

# Rheological study of xanthan and locust bean gum interaction in dilute solution: Effect of salt

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## Abstract

An oscillatory capillary rheometer was used to investigate the effects of NaCl, KCl, and CaCl<sub>2</sub> on visco-elastic properties of xanthan and locust bean gum (LBG) blends in dilute solution. Gums were evaluated for intrinsic viscosity and elastic component. Molecular conformation of the xanthan–LBG complex was assessed by the power-law and Huggins equations. Addition of any of the three salts reduced significantly the intrinsic viscosity and elastic component of the gum blends, with a pronounced effect from divalent ions, compared with monovalent ions. The 60% xanthan–40% LBG blend exhibited the strongest attraction between xanthan and LBG. For the three salts, the attraction weakened when 5-mM salt was added and vanished with the addition of 50-mM salt. The strongest attraction between xanthan and LBG molecules was also evidenced by a positive Huggins miscibility coefficient  $K_m$ , and a positive attraction–repulsion coefficient  $\alpha$ . With addition of 50 mM of any of the three salts, the coefficient  $\alpha$  became negative, suggesting a strong repulsion between the two gums. The power-law coefficient  $b$  increased as salt concentration and LBG fraction increased in the blends for the three salts, suggesting a more flexible xanthan–LBG complex dependent on salt concentrations and LBG.

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## 1. Introduction

Xanthan gum is a polysaccharide produced by fermentation of the microbe *Xanthomonas campestris*, and has been very widely studied by both chemical and physical techniques (Richardson & Ross-Murphy, 1987). Xanthan

gum is composed of a 1-4-linked- $\beta$ -D-glucose backbone substituted with a trisaccharide side chain that is composed of two mannoses and one glucuronic acid at every second residue (Nussinovitch, 1997). The side chains bind to the helical backbone and stabilize the helical structure (Moorhouse, Walkinshaw, & Arnott, 1977), thus making the molecule a rather stiff rod with extraordinary stability to heat, acid, and alkali (Whistler & BeMiller, 1997). Greater viscosity and pseudoplasticity result from xanthan gum solutions due to formation of high-molecular-weight aggregates of stiff rod molecules (Whistler & BeMiller, 1997). Solutions of xanthan gum are one of the most intensively studied polysaccharide systems, both in terms of rheological properties and physical chemistry (Rodd, Dunstan, & Boger, 2000). Xanthan macromolecules in the ordered (helical) structure would stiffen the polysaccharide (Milas & Rinaudo, 1979), and this makes xanthan the stiffest

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natural biopolymer (Coviello, Kajiwaru, Burchard, Dentini, & Crescenzi, 1986). The shape of the xanthan molecule in solution depends on the molecular weight of the polysaccharide. Xanthan is reported to behave as a rigid rod at low molecular weights, whereas the shape would be a stiff wormlike coil at high molecular weight (Morris, 1995; Sato et al., 1984).

Locust bean gum (LBG), or carob gum, is the refined endosperm of the seed of the carob tree (*Ceratonia siliqua*) that grows most extensively in Spain and in other Mediterranean countries (Maier, Anderson, Karl, & Magnuson, 1993). Locust bean gum is a galactomannan composed of a 1-4-linked- $\beta$ -D-mannan backbone with 1-6-linked- $\alpha$ -D-galactose side groups (Dea & Morrison, 1975). Physico-chemical properties of galactomannans are strongly influenced by the galactose content (Morris, 1990) and the distribution of the galactose units along the main chain (Launay, Doublier, & Cuvelier, 1986). Longer galactose side chains yield stronger synergistic interactions with other polymers (Morris, 1990) and result in greater functionality (Launay et al., 1986).

Xanthan gum interacts with galactomannans to form mixed gels with high viscosity at low-total-polysaccharide concentrations (Tako, Asato, & Nakamura, 1984), and this interaction is more pronounced with LBG than with any other polysaccharide or galactomannan (Dea, Morris, Rees, & Welsh, 1977). The interaction between xanthan and LBG is largely exploited in food applications in which thickening or gelling is desired.

Considerable work has been published to elucidate the mechanisms behind xanthan and LBG interaction (Cairns, Miles, & Morris, 1986; Cairns, Miles, Morris, & Brownsey, 1987; Tako et al., 1984; Wang, Wang, & Sun, 2002; Williams, Clegg, Day, Phillips, & Nishinari, 1991). Physical and physicochemical techniques have been used to study xanthan–galactomannans interaction, and results demonstrated the existence of an “order (helix)–disorder (coil)” transition for xanthan; this transition responds to changes in ionic strength and temperature, and may play a major role in such interactions (Morris, 1995a). High temperatures favor the “disordered” transition, whereas high ionic strength favors the “ordered” transition (Morris, 1995a). Lopes, Andrade, Milas, and Rinaudo (1992) studied the interaction of xanthan and guar gum at low temperature in water and 20 mM NaCl by using viscosity methods. The authors noticed a small synergistic effect between the two gums in 20 mM NaCl; the effect became more pronounced in water. They concluded that xanthan adopted a disordered conformation in water, whereas the conformation was in an ordered form in 20 mM NaCl. These findings were supported by Dalbe (1992); he used small-deformation oscillation methods to study xanthan–glucomannan mixture and reported that addition of 85.55 mM NaCl or 67.07 mM KCl to gum mixtures led to a dramatic reduction in gel strength, which was not altered by further addition of electrolytes. Similar results were reported by Wang et al. (2002). The authors used rheological methods to study the conformational role of xanthan in interaction

with LBG, and reported a decrease in viscosity and texture strength for xanthan–LBG solutions and gels, respectively, associated with the addition of 40 mM NaCl. In the absence of electrolytes, the xanthan molecules may self-associate to reduce the interfacial energy. If mannan is present, competition between xanthan self-association and xanthan–mannan occurs, and the later may be favored (Morris, 1995a). In the presence of electrolytes and at lower temperatures, xanthan self-association competes more effectively with xanthan–mannan association because of charge screening and hence, lower viscosity solutions and weaker gels result (Morris, 1995a).

To date, much work has been accomplished on the gelling properties of gums and their inherent visco-elastic properties, as evidenced by many published papers; evaluation of gums in solution has been studied to a lesser extent (Casas & Garcia-Ochoa, 1999; Cuvelier & Launay, 1986), and most of the research was performed by using the Ubbelohde or rotational viscometer. Advancements in capillary rheometry now allow for improved, reproducible observation of the elastic component in a dilute solution, which could not be measured with an Ubbelohde or rotational viscometer (Yaseen, Herald, Aramouni, & Alavi, 2005). The objective of this study was to explore the effect of ionic environment on the interaction of xanthan with LBG in dilute solution. A sensitive oscillatory-rheometry technique was employed to capture this interaction. Different intrinsic-viscosity models and the elastic component for the gum blends were used to determine if any conformational change or interaction occurred between xanthan and LBG in dilute solution.

## 2. Materials and methods

### 2.1. Materials and solutions preparation

Xanthan gum and locust bean gum (LBG) were purchased from Sigma (Sigma–Aldrich, St. Louis, MO). The stock solutions (0.1% m/v) were prepared by dispersing 0.1 g of dry sample in 100 ml of deionized distilled water while continuously stirring at ambient temperature. The solutions were heated for 30 min at 80 °C in a water bath to completely solubilize the gums, then were cooled and immediately dialyzed against deionized distilled water for 72 h with a dialyzing tube (Snakeskin TM Pleated Dialysis Tubing, Pierce, Rockford, IL), which was designed to let through compounds with a molecular weight of 10 kDa or less.

To study the interaction between gums, the following treatments were considered: xanthan 100%, xanthan 60%–LBG 40%, xanthan 40%–LBG 60%, and LBG 100%. Stock solutions were stirred at room temperature after dialysis, diluted with deionized distilled water to a final concentration of 0.025% (m/v), and blended as previously stated. The concentration of each solution was determined by the phenol-sulfuric method (Dubois, Giles, Hamilton, Rebers, & Smith, 1956). To study the effect of

salt on interaction between gums, appropriate amounts of NaCl, KCl, and CaCl<sub>2</sub> were added and completely dissolved to make 5-mM and 50-mM solutions.

## 2.2. Density measurement

The densities of the solutions were determined with a standardized 10-mL pycnometer (Bradly, 1998). The mass of the solution was calculated from the weight difference between the empty pycnometer (Kmax<sup>®</sup>, Kimble Glass Inc., Vineland, NJ) and the filled vessel. The pycnometer filled with each respective gum solution was incubated at 20 °C for 1 h (Equatherm, Lab-Line Instruments Inc., Melrose Park, IL) to equilibrate the sample before density determinations (Yaseen et al., 2005).

## 2.3. Rheological properties

The elastic and viscous components of each gum solution and of the blends were measured as a function of oscillating shear rate by using an oscillating capillary rheometer (Viscoelasticity Analyzer, Vilastic 3, Vilastic Scientific, Inc., Austin, TX), which is specifically engineered to measure the oscillatory viscosity of fluids. The viscoelasticity analyzer is based on the principles of generating oscillatory flow at a selected frequency within a straight, cylindrical, stainless steel tube (0.0504 cm radius and 6.038 cm length).

The fluid in the capillary tube is forced into oscillatory flow at a selected frequency, during which the pressure gradient and the volume flow are monitored. Precise resolution of magnitude and phase of the pressure and volume flow allows calculation of the viscous and elastic components of the shear stress, shear rate, and shear strain at the tube wall (Thurston, 1960, 1996).

The viscosity and elasticity were determined at a frequency of 2 Hz because preliminary work showed reproducible and better results at that frequency. Also, we wanted to compare our results to other published findings, which used the same frequency (Yaseen et al., 2005). The shear rate range was 0.8–20 s<sup>-1</sup>, but data were statistically analyzed at 10 s<sup>-1</sup> (Morris & Taylor, 1982). Rheological measurements were carried out at 20 °C by using a temperature-controlled circulating water bath (Haake DC5, Gebr. Haake GmbH, Karlsruhe, Germany). The viscoelasticity analyzer was calibrated with deionized distilled water at 20 °C before the verification procedure to further ensure that the rheometer was operating at optimum accuracy.

Microsoft Excel 2000 (Microsoft Corporation, Seattle, WA) was used to plot viscosities against concentrations, as well as to obtain linear regression lines with the corresponding equations and correlation coefficients (*R*<sup>2</sup>), to assess the best model.

## 2.4. Intrinsic viscosity determination

The intrinsic viscosity [ $\eta$ ] is a measure of the hydrodynamic volume occupied by a macromolecule, which is clo-

sely related to the size and conformation of the macromolecular chains in a particular solvent (Lai & Chiang, 2002). The intrinsic viscosity [ $\eta$ ] is determined experimentally from measurements of the viscosity of very-low-concentration (*C*) solutions. Denoting solution and solvent viscosity as, respectively,  $\eta_{\text{solution}}$  and  $\eta_{\text{solvent}}$ , [ $\eta$ ] is defined by the following relationships:

$$\text{Relative viscosity: } \eta_{\text{rel}} = \eta_{\text{solution}} / \eta_{\text{solvent}} \quad (1)$$

$$\text{Specific viscosity: } \eta_{\text{sp}} = \eta_{\text{rel}} - 1 \quad (2)$$

$$\text{Intrinsic viscosity: } [\eta] = \lim_{C \rightarrow 0} \frac{\eta_{\text{sp}}}{C} \quad (3)$$

The intrinsic viscosity can be obtained by measuring specific viscosities at different concentrations at the same shear-rate, and extrapolating the course of specific viscosity to infinite dilution (Heitmann & Mersmann, 1995). The intrinsic viscosity [ $\eta$ ] is, therefore, obtained by extrapolating data to zero concentration by using a linear regression, which will be called the graphic double-extrapolation procedure (GDEP) in this study. McMillan (1974) showed that  $\frac{\eta_{\text{sp}}}{C}$ , also called reduced viscosity, could be written in the form of a Huggins equation (Huggins, 1942)

$$\frac{\eta_{\text{sp}}}{C} = [\eta] + k'[\eta]^2 C \quad (4)$$

where *k'* is the Huggins constant. The determination of the intrinsic viscosity is, therefore, the extrapolation of reduced viscosity to the value at zero solute concentration. The extrapolations are usually done in very dilute regimes ( $C \ll C^*$ ) with relative viscosity values between 1.2 and 2.0, the corresponding specific viscosities being between 0.2 and 1.0 (Da Silva & Rao, 1992). *C*<sup>\*</sup> is defined as the overlap concentration, the transition from the dilute to the semi-dilute region which mark the onset of polymer entanglement (Launay, Cuvelier, & Martinez-Reyes, 1997). In the present work, gum solutions were therefore diluted to be within the described range. In addition, McMillan (1974) reported that the intrinsic viscosity could be obtained from the Kraemer equation (Kraemer, 1938) by extrapolation to zero concentration (*C*)

$$\frac{\ln \eta_{\text{rel}}}{C} = [\eta] + k''[\eta]^2 C \quad (5)$$

where *k''* is the Kraemer constant. For very dilute solutions, however, Eq. (5) can be shortened by retaining only the first-order term, and [ $\eta$ ] can be determined from the slope of a plot of *C* against  $\ln \eta_{\text{rel}}$  (Sornsrivichai, 1986). McMillan (1974) showed that methods of determination of the intrinsic viscosity that were based on slopes of plots had higher correlation coefficients and lower standard errors, compared with those based on intercepts of plots. On the basis of such findings, Tanglertpaibul and Rao (1987) used the following equations to obtain the intrinsic viscosity of tomato serum:

$$\eta_{\text{rel}} = 1 + [\eta]C \quad (6)$$

The intrinsic viscosity  $[\eta]$  is the slope obtained by plotting  $\eta_{\text{rel}}$  vs.  $C$

$$\eta_{\text{rel}} = e^{[\eta]C} \quad (7)$$

The intrinsic viscosity  $[\eta]$  is the slope obtained by plotting  $\ln \eta_{\text{rel}}$  vs.  $C$

$$\eta_{\text{rel}} = \frac{1}{1 - [\eta]C} \quad (8)$$

The intrinsic viscosity is the slope obtained by plotting  $1 - \frac{1}{\eta_{\text{rel}}}$  vs.  $C$ .

The intrinsic viscosity  $[\eta]$  was estimated based on the slope of  $\eta_{\text{sp}}$  vs.  $C$  for polyelectrolytes, as suggested by Chou and Kokini (1987); this is similar to the method discussed in Eq. (6). Chou and Kokini (1987) reported that when there is essentially no molecular interaction, as in dilute solutions, the second term of the Huggins equation (Eq. (4)) is negligible, and a plot of  $\eta_{\text{sp}}$  against concentration is linear. In this study, the intrinsic viscosity in the dilute domain was estimated on the basis of Eqs. (3), (6), (7), and (8), and the four methods were statistically compared for a better fit.

### 2.5. Determination of the molecular conformation and polymer interaction

The power-law equation

$$\eta_{\text{sp}} = aC^b \quad (9)$$

was used to estimate the exponent  $b$  from the slope of a double logarithmic plot of  $\eta_{\text{sp}}$  against concentration, and provides an indication of the conformation of polysaccharides (Lai, Tung, & Lin, 2000). To determine xanthan–LBG interaction, the verified theory (Chee, 1990; Sun, Wang, & Feng, 1992), based on the classical Huggins equation expressing the specific viscosity ( $\eta_{\text{sp}}$ ) of a polymer as a function of polymer concentration  $C$ , was used

$$\frac{\eta_{\text{sp}}}{C} = [\eta] + bC \quad (10)$$

and

$$b = K[\eta]^2 \quad (11)$$

where  $[\eta]$  is the intrinsic viscosity, and  $b$  and  $K$  are Huggins parameters. Chou and Kokini (1987) reported that for dilute solutions, the term  $bC$  from Eq. (10) is negligible, and the plot of the specific viscosity against the concentration gives a straight line. The term  $b$  was approximated from the small intercept value of the plot of specific viscosity against the concentration (Higiro, Herald, & Alavi, 2006).

According to this theory, the following equation applies to a ternary polymer–polymer–solvent dilute solution, in which there is no aggregation between molecules

$$\alpha = b_m - \left( \sqrt{b_1}W_1 + \sqrt{b_2}W_2 \right)^2 \quad (12)$$

where  $b_m$ ,  $b_1$ , and  $b_2$  are the Huggins coefficients for blend, polymer 1, and polymer 2, respectively.  $W_i$  is the weight

fraction of polymer  $i$  in the polymer blend ( $i = 1$  or  $2$ ). The equation may be used to provide qualitative information on polymer–polymer interaction: two polymers are attractive in solution when  $\alpha \geq 0$ , whereas they are repulsive when  $\alpha < 0$  (Wang, Sun, & Wang, 2001).

Three types of interaction contribute to the value of  $b_m$  for a ternary polymer–polymer–solvent interaction (Cragg & Bigelow, 1955)

1. Long-range hydrodynamic interaction of pairs of single molecules:

$$b_{m1} = b_1W_1^2 + b_2W_2^2 + 2\sqrt{b_1b_2}W_1W_2 = \left( \sqrt{b_1}W_1 + \sqrt{b_2}W_2 \right)^2 \quad (13)$$

where  $b_i$  is the Huggins parameter  $b$  for component  $i$  ( $i = 1$  or  $2$ ).  $W_i$  is the weight fraction of polymer  $i$  in the blend.

2. The formation of double molecules. This assumes actual contact:

$$b_{m2} = K'([\eta]_2 - [\eta]_1) \quad (14)$$

where  $[\eta]_1$  and  $[\eta]_2$  are intrinsic viscosities of double- and single-molecule species, respectively, and  $K'$  is a constant. In the absence of aggregation between molecules and at sufficiently low concentrations ( $C \ll C^*$ ), this term is neglected.

3. Intermolecular attraction or repulsion:

$$b_{m3} = \alpha \quad (15)$$

where  $\alpha \geq 0$  means attraction, whereas  $\alpha < 0$  means repulsion.

Therefore,

$$b_m = b_{m1} + b_{m2} + b_{m3} \approx b_{m1} + b_{m3} \quad (16)$$

$$b_m = \left( \sqrt{b_1}W_1 + \sqrt{b_2}W_2 \right)^2 + \alpha \quad (17)$$

$$\alpha = b_m - \left( \sqrt{b_1}W_1 + \sqrt{b_2}W_2 \right)^2 \quad (18)$$

By measuring  $b_m$  from the Huggins equation for polymer–polymer–solvent solution,  $\alpha$  was calculated and the interaction was characterized.

### 2.6. Statistical analysis

A two-way factorial design was used to generate the best-fitting intrinsic viscosity model. For each gum blend, the three salts (NaCl, KCl, and NaCl<sub>2</sub>) at three concentrations (0, 5, and 50 mM) were compared for the intrinsic viscosity, the elastic component, the molecular conformation coefficient  $b$ , the miscibility coefficient  $\alpha$ , and the Huggins coefficient  $K$ , in a factorial design. In each instance, three replications were made. The analysis of variance and means comparison were conducted by the general linear models procedure (Proc GLM), with Statistical Analysis System software (version 9.1, SAS Institute, Inc., Cary, NC). Comparisons among treatments were analyzed by using Fisher's least significant difference (LSD), with a significance level at  $P < 0.05$ .

### 3. Results and discussion

#### 3.1. Intrinsic viscosity

The dilute regime ( $C \ll C^*$ ) was obtained by blending the two gums at 0.025 g/dl. This gum concentration (0.025 g/dl) and below corresponded to the Newtonian regime for the whole shear rate range used ( $0\text{--}20\text{ s}^{-1}$ ). Lounay et al. (1997) reported that xanthan at 0.025 g/dl or less corresponded to the Newtonian viscosity. For xanthan and all xanthan–LBG blends, an increase in reduced viscosity was observed as the concentration decreased, regardless of salt type and salt concentration (Figs. 1 and 2). A more pronounced increase was observed for gum concentrations less than 0.008 g/dl. The data did not fit with the linear regression model. Therefore, the GDEP failed to generate the intrinsic viscosity for xanthan gum and xanthan–LBG blends. Lai et al. (2000) reported similar results when determining the intrinsic viscosity of hsian-tiao leaf gum in different salt solutions. Lapasin and Priol (1995) reported that non-ionic polysaccharides (i.e., LBG) exhibited linear plots of lower slope, whereas ionic polysaccharides (i.e., xanthan) displayed a sharp increase in slope, possibly due to expanded coil dimensions and electrostatic repulsion between chain segments. In a study of the rheological properties of sodium pectate at various counter-ion concentrations, Pals and Hermans (1952) reported that the specific viscosity/concentration for sodium pectate increased with a small uniform slope with increasing polymer concentration only in excess of added salts, as is true of non-ionic polysaccharides. As ionic strength decreased, the viscosity increased, and a more complex dependence upon concentration was observed, with the appearance of a maximum, becoming more pronounced and shifting to the left. Without salt addition, the specific viscosity/concentration of sodium pectate increased steadily with dilution, and very rapidly at high dilution. Our results showed an increase

of specific viscosity/concentration for xanthan and xanthan–LBG blends as gum concentration decreased for all salts and salt concentrations, which was consistent with the behavior of ionic polysaccharides.

For LBG, the Huggins and Kraemer plots (Figs. 3 and 4) provided a better fit for all salts and salt concentrations; therefore, the models were selected as the best method for intrinsic viscosity estimation. From Eq. (4), the Huggins constant  $k'$  theoretically should lie between 0.3 and 0.8, and values larger than 1 imply polymer–polymer aggregation (Morris, 1995b). The sum of the Huggins constant  $k'$  and Kraemer constant  $k''$  should equal  $0.5 \pm 10\%$ , and larger or smaller values are attributed to molecule association (Morris, Cutler, Ross-Murphy, & Rees, 1981). Values from this study fall within this range, meaning molecular association was absent.

The non-linear relationship between  $\frac{\eta_{sp}}{C}$  and gum-blend concentration was observed at low xanthan and xanthan–LBG blend concentrations, thus making impossible the determination of the intrinsic viscosity by extrapolation of experimental data; this prompted the use of slope models (Chou & Kokini, 1987; Tanglertpaibul & Rao, 1987) to determine the intrinsic viscosity by plotting relative viscosity vs.  $C$  (Eq. (6)),  $\ln \eta_{rel}$  vs.  $C$  (Eq. (7)), and  $1 - \frac{1}{\eta_{rel}}$  vs.  $C$  (Eq. (8)). Straight-line relationships with large linear regression coefficients were obtained for xanthan and xanthan–LBG blends (Figs. 5–7). McMillan (1974) reported that methods of determination of intrinsic viscosity based on slopes of plots had larger correlation coefficients and smaller standard errors than did those based on intercepts of plots. The values of the intrinsic viscosities resulting from these models differed, but showed similar trends for all salts and salt concentrations. A representative comparison of the different models used to estimate the intrinsic viscosity is shown for xanthan 60%–LBG 40% blend in 50 mM  $\text{CaCl}_2$  (Table 1). Intrinsic viscosity values calculated by using Eq. (6) were larger and significantly different

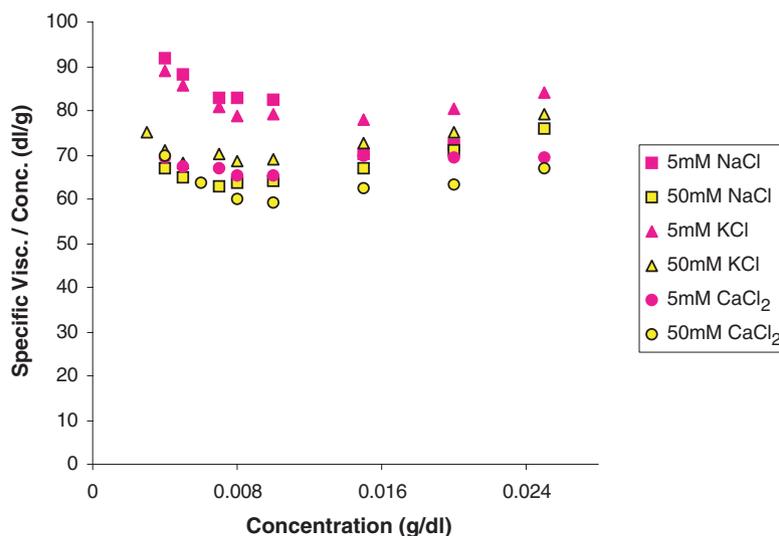


Fig. 1. Huggins plot (specific viscosity/concentration vs. concentration) for xanthan 100% with addition of salts.

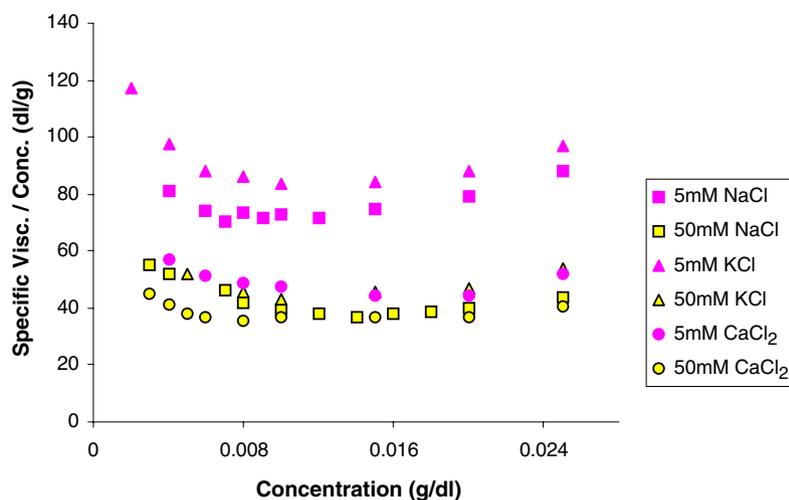


Fig. 2. Huggins plot (specific viscosity/concentration vs. concentration) for xanthan 60%–locust bean gum 40% blend with addition of salts.

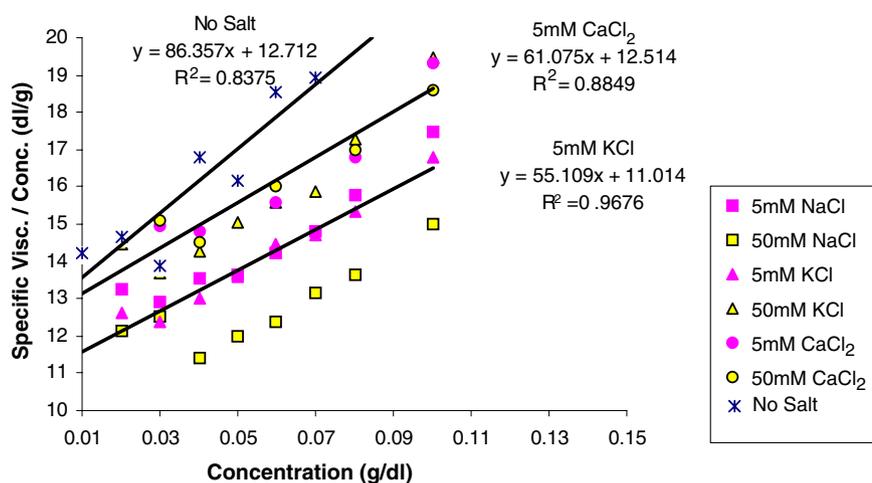


Fig. 3. Huggins plot (specific viscosity/concentration vs. concentration) for locust bean gum with addition of salts.

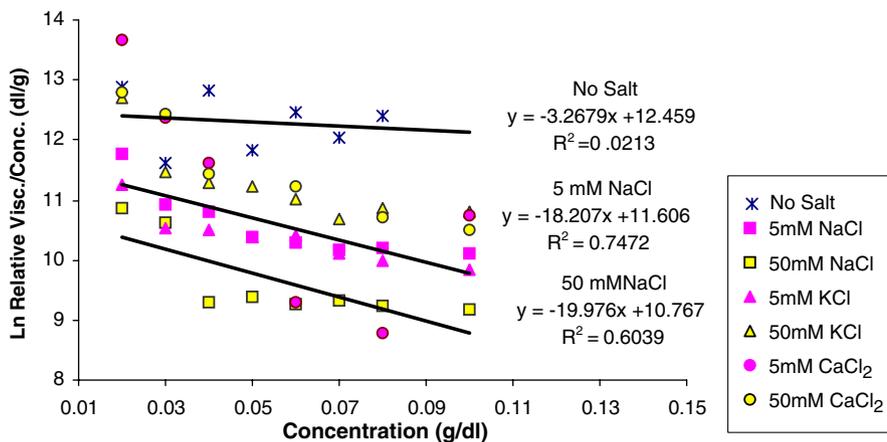


Fig. 4. Kraemer plot (Ln relative viscosity/concentration vs. concentration) for locust bean gum with addition of salts.

from those obtained by using Eqs. (7) and (8) for all salts and salt concentrations. Tanglertpaibul and Rao (1987) successfully implemented the model from Eq. (6) in the

intrinsic viscosity determination of tomato serum. The authors reported higher correlation coefficients and fewer errors. Intrinsic viscosity values obtained in this study for

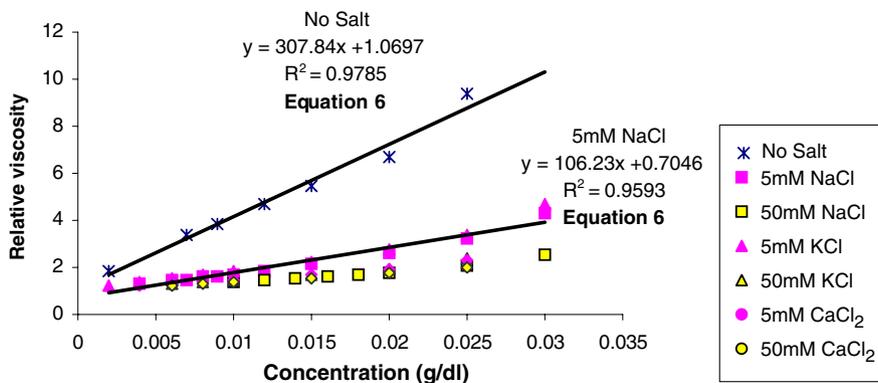


Fig. 5. Relative viscosity for xanthan 60%–locust bean gum 40% blend as a function of gum concentration with addition of salts.

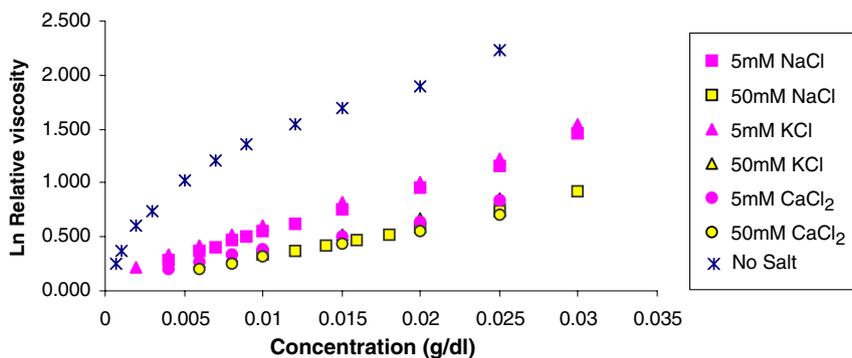


Fig. 6. Ln Relative viscosity for xanthan 60%–locust bean gum 40% blend as a function of gum concentration with addition of salts.

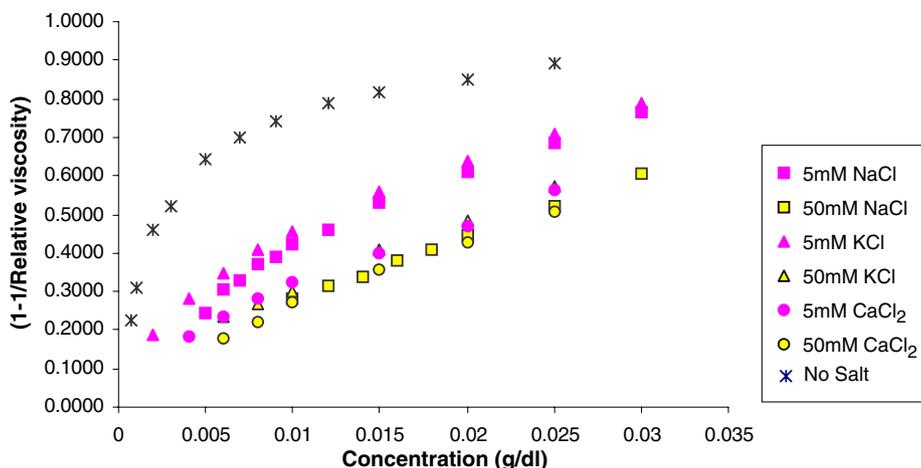


Fig. 7. Plot of  $(1 - 1/\text{Relative viscosity})$  as a function of gum concentration for xanthan 60%–locust bean gum 40% blend with addition of salts.

xanthan alone in any of three salts at 5 and 50 mM by using the Eq. (6), ranged from 77.93 to 59.77 dl/g (Table 2), respectively, and were comparable to those reported by Launay et al. (1997). Significant differences between gum blends were detected with Eq. (6), whereas these differences were few with Eqs. (7) and (8). We used Eq. (6) as the best model for intrinsic viscosity determination because it showed a better linear fit, with higher correlation ( $R^2$ ) for most of the blends, salts, and salt concentrations. Further-

more, the results were similar to other findings (Tanglerpaibul & Rao, 1987).

As shown in Table 2, the hydrodynamic behavior of xanthan and xanthan–LBG blends was strongly affected by ion types and ion concentrations. Within each gum blend, divalent ions from  $\text{CaCl}_2$  showed a more pronounced effect on the intrinsic viscosity, compared with monovalent ions from  $\text{NaCl}$  and  $\text{KCl}$ . The increase in ionic strength of the three salts from 0 to 5 and 50 mM caused a

Table 1  
Intrinsic viscosity values (dl/g) obtained for xanthan, locust bean gum (LBG), and their blends with addition of 50 mM CaCl<sub>2</sub>

Gum	Model				
	Huggins	Kraemer	Tanglertpaibul and Rao		
	Eq. (4)	Eq. (5)	Eq. (6)	Eq. (7)	Eq. (8)
Xanthan 100%	–	–	59.77 <sup>A</sup> <sub>a</sub> ± 0.50	37.74 <sup>B</sup> <sub>a</sub> ± 0.53	23.84 <sup>C</sup> <sub>a</sub> ± 0.18
Xanthan 60%–LBG 40%	–	–	44.86 <sup>A</sup> <sub>b</sub> ± 6.30	31.2 <sup>B</sup> <sub>a</sub> ± 1.73	24.87 <sup>C</sup> <sub>a</sub> ± 1.80
Xanthan 40%–LBG 60%	–	–	33.15 <sup>A</sup> <sub>c</sub> ± 2.10	22.97 <sup>B</sup> <sub>b</sub> ± 1.21	15.74 <sup>C</sup> <sub>b</sub> ± 1.26
LBG 100%	13.01 ± 0.22	13.1 ± 0.10	18.71 <sup>A</sup> <sub>d</sub> ± 1.01	9.96 <sup>B</sup> <sub>c</sub> ± 1.60	5.59 <sup>C</sup> <sub>c</sub> ± 1.24

Results are expressed as means ± SD for three replications.

<sup>A–C</sup> Means followed by the same letters in the same row are not significantly different ( $P < 0.05$ ).

<sup>a–c</sup> Means followed by the same letters in the same column are not significantly different ( $P < 0.05$ ).

–: Not estimated by corresponding equations.

Table 2  
Intrinsic viscosity values for xanthan, locust bean gum (LBG), and their blends with addition of three salts at selected concentrations

Gum blend	Salts			
	Salt concentration (mM)	NaCl (dl/g)	KCl (dl/g)	CaCl <sub>2</sub> (dl/g)
Xanthan 100% <sup>†</sup>	0	214.21 <sub>a</sub> ± 0.2	214.21 <sub>a</sub> ± 0.2	214.21 <sub>a</sub> ± 0.2
	5	75.44 <sup>A</sup> <sub>b</sub> ± 1.1	77.93 <sup>A</sup> <sub>b</sub> ± 2.4	63.08 <sup>B</sup> <sub>b</sub> ± 0.8
	50	68.72 <sup>A</sup> <sub>b</sub> ± 1.7	73.82 <sup>A</sup> <sub>b</sub> ± 1.1	59.77 <sup>B</sup> <sub>c</sub> ± 0.5
Xanthan 60%–LBG 40% <sup>†</sup>	0	306.61 <sub>a</sub> ± 4.3	306.61 <sub>a</sub> ± 4.3	306.61 <sub>a</sub> ± 4.3
	5	106.23 <sup>B</sup> <sub>b</sub> ± 1.6	113.48 <sup>A</sup> <sub>b</sub> ± 0.5	48.30 <sup>C</sup> <sub>b</sub> ± 2.6
	50	55.86 <sup>A</sup> <sub>c</sub> ± 0.7	54.21 <sup>A</sup> <sub>c</sub> ± 2.9	44.86 <sup>B</sup> <sub>b</sub> ± 6.3
Xanthan 40%–LBG 60% <sup>†</sup>	0	216.89 <sub>a</sub> ± 0.6	216.89 <sub>a</sub> ± 0.6	216.89 <sub>a</sub> ± 0.6
	5	76.75 <sup>A</sup> <sub>b</sub> ± 4.4	61.6 <sup>B</sup> <sub>b</sub> ± 5.3	35.71 <sup>C</sup> <sub>b</sub> ± 0.8
	50	37.91 <sup>A</sup> <sub>c</sub> ± 1.3	31.92 <sup>C</sup> <sub>c</sub> ± 2.5	33.15 <sup>B</sup> <sub>b</sub> ± 2.1
LBG 100% <sup>‡</sup>	0	12.49 <sub>a</sub> ± 0.1	12.49 <sub>a</sub> ± 0.1	12.49 <sub>a</sub> ± 0.1
	5	10.36 <sup>C</sup> <sub>b</sub> ± 0.4	11.01 <sup>B</sup> <sub>b</sub> ± 0.1	13.65 <sup>A</sup> <sub>b</sub> ± 0.6
	50	10.75 <sup>B</sup> <sub>b</sub> ± 0.3	11.25 <sup>B</sup> <sub>b</sub> ± 0.2	13.21 <sup>A</sup> <sub>b</sub> ± 0.1

Results are expressed as means ± SD for three replications.

<sup>A–C</sup> Means followed by the same letters in the same row are not significantly different ( $P < 0.05$ ).

<sup>a–c</sup> Means followed by the same letters in the same sub column are not significantly different ( $P < 0.05$ ).

<sup>†</sup> Intrinsic viscosity values estimated with Tanglertpaibul and Rao (1987) equation:  $\eta_{rel} = 1 + [\eta]C$ .

<sup>‡</sup> Intrinsic viscosity values estimated with Huggins equation:  $\frac{\eta_{sp}}{C} = [\eta] + k'[\eta]^2C$ .

significant decrease in intrinsic viscosity for all gum blends. The intrinsic viscosity dropped to less than 50% of the value obtained with dialyzed xanthan and xanthan–LBG blends when only 5 mM salt was added to the gum blends. Similar results were reported elsewhere (Lai & Chiang, 2002; Launay et al., 1997; Wang et al., 2002). Such behavior is to be attributed to the native xanthan. In aqueous solution with no added ions, the xanthan molecule is extended due to electrostatic repulsion of the negatively charged acetyl groups (Carrington, Odell, Fisher, Mitchell, & Hartley, 1996). When salt is added, charge screening causes the side chains to collapse down to the backbone, hence giving the xanthan molecule a rod-like shape and reducing the hydrodynamic volume (Rochefort & Middleman, 1987). The more pronounced effect of the divalent ions (calcium) on the intrinsic viscosity, compared with that of monovalent ions (sodium and potassium), was pos-

sibly due to molecular crosslinking between xanthan and calcium ions, which resulted in a greater extent of molecular contraction (Lai & Chiang, 2002). In contrast, addition of salt did not affect the intrinsic viscosity of LBG, for which values ranged between 10.36 and 13.65 dl/g in aqueous solution and in 5 or 50 mM of salts. Locust bean gum is a non-polyelectrolyte, and salt has little effect on the hydrodynamic volume (Wang et al., 2002).

Results in Table 2 for the three salts show that the intrinsic viscosities of xanthan–LBG blends were greater than those calculated from the weight average of the individual gums at 0 and 5 mM of salt. But the intrinsic viscosity of the blends dropped below those calculated from the weight average of the individual gums when salt content increased to 50 mM. These results may suggest an intermolecular attraction between xanthan and LBG, which was greater for the 60% xanthan and 40% LBG blend, as long

as the salt content was low (0–5 mM). Double molecule formation was unlikely because of the dilute regime ( $C \ll C^*$ ). A similar trend was reported from studies at higher xanthan and LBG concentration in 10 mM KCl, in which the measured viscosities passed through a maximum when the two polymers were present in roughly equal concentrations (Goycoolea, Morris, & Gidley, 1995). From a molecular viewpoint, LBG may have destabilized the xanthan helical structure and rendered a more flexible xanthan chain, thus facilitating formation of junctions with xanthan (Wang et al., 2002; Zhan, Ridout, Brownsey, & Morris, 1993). Whether this synergism is accompanied by a change in molecular weight of both species remains unclear, and is subject to further investigations. The observed drop of the intrinsic viscosity of the blends below the viscosity calculated from weight average of the individual gums when salt concentration increased to 50 mM suggests the absence of synergism between xanthan and LBG molecules at that particular ionic strength. Salt addition stabilizes the xanthan helix structure and hinders xanthan–galactomannan interaction (Foster & Morris, 1994). As a consequence, the xanthan–xanthan interactions are more favored at the expense of xanthan–galactomannan interactions (Dalbe, 1992). Results (Table 2) showed significant differences between salts and salt concentrations in terms of intrinsic viscosity for LBG 100%. A limited, but significant decrease of the intrinsic viscosity was noted from 0 to 5 mM salt for NaCl and KCl, whereas the intrinsic viscosity increased for CaCl<sub>2</sub>. Similar results were reported by Launay et al. (1997). The authors noted a decrease in intrinsic viscosity for LBG from 14.1 to 9.8 dl/g in aqueous and 34.22 mM NaCl, respectively. The authors pointed out that, since these results were stable in the course of the experiment, no depolymerization or debranching occurred by increasing the ionic strength for

LBG. They suggested that the decrease was due to changes in solvent quality. The viscosity of LBG, a non-polyelectrolyte polysaccharide, is less affected by salts (Wang et al., 2002).

### 3.2. Molecular conformation and polymer interaction

The power-law model (Eq. (9)) was used to estimate the exponent  $b$  from the slope of a double logarithmic plot of  $\eta_{sp}$  against concentration (Table 3). Results showed that  $b$  increased with the increase of salt concentration in all gum blends, with significant differences among salt concentrations. Larger  $b$  values were obtained with 50 mM salt, compared with values at 0 and 5 mM. Also, significant differences were found among salt types within each gum blend, with larger values for NaCl and smaller ones for CaCl<sub>2</sub>. Xanthan alone showed smaller  $b$  values, compared with  $b$  values of xanthan–LBG blends and LBG alone in all salts and salt concentrations. The addition of LBG in the blend (from 40% to 60% LBG) resulted in an increase in  $b$  values.

The slope of the power-law equation was used to determine the molecular conformational structure of polymers. In dilute regimes and at different ionic strengths, slope values greater than unity have been reported to be associated with random coil conformation (Lapasin & Priel, 1995). In dilute regimes, slope values less than unity have been reported to be associated with rod-like conformation (Lai & Chiang, 2002). In dilute regimes, slope values reported for xanthan alone in 100 mM NaCl varied from 1.07 to 1.25, whereas they were between 1.1 and 1.4 for LBG alone (Lapasin & Priel, 1995). In an investigation of hsian-tso leaf gum, Lai and Chiang (2002) found slopes in the dilute regime ranging from 0.78 to 0.8. They concluded that the molecular conformation of hsian-tso gum was more

Table 3

Slope values of log specific viscosity versus log concentration from the power-law equation for xanthan, locust bean gum (LBG), and their blends with addition of three salts at selected concentrations

Gum blend	Salts			
	Salt concentration (mM)	NaCl	KCl	CaCl <sub>2</sub>
Xanthan 100%	0	0.78 <sub>a</sub> ± 0.01	0.78 <sub>a</sub> ± 0.01	0.78 <sub>a</sub> ± 0.01
	5	0.91 <sub>b</sub> <sup>A</sup> ± 0.01	0.93 <sub>b</sub> <sup>A</sup> ± 0.03	0.92 <sub>b</sub> <sup>A</sup> ± 0.01
	50	1.06 <sub>c</sub> <sup>A</sup> ± 0.02	1.04 <sub>c</sub> <sup>A</sup> ± 0.02	0.91 <sub>b</sub> <sup>B</sup> ± 0.01
Xanthan 60%–LBG 40%	0	0.88 <sub>a</sub> ± 0.01	0.88 <sub>a</sub> ± 0.01	0.88 <sub>a</sub> ± 0.01
	5	1.13 <sub>b</sub> <sup>A</sup> ± 0.01	0.99 <sub>b</sub> <sup>B</sup> ± 0.02	0.92 <sub>a</sub> <sup>C</sup> ± 0.03
	50	1.23 <sub>c</sub> <sup>A</sup> ± 0.01	1.05 <sub>c</sub> <sup>B</sup> ± 0.03	1.07 <sub>b</sub> <sup>B</sup> ± 0.03
Xanthan 40%–LBG 60%	0	0.91 <sub>a</sub> ± 0.02	0.91 <sub>a</sub> ± 0.02	0.91 <sub>a</sub> ± 0.02
	5	1.36 <sub>a</sub> <sup>A</sup> ± 0.04	1.15 <sub>c</sub> <sup>B</sup> ± 0.01	1.08 <sub>b</sub> <sup>C</sup> ± 0.04
	50	1.15 <sub>b</sub> <sup>A</sup> ± 0.04	1.05 <sub>b</sub> <sup>B</sup> ± 0.08	1.11 <sub>b</sub> <sup>A</sup> ± 0.04
LBG 100%	0	1.23 <sub>a</sub> ± 0.05	1.23 <sub>a</sub> ± 0.05	1.23 <sub>a</sub> ± 0.05
	5	1.28 <sub>a</sub> <sup>A</sup> ± 0.04	1.18 <sub>b</sub> <sup>B</sup> ± 0.01	1.03 <sub>b</sub> <sup>C</sup> ± 0.01
	50	1.12 <sub>b</sub> <sup>B</sup> ± 0.01	1.18 <sub>b</sub> <sup>A</sup> ± 0.02	1.06 <sub>b</sub> <sup>C</sup> ± 0.01

Results are expressed as means ± SD for three replications.

<sup>A–C</sup> Means followed by the same letters in the same row are not significantly different ( $P < 0.05$ ).

<sup>a–c</sup> Means followed by the same letters in the same sub column are not significantly different ( $P < 0.05$ ).

rod-like than random coil. In this study, the slope value for the dialyzed gum blends and xanthan alone was less than unity. The slope value increased as salt was added, however, regardless of the salt type. The increase in slope values with the addition of LBG to the gum blends may suggest a possible conformational change of xanthan–LBG complex molecules from rod-like to random coil, which becomes more evident as salt concentration is increased. Sodium chloride seems to cause a molecular-conformation shift to random coil; whereas  $\text{CaCl}_2$  does not. The reason is not clear and is subject to further investigations.

Huggins parameters  $K$  and  $\alpha$  were used to determine the presence or absence of interaction between polymers. In this study, the  $b$  parameter was calculated, but not reported as an estimator for the polymer interaction. Sun et al. (1992) reported that parameter  $b$  should not be regarded as the only criterion for determining the presence of intermolecular interaction between polymers. The authors pointed out that, because  $b$  is a measure of the interaction between attractive molecules, this term may erroneously be estimated when repulsive molecules interact due to shear forces. The Huggins parameters were estimated for xanthan and LBG alone, and for the blends in all salts and salt concentrations. The  $K$  value indicates the ability for the polymers to aggregate. A  $K$  value greater than unity indicates aggregates, whereas  $K$  values less than unity indicate no aggregation (Millard, Dintzis, Wilett, & Klavons, 1997). The results in this study (Table 4) showed that the  $K$  values were all less than unity, suggesting no aggregation of the gums in the solution. The  $K$  values reported in this study were smaller than those reported by Sun et al. (1992) and Wang et al. (2001), suggesting that our solutions had a reduced propensity to aggregate.

The terms  $b_m$ ,  $b_1$ , and  $b_2$  were approximated from the intercept of the plot of specific viscosity against the concentration for the blend, xanthan alone, and LBG alone, respectively, and polymer interaction coefficient  $\alpha$  calcu-

lated (Eq. (18)). Results are shown in Table 5. Sun et al. (1992) reported that  $\alpha$  values greater than zero indicate a strong attraction between polymers, whereas negative values indicate strong repulsion. In this study, the values for the two gum blends in 0 and 5 mM salt were all greater than zero, whereas these values were negative in 50 mM salt. These results suggest that a strong attraction between xanthan and LBG occurred at 0 and 5 mM salt, whereas these gums repelled each other in 50 mM salt.

Wang et al. (2002) reported that NaCl reduced xanthan interaction with LBG due to electrostatic shielding. In aqueous solution with no added ions, the xanthan molecule is extended due to electrostatic repulsion of the negatively charged side chains (Carrington et al., 1996). When salt is added, charge screening causes the side chains to collapse down to the backbone, hence giving the xanthan molecule a rod-like shape and reducing the hydrodynamic volume (Rochefort & Middleman, 1987). The strong attraction observed in this study may have resulted from the increase in xanthan side-chain flexibility due to removal of counterions. Addition of 5 mM salt did not change the course of xanthan–LBG interaction. But electrostatic shielding was greater with 50 mM salt and a strong repulsion between xanthan and LBG resulted. These results are in agreement with the sharp drop in intrinsic viscosity observed with addition of 50 mM salt for xanthan and xanthan–LBG blends. Values for  $\alpha$  obtained in our study with addition of salt were larger than those reported by Sun et al. (1992) and Wang et al. (2001), and our results may suggest greater interaction magnitudes between xanthan and LBG related to the presence of salt.

### 3.3. Elastic component of gum solutions

The elastic component of xanthan, LBG, and their blends with all salts and salt concentrations is given Table 6. The elastic component was obtained with the highest

Table 4  
 $K$  values for xanthan, locust bean gum (LBG), and their blends with addition of three salts at selected concentrations

Gum blend	Salts			
	Salt concentration (mM)	NaCl	KCl	$\text{CaCl}_2$
Xanthan 100%	0	0.0011 <sub>c</sub> ± 0.0001	0.0011 <sub>b</sub> ± 0.0001	0.0011 <sub>a</sub> ± 0.0001
	5	0.012 <sub>b</sub> <sup>A</sup> ± 0.0004	0.011 <sub>a</sub> <sup>A</sup> ± 0.003	0.0084 <sub>b</sub> <sup>B</sup> ± 0.0012
	50	0.016 <sub>a</sub> <sup>A</sup> ± 0.0020	0.0092 <sub>a</sub> <sup>B</sup> ± 0.002	0.011 <sub>b</sub> <sup>B</sup> ± 0.0007
Xanthan 60%–LBG 40%	0	0.0003 <sub>c</sub> ± 0.0001	0.0003 <sub>b</sub> ± 0.0001	0.0003 <sub>b</sub> ± 0.0001
	5	0.0047 <sub>b</sub> <sup>B</sup> ± 0.0002	0.004 <sub>a</sub> <sup>B</sup> ± 0.0005	0.011 <sub>a</sub> <sup>A</sup> ± 0.0011
	50	0.011 <sub>a</sub> <sup>A</sup> ± 0.0021	0.006 <sub>a</sub> <sup>B</sup> ± 0.0004	0.008 <sub>a</sub> <sup>A</sup> ± 0.0012
Xanthan 40%–LBG 60%	0	0.0003 <sub>c</sub> ± 0.0001	0.0003 <sub>b</sub> ± 0.0001	0.0003 <sub>c</sub> ± 0.0001
	5	0.008 <sub>b</sub> <sup>C</sup> ± 0.0004	0.011 <sub>a</sub> <sup>B</sup> ± 0.0031	0.02 <sub>a</sub> <sup>A</sup> ± 0.0032
	50	0.02 <sub>a</sub> <sup>A</sup> ± 0.0030	0.011 <sub>a</sub> <sup>B</sup> ± 0.0022	0.02 <sub>b</sub> <sup>A</sup> ± 0.0021
LBG 100%	0	0.008 <sub>c</sub> ± 0.0031	0.008 <sub>c</sub> ± 0.0031	0.008 <sub>b</sub> ± 0.0031
	5	0.061 <sub>a</sub> <sup>A</sup> ± 0.0042	0.045 <sub>b</sub> <sup>B</sup> ± 0.0020	0.029 <sub>a</sub> <sup>C</sup> ± 0.0051
	50	0.031 <sub>b</sub> <sup>B</sup> ± 0.0051	0.059 <sub>a</sub> <sup>A</sup> ± 0.0025	0.029 <sub>a</sub> <sup>B</sup> ± 0.0010

Results are expressed as means ± SD for three replications.

<sup>A–C</sup> Means followed by the same letters in the same row are not significantly different ( $P < 0.05$ ).

<sup>a–c</sup> Means followed by the same letters in the same sub column are not significantly different ( $P < 0.05$ ).

Table 5  
Alpha values for xanthan–locust bean gum (LBG) blends with addition of three salts at selected concentrations

Gum blend	Salts			
	Salt concentration (mM)	NaCl	KCl	CaCl <sub>2</sub>
Xanthan 60%–LBG 40%	0	0.05 <sub>b</sub> ± 0.01	0.05 <sub>b</sub> ± 0.01	0.05 <sub>b</sub> ± 0.01
	5	17.2 <sub>a</sub> <sup>A</sup> ± 3.25	14.39 <sub>a</sub> <sup>A</sup> ± 6.62	6.24 <sub>b</sub> <sup>B</sup> ± 0.29
	50	−4.30 <sub>c</sub> <sup>B</sup> ± 0.01	−16.43 <sub>c</sub> <sup>A</sup> ± 1.53	−6.75 <sub>c</sub> <sup>B</sup> ± 1.42
Xanthan 40%–LBG 60%	0	0.02 <sub>b</sub> ± 0.01	0.02 <sub>b</sub> ± 0.01	0.02 <sub>b</sub> ± 0.01
	5	8.44 <sub>a</sub> <sup>A</sup> ± 1.83	2.49 <sub>c</sub> <sup>C</sup> ± 0.32	5.66 <sub>a</sub> <sup>B</sup> ± 2.53
	50	−12.79 <sub>c</sub> <sup>B</sup> ± 5.93	−21.06 <sub>c</sub> <sup>A</sup> ± 0.41	−6.41 <sub>c</sub> <sup>C</sup> ± 0.38

Results are expressed as means ± SD for three replications.

<sup>A–C</sup> Means followed by the same letters in the same row are not significantly different ( $P < 0.05$ ).

<sup>a–c</sup> Means followed by the same letters in the same sub column are not significantly different ( $P < 0.05$ ).

Table 6  
Elasticity values at the highest gum concentration corresponding to the dilute regime ( $C < C^*$ ) for xanthan, locust bean gum (LBG), and their blends with addition of three salts at selected concentrations

Gum blend	Salts			
	Salt concentration (mM)	NaCl (mPa s)	KCl (mPa s)	CaCl <sub>2</sub> (mPa s)
Xanthan 100% <sup>†</sup>	0	3.57 <sub>a</sub> ± 0.09	3.57 <sub>a</sub> ± 0.09	3.57 <sub>a</sub> ± 0.09
	5	0.77 <sub>b</sub> <sup>C</sup> ± 0.001	0.87 <sub>b</sub> <sup>B</sup> ± 0.018	0.95 <sub>b</sub> <sup>A</sup> ± 0.003
	50	0.72 <sub>a</sub> <sup>B</sup> ± 0.009	0.85 <sub>b</sub> <sup>A</sup> ± 0.005	0.74 <sub>b</sub> <sup>A</sup> ± 0.001
Xanthan 60%–LBG 40% <sup>†</sup>	0	22.02 <sub>a</sub> ± 0.58	22.02 <sub>a</sub> ± 0.58	22.02 <sub>a</sub> ± 0.58
	5	1.59 <sub>b</sub> <sup>B</sup> ± 0.15	2.38 <sub>b</sub> <sup>A</sup> ± 0.34	0.81 <sub>b</sub> <sup>C</sup> ± 0.07
	50	0.41 <sub>c</sub> <sup>B</sup> ± 0.02	0.91 <sub>c</sub> <sup>A</sup> ± 0.15	0.47 <sub>c</sub> <sup>B</sup> ± 0.05
Xanthan 40%–LBG 60% <sup>†</sup>	0	17.69 <sub>a</sub> ± 0.5	17.69 <sub>a</sub> ± 0.5	17.69 <sub>a</sub> ± 0.5
	5	0.44 <sub>b</sub> <sup>A</sup> ± 0.02	0.45 <sub>b</sub> <sup>A</sup> ± 0.01	0.31 <sub>b</sub> <sup>B</sup> ± 0.01
	50	0.28 <sub>b</sub> <sup>B</sup> ± 0.02	0.39 <sub>b</sub> <sup>A</sup> ± 0.03	0.26 <sub>b</sub> <sup>B</sup> ± 0.007
LBG 100% <sup>‡</sup>	0	0.31 <sub>a</sub> ± 0.002	0.31 <sub>a</sub> ± 0.002	10.31 <sub>a</sub> ± 0.002
	5	0.14 <sub>b</sub> <sup>A</sup> ± 0.02	0.11 <sub>c</sub> <sup>A</sup> ± 0.02	0.15 <sub>c</sub> <sup>A</sup> ± 0.01
	50	0.11 <sub>c</sub> <sup>B</sup> ± 0.01	0.25 <sub>b</sub> <sup>A</sup> ± 0.02	0.16 <sub>b</sub> <sup>B</sup> ± 0.012

Results are expressed as means ± SD for three replications.

<sup>A–C</sup> Means followed by the same letters in the same row are not significantly different ( $P < 0.05$ ).

<sup>a–c</sup> Means followed by the same letters in the same sub column are not significantly different ( $P < 0.05$ ).

<sup>†</sup> Highest xanthan and xanthan–LBG concentration (0.025%).

<sup>‡</sup> Highest LBG concentration (0.1%).

gum concentration for xanthan and the xanthan–LBG blend (0.025%), as well as for LBG alone (0.1%). Launay et al. (1997) reported that the Newtonian viscosity of xanthan corresponded to a xanthan concentration of 0.025 g/dl or less. For LBG, the elastic component response could be obtained for a concentration of 0.1% or higher. Locust bean gum concentrations lower than 0.1% appeared viscous. A concentration of 0.025% for LBG corresponded with the absence of the elastic component. By definition, fluids that do not contain an elastic component are considered purely viscous. According to that criterion, LBG was viscous in the 0.025 g/dl range, whereas xanthan and xanthan–LBG blends were visco-elastic. The results in this study showed that, as salt was added to the gum blends, the elastic component dropped significantly. A greater gum synergy was noticed with dialyzed xanthan 60%–LBG 40%, as evidenced by an elastic-component value two to three times larger than the average elastic-component value of the two gums. The synergy was still present,

but with less magnitude when 5 mM of salt was added to the blend, but it vanished with the addition of 50 mM of salt. These results were in agreement with the greater intrinsic viscosity, significantly different from that of the rest of the blend. But a significant decrease of the intrinsic viscosity resulted from the addition of salt in the blends.

Thurston (1996) used the oscillatory flow method and reported that the elastic component of 0.05% (w/v) xanthan was about 1.25 mPa. No other elastic-component values were found in the literature for gum solutions. Results in this study showed comparable, or even larger values for 100% xanthan and for xanthan 60%–LBG 40% blend with reduced gum concentration (0.025%), as long as salt concentration was kept low ( $\leq 5$  mM). In as much as the gum solutions were Newtonian, these results suggest that a critical role is played by the elastic component of the polymers in xanthan–LBG synergism. This role may be hindered by the addition of large amounts of salt.

#### 4. Conclusion

A strong interaction occurred between xanthan and LBG, even in dilute gum solution. But this interaction vanished with the addition of salts. The methods characterized the molecular conformation of xanthan and LBG alone, and suggested a conformational change of the complex xanthan–LBG, in which LBG and salts may play a significant role. The molecular conformation change may coincide with an increase or a decrease in the elastic component, due to removal or addition of salts, respectively.

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