Fe	1.57	2.49	1.92	0.013
As × Fe	2.29	2.64	0.28	−0.022
R^2	67.00	89.30	61.60	73.200

Lower levels of Ti and Fe produced the greatest yields, as can be seen in Fig. 6a. The lower levels of inlet drying air temperature caused a minimal thermal degradation of the mucilage; the increase in feed flow suggests that the fluid being fed in has a short time of residency, reducing drying time and thus further decreasing degradation. The principle effect was linear or first order; the quadratic effect was not significant, indicating that the interactions between the individual variables did not have a significant influence on yield. The model's negative sign coefficient values between As and yield suggest that a greater yield is obtained at lower As. Furthermore, the positive correlations indicate that the relationships between the variables are direct (Oomah & Mazza, 2001). The behavior of the surface response of graphics displayed increases in yield; however, the maximum points could not be observed in the experimental zone.

Viscous response was analyzed with the following Cross model parameter values: low shear rate limit viscosity, characteristic time, and flow index. The mucilage displayed greater η_0 with increases in Ti and Fe. Oomah and Mazza (2001) reported a negative correlation between yield and rheological properties, indicating that optimization for a high yield leads to the production of low viscosity gums. However, this tendency was not observed in the present study, as optimization for high yields produced final products with acceptable rheological properties.

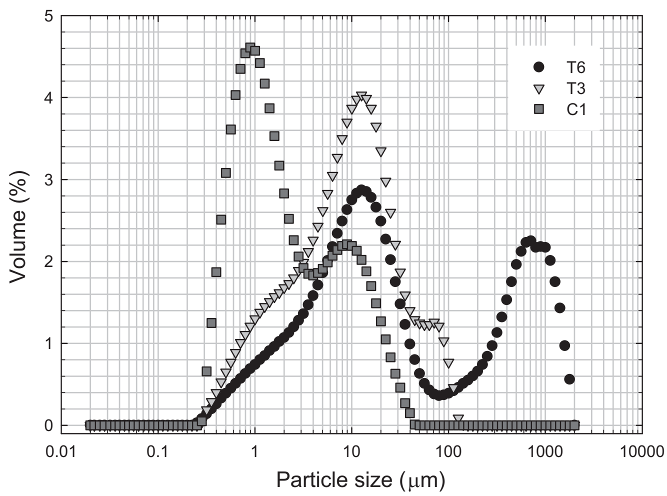

Fig. 7. DTP of reconstituted pitahaya mucilage at 6% (w/v).

Characteristic time is an estimation of the amount of response time needed for the fluid to deform; this parameter was found to increase with higher levels of Ti and Fe. The surface response graphic in Fig. 6d shows how greater flow index values were obtained with central Ti values and high Fe values. This reveals the thinning or pseudoplastic nature of the reconstituted pitahaya mucilage samples, as *m* tends towards unity in the Cross model, describing a greater tendency towards a shear thinning fluid.

3.9. *Particle size distribution (PSD)*

As mentioned in Section 3.1, rheology is influenced by parameters such as particle size and size distribution. In general, the 6% (w/v) reconstituted pitahaya mucilage samples exhibited a quasi-modal distribution and a particle size smaller than 20 μm, indicating that the samples have a low degree of polydispersity and are constituted of very fine particles. Particle size distribution for the T3 and T6 treatments, as well as the central point (C1), are shown in Fig. 7; the highest degree of polydispersity was found in the T6 and C samples. The presence of very small particles facilitates a greater number of particle–particle interactions, increasing resistance to flow; this is most visible at high shear rates (Hill & Carrington, 2006). Servais, Jones, & Roberts (2002) found that a multimodal DTP considerably reduces viscosity in suspensions. The particle size of the pitahaya mucilage powders was smaller than that of *Ofi* mucilage, as reported by León-Martínez et al. (2010). In addition, the pitahaya mucilage powders had a more homogenous distribution, confirming that the mucilage studied in this work has a higher viscosity and stability than *Ofi* mucilage. Increasing drying Ti caused an increase in particle size; this agrees with the results found by Chegini and Ghobadian (2007) for orange juice powder, and Gharsallaoui et al. (2007), who suggested that drying temperature and moisture content are related to changes in particle size and morphology.

4. Conclusions

The physical properties, in particular the rheological properties, of pitahaya (*H. undatus*) mucilage have not been previous studied. The present work demonstrates that this mucilage is an important potential source of hydrocolloids. In addition, the results of this study demonstrate that spray drying is an appropriate method for prolonging the shelf life of this mucilage and preserving its properties. Analysis of processing variables and functional properties provided precise information applicable in the identification of potential industrial applications for this polysaccharide. Inlet air temperature was found to be the variable that most significantly affected the yield of the spray-drying process. A further significant finding was the superior yield obtained for pitahaya mucilage, in comparison with yields obtained for mucilages of other species of the Cactacea family.

Storage stability at room temperature is of great importance in polysaccharides. The low moisture content and glass transition temperature of this polysaccharide suggest that the state of the powdered mucilage is glassy, assuring its stability.

The shear-thinning behavior of reconstituted pitahaya mucilage was found to be stable in flow; this was confirmed through application of the Cox–Merz rule. Non-Newtonian behavior was described with the Cross model. The elastic properties of this mucilage were the most affected during the drying process (G'). In addition, reconstituted pitahaya mucilage exhibited a viscoelastic liquid tendency, and as such can be concluded not to have the capacity to form gels, but rather only to increase in viscosity. The rheological properties of the material were affected by moisture content, concentration, and particle size distribution and shape. These results suggest a random coil structure for the polysaccharide. The flow properties studied revealed that spray-dried pitahaya mucilage has potential for applications as a thickening agent.

Finally, yield and rheological properties were found to be directly linked to drying variables. Optimization of drying conditions achieved a powder yield of up to 85%, and the resulting reconstituted mucilage retained rheological properties suitable for its use in foodstuffs. Manipulation of operating conditions allows specific characteristics of interest to be attained in the final product; as such, further studies on this topic are needed.

References

Abadio, F. D. B., Domingues, A. M., Borges, S. V., & Oliveira, V. M. (2004). Physical properties of powdered pineapple (*Ananas comosus*) juice-effect of malt dextrin concentration and atomization speed. *Journal of Food Engineering*, *64*, 285–287.

Abu-Jdayil, B., Banat, F., Jumah, R., Al-Asheh, S., & Hammad, S. (2004). A comparative study of rheological characteristics of tomato paste and tomato powder solutions. *International Journal of Food Properties*, *7*(3), 483–497.

Association of Official Analytical Chemist (A.O.A.C.). (1984). Edited by Sidney Williams, 14 editions, USA.

Barnes Howard, A. (2000). *A handbook of elementary rheology*. University of Wales.

Bhandari, B. R., Datta, N., & Howes, T. (1997). Problems associated with spray drying of sugar-rich foods. *Drying Technology*, *15*(2), 671–684.

Busso, C. C., Schebor, C., Zamora, C. M., & Chirife, J. (2007). Glass transition temperatures and some physical and sensory changes in stored spray-dried encapsulated flavors. *LWT – Food Science and Technology*, *40*, 1792–1797.

Calderas, F., Sánchez-Solís, A., Maciel, A., & Manero, O. (2009). The transient flow of the PEN-Montmorillonite clay nanocomposite. *Macromolecular Symposia*, *283–284*, 354–360.

Chegini, G. R., & Ghobadian, B. (2007). Spray dryer parameters for fruit juice drying. *World Journal of Agricultural Sciences*, *3*(2), 230–236.

Chin, N. L., Chan, S. M., Yusof, Y. A., Chuah, T. G., & Talib, R. A. (2009). Modelling of rheological behaviour of pummelo juice concentrates using master-curve. *Journal of Food Engineering*, *93*, 134–140.

Chiou, D., & Langrish, T. A. G. (2007). Development and characterisation of novel nutraceuticals with spray drying technology. *Journal of Food Engineering*, *82*, 84–91.

Chuah, G. T., Lin, L. H., Ling, C. N., Choong, T. S. Y., & Fakhru'l-Razi, A. (2008). Effects of temperatures on rheological behavior of dragon fruit (Hylocereus sp.) juice. *International Journal of Food Engineering*, *4*(7), art. no. 4.

Cox, W. P., & Merz, E. H. (1958). Correlation of dynamic and steady flow viscosities. *Journal of Polymer Science*, *28*, 619–622.

da Silva, J. A. L., & Rao, M. A. (1992). Viscoelastic properties of food hydrocolloid dispersions. In M. A. Rao, & J. F. Steffe (Eds.), *Viscoelastic properties of foods* (pp. 285–315). Barking, England: Elsevier Applied Science.

Gharsallaoui, A., Roudaut, G., Chambin, O., Voilley, A., & Saurel, R. (2007). Applications of spray-drying in microencapsulation of food ingredients: An overview. *Food Research International*, *40*, 1107–1121.

González-Aguirre, P., Medina-Torres, L., Schrauwen, C., Fonteix, C., Pla, F., & Herrera-Nájera, R. (2009). Morphology and rheological behavior of maltene-polymer blends. I. Effect of partial hydrogenation of poly(styrene-block-butadiene-block-styrene-block)-type copolymers. *Journal of Applied Polymer Science*, *112*, 1330–1344.

Goula, A., & Adamopoulos, K. G. (2005). Spray drying of tomato pulp in dehumidified air. I. The effect on product recovery. *Journal of Food Engineering*, *66*, 25–34.

Goula Athanasia, M., & Adamopoulos Konstantinos, G. (2010). A new technique for spray drying orange juice concentrate. *Innovative Food Science and Emerging Technologies*, *11*, 342–351.

Goycoolea, F. M., & Cardenas, A. (2003). Pectins from *Opuntia* spp.: A short review. *Journal of the Professional Association for Cactus Development*, *5*, 17–29.

Gunasekaran, S., & Mehmet, Ak. M. (2000). Dynamic oscillatory shear testing of foods-selected applications. *Trends in Food Science & Technology*, *11*, 115–127.

Hill, A., & Carrington, S. (2006). *Understanding the links between rheology and particle parameters*. American Laboratory News. Malvern Instruments Ltd.

Kasapis, Stefan. (2005). Glass transition phenomena in dehydrated model systems and foods: A review. *Drying Technology*, *23*, 731–757.

Kirkwood, D. H., & Ward, P. J. (2008). Comment on the power law in rheological equations. *Materials Letters*, *62*(24), 3981–3983.

Koocheki, A., Mortazavi, S. A., Shahidi, F., Razavi, S. M. A., & Taherian, A. R. (2009). Rheological properties of mucilage extracted from *Alyssum homolocarpum* seed as a new source of thickening agent. *Journal of Food Engineering*, *91*, 490–496.

León-Martínez, F. M., Rodríguez-Ramírez, J., Medina-Torres, L., Méndez Lagunas, L. L., & Bernad-Bernad, M. J. (2011). Effects of drying conditions on the rheological properties of reconstituted mucilage solutions (*Opuntia ficus-indica*). *Carbohydrate Polymers*, *84*, 439–445.

León-Martínez, F. M., Méndez-Lagunas, L. L., & Rodríguez, R. J. (2010). Spray drying of nopal mucilage (*Opuntia ficus-indica*): Effects on powder properties and characterization. *Carbohydrate Polymers*, *81*, 864–870.

Li, X., Fang, Y., Saphwan, A., Phillips, G. O., Nishinari, K., & Zhang, H. (2009). Rheological study of gum arabic solutions: Interpretation based on molecular self-association. *Food Hydrocolloids*, *23*, 2394–2402.

Liaotrakoon, W., De Clercq, N., Van Hoed, V., Van de Walle, D., Lewille, B., & Dewettinck, K. (2011). Impact of thermal treatment on physicochemical, antioxidative and rheological properties of white-flesh and red flesh dragon fruit (*Hylocereus* spp.) Purees. *Food and Bioprocess Technology*, 1–15. http://dx.doi.org/10.1007/s11947-011-0722-4 online version, 12 November 2011

Manero, O., Bautista, F., Soltero, J. F. A., & Puig, J. E. (2002). Dynamics of worm-like micelles: The Cox–Merz rule. *Journal of NonNewtonian Fluid Mechanics*, *106*, 1–15.

Medina-Torres, L., Brito-De La Fuente, E., Torrestiana Sánchez, B., & Katthain, R. (2000). Rheological properties of the mucilage gum (*Opuntia ficus indica*). *Food Hydrocolloids*, *14*, 417–424.

Medina-Torres, L., Calderas, F., Gallegos-Infante, J. A., González-Laredo, R. F., & Rocha-Guzmán, N. (2009). Stability of alcoholic emulsions containing different caseinates as a function of temperature and storage time. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, *352*(1-3), 38–46.

Morris, E. R., Cutler, A. N., Ross-Murphy, S. B., Rees, D. A., & Price, J. (1981). Concentration and shear rate dependence of viscosity in random coil polysaccharide solutions. *Carbohydrate Polymers*, *1*, 5–21.

Nindo, C. I., Tang, J., Powers, J. R., & Takhar, P. S. (2007). Rheological properties of blueberry puree for processing applications. *LWT – Food Science and Technology*, *40*, 292–299.

Obón, J. M., Castellar, M. R., Alacid, M., & Fernández-López, J. A. (2009). Production of a red–purple food colorant from *Opuntia stricta* fruits by spray drying and its application in food model systems. *Journal of Food Engineering*, *90*, 471–479.

Oomah, B. D., & Mazza, G. (2001). Optimization of a spray drying process for flaxseed gum. *International Journal of Food Science and Technology*, *36*, 135–143.

Ramírez, M. E. (1972). Cultivo, explotación y aprovechamiento del Nopal. Mundo Científico. I.P.N. COFFA. SEDICT. No. 4, 10–14.

Rodríguez-Hernández, G. R., González-García, R., Grajales-Lagunés, A., & Ruiz-Cabrera, M. A. (2005). Spray-drying of cactus pear juice (*Opuntia streptacantha*): Effect on the pysicochemical properties of powder and reconstituted product. *Drying Technology*, *23*, 955–973.

Sáenz, C., Sepúlveda, E., & Matsuhiro, B. (2004). *Opuntia* spp. mucilages: A functional component with industrial perspectives. *Journal of Arid Environments*, *57*, 275–290.

Sáenz, C., Tapia, S., Chávez, & Robert, P. (2009). Microencapsulation by spray drying of bioactive compounds from cactus pear (*Opuntia ficus-indica*). *Food Chemistry*, *114*, 616–622.

Servais, C., Jones, R., & Roberts, I. (2002). The influence of particle size distribution on the processing of food. *Journal of Food Engineering*, *51*, 201–208.

Simas-Tosin, F. F., Barraza, R. R., Petkowicz, C. L. O., Silveira, J. L. M., Sassaki, G. L., Santos, E. M. R., et al. (2010). Rheological and structural characteristics of peach tree gum exudates. *Food Hydrocolloids*, *24*, 486–493.

Sopade, P. A., Halley, P. J., Cichero, J. A. Y., Ward, L. C., Hui, L. S., & Teo, K. H. (2008). Rheological characterization of food thickeners marketed in Australia in various media for the management of dysphagia. II. Milk as a dispersing medium. *Journal of Food Engineering*, *84*, 553–562.

Truong, V., Bhandari, B. R., & Howes, T. (2005). Optimization of cocurrent spray drying process for sugar-rich foods. Part II – Optimization of spray drying process based on glass transition concept. *Journal of Food Engineering*, *71*, 66–72.

Walton, D. E., & Mumford, C. J. (1999). Spray dried products-characterization of particle morphology. *Institution of Chemical Engineers*, *77*, 21–38.

Wang, Y., Li, D., Wang, L.-J., Li, S.-J., & Adhikari, B. (2010). Effects of drying methods on the functional properties of flaxseed gum powders. *Carbohydrate Polymers*, *81*, 128–133.

Xu, C., Willfor, S., Holmlund, P., & Holmbom, B. (2009). Rheological properties of water-soluble spruce O-acetyl galactoglucomannans. *Carbohydrate Polymers*, *75*, 498–504.