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RESEARCH NOTE

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Dynamic Rheological Comparison of Selected Gum Solutions

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Abstract Dynamic rheological properties of commercial 0.8, 1.0, and 1.2% gums [carboxylmethylcellulose (CMC), guar gum, hydroxypropylmethylcellulose (HPMC), tara gum, and xanthan gum], which can be dissolved in cold water, were investigated by small-deformation oscillatory measurements. Magnitudes of storage (G') and loss (G") moduli increased with increasing concentration of gum solutions except for xanthan gum. Guar gum exhibited greatest G' and G" values among all gums except for G' value at 0.8% concentration. Slopes of G' and G" decreased with increasing concentration of gum solutions except for xanthan gum. Tan δ (G"/G') values decreased with increasing concentration of gum solutions except for xanthan gum. Tan δ values of xanthan gum solutions were much lower than those of other gum solutions, indicating that xanthan gum solutions were predominantly more elastic than viscous.

Key words: rheology, gum, storage modulus, loss modulus

Introduction

Gums are high-molecular-weight hydrophilic biopolymers used as functional ingredients in the food industry for the control of microstructure, texture, flavor, and shelf-life (1). Rheological properties of gums are of significance when they are used to control and modify textural properties of food products. In general, rheological properties of gum solutions are dependent on the structure, molecular weight, and concentration of the gums dissolved or dispersed in the solvent. Recently, many researchers have studied the rheological properties of gums in an aqueous solution (2-10) or in the formulated products (9, 11-14). However, only a few researchers (2, 6, 7, 9) have studied the rheological comparison of three different gum solutions or more under similar rheological conditions (concentration, pH, temperature, and frequency) mainly using the capillary or rotational viscometers. They found that the rheological properties of gums in an aqueous solution depended on the type of gum, concentration, and temperature. Most aqueous gum solutions in the literature are prepared by dispersing gums at high temperatures. However, in the food systems dry gums, which are dissolved or dispersed in water at room temperature, are added directly into some semi-solid foods, such as hot popper-soybean paste (5), salep (9), and tomato ketchup (14), to control and modify the textures of the products. Little comprehensive information is also available on comparative dynamic rheological properties of gum solutions, which can be dissolved in cold water, under similar rheological conditions.

In general, dynamic rheometry for small-deformation oscillatory measurement has been used to obtain valuable information on the viscoelastic properties of biopolymers without breaking their structural elements. This test allows researchers to relate dynamic rheological parameters to the molecular structure of the sample (15). Therefore, the dynamic rheometry can be a sensitive method for possible observation of the molecular structure properties of the gum solution system. The objectives of this study were to compare dynamic rheological differences between various gum solutions, which are dispersed in cold water, under similar rheological conditions by small-deformation oscillatory measurements, and to investigate the effect of gum concentration on the dynamic rheological properties of gum solutions.

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Materials and Methods

Materials and preparation of gum solutions Experimental studies on dynamic rheological properties of gums in solution were conducted with five commercial gums, including carboxylmethylcellulose (CMC) (type 7HOF; Hercules, Wilmington, DE, USA), guar gum (Rama Inst., Near G.I.D.C., India), hydroxypropylmethylcellulose (HPMC) (type MP874; Hercules), tara gum (Silva team Co., Buenous Aires, Argentina), and xanthan gum (CP Kelco, San Diego, CA, USA).

All gums used in this study were dissolved or dispersed in distilled water at room temperature. Aqueous solutions were prepared by mixing the gums with distilled water to obtain 0.8, 1.0, and 1.2% gum levels. A weighed amount of gum was gradually added to the appropriate amount of distilled water under stirring. The gum solution was continuously dispersed for 2 hr at room temperature with constant mild agitation provided by a magnetic stirrer to avoid agglomeration. The gum solutions were allowed to stand overnight in a refrigerator at 5°C, and for 2 hr at room temperature prior to conducting rheological measurements. Each solution was prepared in triplicate.

Dynamic rheological measurements Small-amplitude oscillatory rheological measurements of gum solutions were conducted with a TA AR1000 rheometer (TA Instruments Inc., New Castle, DE, USA) at 25°C with a plate-plate system (4 cm diameter) at a gap of 500 μm. Dynamic rheological data were obtained from frequency sweeps over the range of 0.63-62.8 rad/sec at 3% strain.

The 3% strain was in the linear viscoelastic region for each sample. Frequency sweep tests were performed after equilibration at 25°C for 5 min. TA rheometer Data Analysis Software (ver. VI. 1.76) was used to obtain the experimental data and to calculate storage modulus (G') and loss modulus (G"). All rheological measurements were in triplicate. Results reported are average of the three measurements.

Statistical analysis All results are expressed as mean± standard deviation. Analysis of variance (ANOVA) was performed using the Statistical Analysis System software (version 9.1). Differences in means were determined using Duncan's multiple-range test.

Results and Discussion

Changes in storage modulus (G') and loss modulus (G") as functions of the frequency (ω) for gum solutions at a typical concentration (0.8%) are shown in Fig. 1. In general, G' and G" values of gum solutions increased with increase in ω. Table 1 also shows G' and G" values at 6.3 rad/sec of gum solutions with different concentrations at 25°C. The G' and G" values of gum solutions increased with increase in gum concentration except for xanthan gum solution, in which there was no particular trend. The CMC, guar gum, and tara gum, in particular, exhibited a proportional increase in dynamic moduli (G' and G") within the concentration range of 0.8-1.2%. Da Silva and Rao (16) and Rao (17) also reported that the dynamic moduli increased when gum concentration in solution increased. Guar and xanthan gums showed higher G' values (15.3 and 15.7 Pa) in comparison to other gum solutions tested at 0.8% concentration, indicating that the guar and xanthan gums at lower concentrations were very effective in increasing the elastic property. The G' values (31.2 and 53.1 Pa) of guar gums at 1.0 and 1.2% were much higher than those of other gum solutions. In addition, the G" values of guar gum at all gum concentrations were relatively much higher than those of other gum solutions including tara gum, which is another galactomannan. Such lower values of dynamic moduli (G' and G") of tara gum may be attributed to its lower solubility (about 70%) in cold water when compared to guar gum (18). These results indicate a more pronounced effect of guar gum on the viscoelastic properties. The observed higher dynamic moduli of guar gum may be attributed to its long, soluble, and rigid chains that have a large hydrodynamic volume, as explained by Whistler and BeMiller (19). In general, it is well known that the guar gum produces high apparent viscosity and consistency index (K) due to its high-water binding capacity (20). Kayacier and Dogan (9) also reported that the increase in K values with respect to gum concentration (0.25-1.0%) was more prominent for guar gum compared to xanthan and alginate solutions. The changes in dynamic moduli (G'

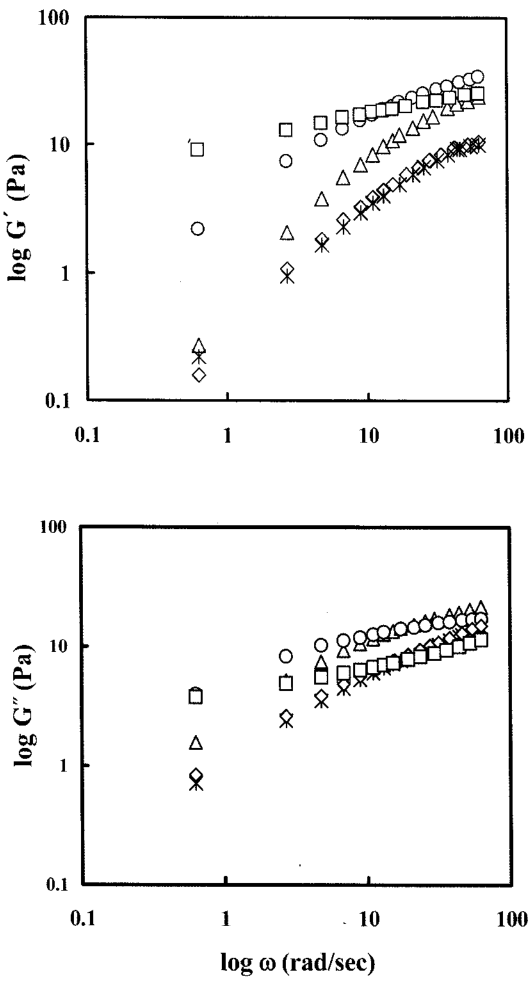

Fig. 1. Plots of log G' and log G" for gum solutions at 0.8% concentration: (○) guar gum; (□) xanthan gum; (△) tara gum; (◇) CMC; (*) HPMC.

Table 1. Storage (G') and loss (G") moduli at 6.3 rad/sec of gum solutions at 25°C

Gum Type	Concentration (%)	G' (Pa)	G" (Pa)	Tan δ
CMC	0.8	$2.07\pm0.03^{j1)}$	4.22 ± 0.02^{k}	2.04 ± 0.02^{a}
	1.0	4.59 ± 0.01^{i}	7.54 ± 0.04^{h}	1.65 ± 0.01^{d}
	1.2	7.84 ± 0.09^{h}	11.2 ± 0.01^{g}	1.42 ± 0.01^{e}
Guar	0.8	15.3 ± 0.01^{d}	11.9 ± 0.00^{f}	0.78 ± 0.01^{i}
	1.0	31.2 ± 0.76^{b}	21.1 ± 0.55^{c}	0.68 ± 0.01^{j}
	1.2	53.1 ± 0.80^{a}	30.5 ± 0.21^{a}	0.57 ± 0.01^{k}
HPMC	0.8	1.99 ± 0.00^{j}	3.97 ± 0.07^{k}	1.99 ± 0.02^{b}
	1.0	9.00 ± 0.32^{g}	12.0 ± 0.34^{f}	1.37 ± 0.02^{f}
	1.2	15.5 ± 0.02^{d}	19.9 ± 0.27^{d}	1.29 ± 0.01^{g}
Tara	0.8	4.11 ± 0.17^{i}	7.67 ± 0.30^{h}	1.86 ± 0.01^{c}
	1.0	10.7 ± 0.01^{f}	15.4 ± 0.15^{e}	1.43 ± 0.01^{e}
	1.2	21.3 ± 0.39^{c}	25.6 ± 0.17^{b}	1.20 ± 0.01^{h}
Xanthan	0.8	15.7 ± 0.19^{d}	6.07 ± 0.08^{i}	0.38 ± 0.01^{m}
	1.0	12.8 ± 0.15^{e}	5.48 ± 0.01^{j}	0.43 ± 0.01^{l}
	1.2	13.1 ± 0.22^{e}	5.50 ± 0.05^{j}	0.42 ± 0.00^{l}

[1)] Values are given as mean±standard deviation of triplicate; values in the same column with different superscripts are significantly different ($p < 0.05$).

and G") at gum concentrations of 0.8 and 1.0% also were steeper with HPMC solution in comparison to other gum solutions (Table 1). From the above observations, it can be concluded that the changes in dynamic moduli of gum solutions were influenced by the type of gum, and depended on the concentration of gum solutions except for xanthan gum.

In order to illustrate the differences in the viscoelastic behavior, the loss tangent 'tan δ' directly stating the G"/G' ratio, can be described as a characteristic parameter. The tan δ smaller than one indicates predominantly elastic behavior, while that greater than one indicates predominantly viscous behavior. The tan δ values of guar and xanthan gums were lower than one, indicating that these gum solutions were more elastic than viscous (Table 1). In particular, the tan δ values of xanthan gum solutions were much lower than those of other gum solutions, indicating that there was a more pronounced effect of xanthan gum on the elastic properties in the gum solution system. The viscoelastic properties of xanthan gum can be related to its relatively higher stiffness when compared to other gums. This rigidity implies a much more limited mobility of the chains and hence much longer relaxation times, resulting in higher elastic properties (21). However, the tan δ values of CMC were generally much higher than those of other gums. This observed result suggests that CMC is more viscous than elastic. The tan δ values also decreased with increasing concentration of gum solutions except for xanthan gum, in which only slight differences were observed between tan δ values with different gum concentrations. Literature did not list tan δ values for all gum solutions examined in this study, and gums listed were not always compared under similar rheological conditions.

The dynamic rheological data of log (G', G") versus log ω were also subjected to linear regression; the magnitudes of slopes and determination coefficients (R^2) are summarized in Table 2. The slopes of G' (0.19-0.72) and G" (0.08-0.55) were positive with high R^2 (0.93-0.99), and decreased with increasing concentration of gum solutions except for xanthan gum, indicating that the dynamic moduli greatly depended on frequency at lower gum concentrations. These results suggest that the viscoelastic properties of all gum solutions except for xanthan gum depended on gum concentration. The slope values of G' and G" for guar gum and xanthan gum with tan δ < 1 were much lower than those of other gums (Tables 1 and 2), indicating that guar gum and xanthan gum are more viscoelastic as compared to other gums. The slope values (0.19-0.21 Pa·sec) of G" for xanthan gum also were relatively higher than those (0.28-32 Pa·sec) of G', suggesting that xanthan gum solutions are more elastic than viscous. The xanthan gum also exhibited a relatively much lower frequency dependence of G' in comparison to other gums, indicating a much more limited mobility of the chains due to stiffer conformation of xanthan in aqueous solution (21, 22). This finding supports that xanthan gum solutions are predominantly more elastic than viscous, as previously discussed.

All gum solutions at 1.0% concentration except for xanthan gum showed the cross-point of G' and G" with positive slopes (Fig. 2). In general, at low frequency, G"

Table 2. Slope values of log G' and log G" vs. log ω curve of gum solutions with different gum concentrations at 25°C

Gum Type	Concentration (%)	Slope of G' (Pa·sec)	R^2	Slope of G" (Pa·sec)	R^2
CMC	0.8	$0.69\pm0.01^{b1)}$	0.97	0.53 ± 0.01^{b}	0.99
	1.0	0.65 ± 0.01^{cd}	0.98	0.47 ± 0.01^{c}	0.99
	1.2	0.61 ± 0.01^{ef}	0.99	0.42 ± 0.01^{e}	0.99
Guar	0.8	0.34 ± 0.01^{h}	0.97	0.16 ± 0.00^{j}	0.99
	1.0	0.30 ± 0.01^{i}	0.98	0.12 ± 0.00^{k}	0.98
	1.2	0.27 ± 0.01^{j}	0.98	0.08 ± 0.00^{l}	0.96
HPMC	0.8	0.66 ± 0.01^{c}	0.96	0.55 ± 0.01^{a}	0.99
	1.0	0.63 ± 0.01^{de}	0.99	0.43 ± 0.01^{d}	0.99
	1.2	0.60 ± 0.01^{f}	0.99	0.38 ± 0.01^{f}	0.99
Tara	0.8	0.72 ± 0.01^{a}	0.97	0.42 ± 0.01^{e}	0.98
	1.0	0.63 ± 0.01^{e}	0.98	0.33 ± 0.01^{g}	0.97
	1.2	0.55 ± 0.01^{g}	0.98	0.26 ± 0.01^{i}	0.96
Xanthan	0.8	0.21 ± 0.01^{k}	0.99	0.28 ± 0.00^{h}	0.99
	1.0	0.19 ± 0.00^{l}	0.93	0.32 ± 0.01^{g}	0.98
	1.2	0.19 ± 0.00^{l}	0.94	0.32 ± 0.00^{g}	0.99

1)Values are given as mean±standard deviation of triplicate; values in the same column with different superscripts are significantly different (p<0.05).

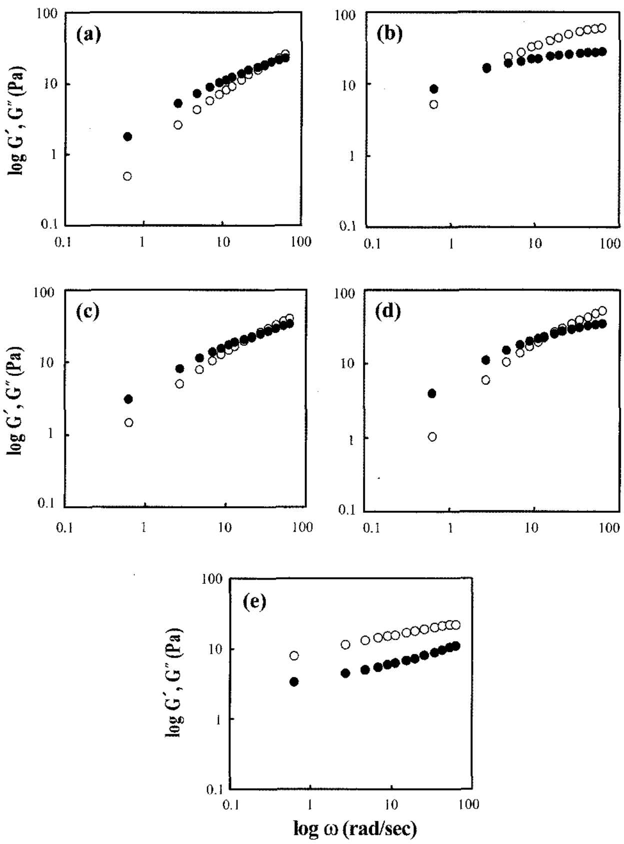

Fig. 2. Plots of log G' (○), log G" (●) vs. log ω for gum solutions at 1.0% concentration: (a) CMC, (b) guar gum, (c) HPMC, (d) tara gum, (e) xanthan gum.

values were higher than those of G' with frequency dependence, showing a liquid-like behavior (23). Such behavior is in good agreement with those found in most biopolymer solutions (24). CMC showed the cross-point at much higher frequency when compared to other gum solutions, indicating that CMC was predominantly more viscous than elastic. This was further supported by much higher tan δ value of CMC than those of other gum solutions (Table 1). The xanthan gum also showed little frequency dependence of G' and G" in comparison to other gum solutions. Moreover, G' was greater than G" over most of frequency range, typical of weak gels, as described by Doublier and Cuvelier (21). These properties of xanthan gum can be due to the association of ordered chain segments, giving rise to weak three-dimensional network (25). Izydorczyk *et al.* (26) also reported that, at sufficiently high concentration, xanthan gum exhibits weak gel-like properties due to its rather stiff, rod-like structure that shows good stability in cold solutions. In a comparison of dynamic moduli of xanthan gum and other gums, a more pronounced effect of xanthan gum on the elastic properties was observed in the gum solution system, as discussed previously. Therefore, it can be concluded that the dynamic rheological properties of gum solutions except for xanthan gum are governed by gum concentration and, more importantly, depend on the type of gum.

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