Food Sci. Biotechnol. Vol. 15, No. 4, pp. 589 ~ 594 (2006)

Food Science and Biotechnology

Effect of Gum Addition on the Rheological Properties of Rice Flour Dispersions

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Abstract The effect of five commercial gums (carboxylmethylcellulose, CMC; guar gum, GG; hydroxypropylmethylcellulose, HPMC; locust bean gum, LBG; and xanthan gum) at a concentration of 0.25% on the rheological properties of rice flour (RF) dispersions was investigated in steady and dynamic shear. The steady shear rheological properties showed that RF-gum mixture dispersions (5%, w/w) at 25°C had high shear-thinning flow behavior (n= 0.20-0.31) exhibiting a yield stress. Magnitudes of consistency index (K), apparent viscosity ($\eta_{a,100}$), and Casson yield stress (σ_{oc}) of RF-gum mixtures were much higher than those of RF dispersion with no added gum (control). Activation energy values (6.67-10.8 kJ/mole) of RF-gum mixtures within the temperature range of 25-70°C were lower than that (11.9 kJ/mole) of the control. Dynamic rheological data of log (G', G") versus log frequency (ω) of RF-gum mixtures had positive slopes (0.15-0.37) with G' greater than G" over most of the frequency range (0.63-63 rad/sec), demonstrating a frequency dependency. Tan δ (G"/G') values of RF-gum mixtures, except for xanthan gum, were much higher than that of the control.

Key words: rice flour, gum, rheological properties, apparent viscosity, storage modulus, loss modulus

Introduction

Gums (hydrocolloids), consisting of high-molecular-weight hydrophilic biopolymers, and starches or flours are often used together in many food systems to modify and control the rheological properties of starches or starch-based foods, control moisture retention, improve overall product quality and stability, reduce costs, and facilitate processing (1). The specific adjustment of the rheological properties of starch or flour is significant in the regulation of production processes and the optimization of applicability, stability, and sensory properties of food products (2). Therefore, knowledge about the rheological properties of starch-gum mixtures is important for understanding molecular interactions between starches and gums. In general, it is well known that the addition of gum may influence the viscosity and retrogradation of starch dispersions, as well as the syneresis of starch gels (3-8).

Rice is the main staple of Oriental nations. Their traditional foods, in general, are prepared from rice and rice-related materials. There are many different kinds of commercially available processed rice products, such as puffed grains, soups, candies, drinks, cakes, puddings, modified flours, and breakfast cereals (9, 10). The rice cake, which is made from rice flour (RF), is a very popular traditional food in Korea. Several researchers have studied RF's rheological properties (9-14).

Recently, only a few studies examined the effect of gums on rheological properties of flours, such as wheat flour (15) and acorn flour (16). They found that the addition of gums to flour dispersions had a great effect on the rheological properties of flour-gum mixtures, and that the extent of variation greatly depended on the chemical structure and concentration of the gum added. Yoo *et al.* (17) and Kim and Yoo (18) also reported that the rheological properties of mixtures of rice starch and gums such as galactomannans (guar gum and locust bean gum) and xanthan gum, depended on the type and concentration of gum and temperature. However, no studies as yet have been published examining the effect of various added gums on the dynamic rheological properties of RF dispersions. The dynamic rheological studies of RF-gum mixtures can provide valuable information through a molecular interpretation on the synergistic interactions of gums with RF dispersions. The present study investigates the steady and dynamic shear rheological properties of RF dispersions in the presence of various commercial gums.

Materials and Methods

Materials Rice was from Ansung, Korea. It was milled and ground to flour at a local mill, and then passed through a 120-mesh standard sieve (Chung Gye Inc., Seoul, Korea) with 125 μm openings using an analytical sieve shaker (Model AS200; Retsch GmbH & Co., Haan, Germany). The proximate RF composition was determined, in triplicate, according to AOAC (19). The five commercial food gums tested were carboxylmethylcellulose (CMC) (7HOF; Hercules, Wilmington, DE, USA), Guar gum (GG) (200/600; Sigma Co., St. Louis, MO, USA), hydroxypropylmethylcellulose (HPMC) (MP874; Hercules), locust bean gum (LBG) (Sigma Co.), and xanthan gum (CP Kelco, San Diego, CA, USA).

Preparation of rice flour-gum mixture dispersions RF-gum mixture dispersions (5%, w/w) were prepared by mixing RF with distilled water and gums to obtain a gum level of 0.25% (weight basis) as described by Yoo *et al.* (16) and Kim and Yoo (18). Flour dispersion with no added gum was also prepared as a control (0% gum). The mixture was moderately stirred in an Erlenmeyer flask with a screw cap for 1 hr at room temperature, and then

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Received April 14, 2006; accepted May 24, 2006

heated at 95°C in a water bath for 30 min with mild agitation provided by a magnetic stirrer. At the end of the heating period, the hot sample (95°C) was immediately transferred to the rheometer to measure the rheological properties. Each sample was prepared and measured in triplicate.

Rheological measurements Steady shear rheological measurements were performed in a concentric cylinder viscometer (VT550; Haake GmbH, Karlsruhe, Germany) at 25°C using the MV2 system. The rotating bob-to-cup diameter ratio was 0.876 and 53 mL samples were used. Temperature control was carried out with a constant temperature circulator (DS50-K10; Haake GmbH) which provided a working temperature range of 0-90°C (±0.1°C). The exposed sample was covered with a thin layer of light paraffin oil to minimize moisture loss during measurements. After equilibration at the sample measurement temperature for 20 min, the sample was sheared continuously from 0.1 to 500 1/sec. In order to describe the variation in the rheological properties of RF-gum mixtures under steady shear, the data were fitted to the well-known power law (Eq. 1) and Casson (Eq. 2) models:

$$\sigma = K\dot{\gamma}^{n} \qquad (1)$$

$$\sigma^{0.5} = K_{oc} + K_{c}\dot{\gamma}^{0.5} \qquad (2)$$

where, σ is the shear stress (Pa), $\dot{\gamma}$ is the shear rate (1/sec), K is consistency index (Pa·sec^{n}), and n is the flow behavior index (dimensionless). Casson yield stress (σ_{oc}) was determined as the square of the intercept (K_{oc}) that was obtained from linear regression of the square roots of shear rate-shear stress data. Using magnitudes of K and n, apparent viscosity ($\eta_{a,100}$) at 100 1/sec was calculated. The steady shear rheological measurements were also conducted at various temperatures in the range of 25-70°C in order to investigate the effect of temperature on $\eta_{a,100}$.

Dynamic shear rheological properties were measured in dynamic (oscillation) shear mode with a rheometer (AR 1000; TA Instruments Inc., New Cattle, DE, USA) at 25 °C, using parallel plate geometry with a 40 mm diameter upper plate and a gap of 500 μm. Dynamic rheological data were obtained from frequency sweeps over the range of 0.63-63 rad/sec at 3% strain. The 3% strain was in the linear viscoelastic region for each sample. The sample measurement temperature was kept constant at 25°C. Each sample was also held for 5 min before the dynamic shear rheological measurement for stress relaxation and temperature equilibrium. TA rheometer Data Analysis software (version VI. 1.76) was used to obtain all experimental data and to calculate storage modulus (G'), loss modulus (G''), and complex viscosity (η*). All steady and dynamic shear rheological experiments were conducted in triplicate. Results reported are the average of three measurements. Each curve presented is the most typical of three runs.

Results and Discussion

Proximate RF composition The rice flour (RF) to be tested had the following composition: 12.70% moisture, 7.50% protein (N×6.25), 1.32% fat, 0.97% ash, and 77.51% carbohydrate (by difference).

Steady shear properties The shear stress (σ) versus shear rate ($\dot{\gamma}$) data for RF-gum mixture dispersions at 0.25% gum concentration at 25°C are shown in Fig. 1. Experimental data of σ and $\dot{\gamma}$ were well fitted to two models, power law and Casson, with high determination coefficients (R^2= 0.93-0.99) (Table 1). All RF-gum mixture samples exhibited high shear-thinning behavior with values of flow behavior indexes (n) as low as 0.20-0.31, which were lower than that (0.36) of RF dispersion without gum (control), indicating the more pseudoplastic behavior of the RF-gum mixtures. This result is in good agreement with those found in rice starch-gum mixtures at low concentrations (<0.4%) (1, 17, 18). Such higher shear-thinning behavior of RF-gum mixtures may be attributed to the increased breakage of the intra- and inter-molecular associative bonding systems in the starch-gum mixtures due to higher shear rates. In particular, the RF-GG mixture exhibited the lowest n value (0.20) among the five samples. This result may be due to the molecule structure of GG in an extended form, which can readily interact with amylose molecules through non-covalent hydrogen bonding, producing an extended conformation that, in turn, increases the degree of pseudoplasticity (20, 21).

In general, the magnitudes of apparent viscosity ($\eta_{a,100}$), consistency index (K), and Casson yield stress (σ_{oc}) of RF-gum mixtures were much higher than those of the control (0% gum) (Table 1), indicating a higher synergism with gum due to starch-gum interaction. These results confirmed that the RF-gum mixtures are high shear-thinning fluids with high magnitudes of K and σ_{oc} compared to the control. Comparison of the K and σ_{oc} values for RF-gum mixture samples showed the RF-GG mixture to have higher values (K=29.5 Pa·sec^{n}; σ_{oc}=30.0 Pa), indicating the higher synergism with GG due to its greater hydration capacity as explained by Sudhaker *et al.* (21). According to Alloncle and Doublier (22), starch dispersions may be regarded as a composite material

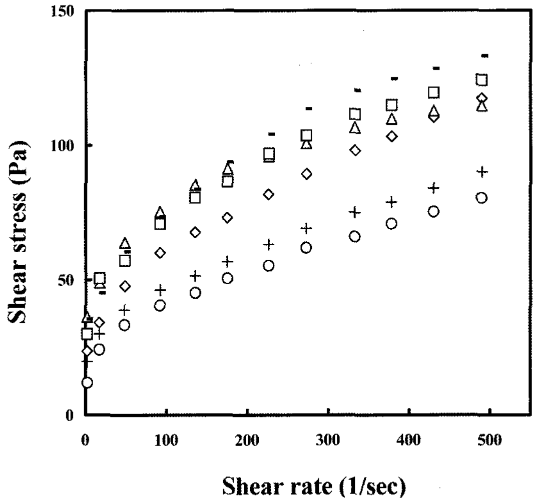

Fig. 1. Shear stress-shear rate plots of rice flour-gum mixtures at 25°C: (○) control (no gum), (□) CMC, (△) GG, (◇) MHPC, (−) LBG, (+) xanthan gum.

Table 1. Effect of gum addition on steady shear rheological properties of rice flour-gum mixtures at 25°C [1)]

Gum type	Apparent viscosity $\eta_{a,100}$ [2)] (Pa·sec)	Consistency index K (Pa·sec^n)	Flow behavior index n (-)	Yield stress σ_{oc} (Pa)
Control (no gum)	0.48±0.03	9.26±0.76	0.36±0.01	9.49±0.26
CMC	0.77±0.02	24.3±0.44	0.25±0.00	26.5±0.24
GG	0.74±0.02	29.5±0.38	0.20±0.01	30.0±0.70
HPMC	0.68±0.04	16.5±0.61	0.31±0.00	15.2±0.90
LBG	0.75±0.02	26.1±0.07	0.23±0.01	26.0±0.79
Xanthan	0.50±0.03	16.3±0.24	0.24±0.00	16.6±0.32

[1)] Values are given as mean±standard deviation of triplicate measurements.
[2)] $\eta_{a,100}$: viscosity at a shear rate of 100 1/sec.

consisting of swollen granules dispersed in a continuous biopolymer matrix (amylose). Therefore, gum is located within the continuous phase (amylose) which reduces the volume of this phase, leading to a dramatic increase in the concentration of gum in the continuous phase due to loss of water into swollen starch granules, thereby resulting in a very high viscosity (4). This result is in good agreements with those reported for mixtures of gums with wheat flour (15), acorn flour (16), and rice starch (2, 17, 18, 23).

Effect of temperature on apparent viscosity The effect of temperature on viscosity needs to be documented because a wide range of temperatures is encountered during processing and storage of fluid foods (24). The temperature dependency of apparent viscosity of RF-gum mixture dispersions at a specified shear rate can be analysed using Arrhenius model (Eq. 3), in which the apparent viscosity decreases in an exponential function with temperature:

$$\eta_{a,100} = A\cdot\exp(Ea/RT) \qquad (3)$$

where, $\eta_{a,100}$ is the apparent viscosity (Pa·sec) at 100 1/sec, A is a constant (Pa·sec), T is the absolute temperature (K), R is the gas constant (8.3144 J/mol·K), and Ea is the activation energy (kJ/mole). The magnitudes of Ea and A were determined at each guar gum concentration from regression analysis of 1/T vs. ln $\eta_{a,100}$. Table 2 illustrates that the apparent viscosity for all samples in relation to temperature obeyed the Arrhenius model, showing that the calculated values of Ea and constant A were in the range of 6.67-11.9 kJ/mole and 0.004-0.047 Pa·sec, respectively, with high determination coefficients (R^2=0.96-0.99). This Ea value is used to measure the relative sensitivity of the sample viscosity: a higher Ea value indicates a lower effect of temperature on the sample viscosity. Ea values (6.77-10.8 kJ/mol) of RF dispersions mixed with gums were lower than that (11.9 kJ/mol) of the control, indicating that the decrease in viscosity with temperature is more pronounced with the control. A similar trend was reported with acorn flour-guar (16), rice starch-galactomannans (17), and rice starch-xanthan mixtures (18).

Table 2. Activation energies of rice flour-gum mixture dispersions

Gum type	A ($\times10^{-2}$ Pa·sec)	Activation energy (kJ/mol)	R^2
Control (no gum)	0.40	11.9	0.97
CMC	1.05	10.7	0.97
GG	4.70	6.80	0.99
HPMC	0.95	10.7	0.96
LBG	0.99	10.8	0.96
Xanthan	3.47	6.67	0.97

Among the five RF-gum mixture samples examined, CMC, HPMC, and LBG showed higher Ea values (10.7-10.8) than GG (6.80) and xanthan gum (6.67), indicating that their apparent viscosity was more affected by temperature. Xanthan gum was also the least temperature dependent of all the gums studied. This is in good agreement with Kim and Yoo (18) who found similar results with rice starch-xanthan gum mixtures. Marcotte *et al.* (25) also reported that xanthan gum solutions had small viscosity changes with increasing temperature in comparison to other gums, and that the temperature effect was more pronounced at higher gum concentrations. Therefore, it can be concluded that the viscosity of the RF-xanthan gum mixture is less affected by temperature as compared to other mixtures, and that the Ea value of RF dispersions with different gums in the temperature range of 25-70°C is also dependent on the gum type. This result is in agreement with Marcotte *et al.* (25) who found a similar trend with different gum solutions.

Dynamic shear properties Figure 2 shows changes in G', G", and η^* as a function of the frequency (ω) for RF-gum mixtures at 25°C. G' and G" values increased with increasing ω, and G' was much higher than G" at all values of ω and displayed only a small frequency dependency. Log η^* vs. log ω plots also show shear-thinning behavior following the power law model. Such behavior is similar to those found in other starch or flour dispersions mixed with gums (2, 16-18, 22, 26). Table 3 shows G', G", and η^* values at 30 rad/sec for RF-gum mixtures at 25°C. In general, the dynamic moduli (G', G", and η^*) of RF-gum mixtures, except for CMC, were higher than those of the control (0% gum), indicating that the addition of gum to RF dispersion has a synergistic effect on viscoelastic properties. In particular, GG and xanthan gum showed much higher G' values (66.1 and 79.5 Pa) in comparison to other mixtures, indicating that GG and xanthan gum at low

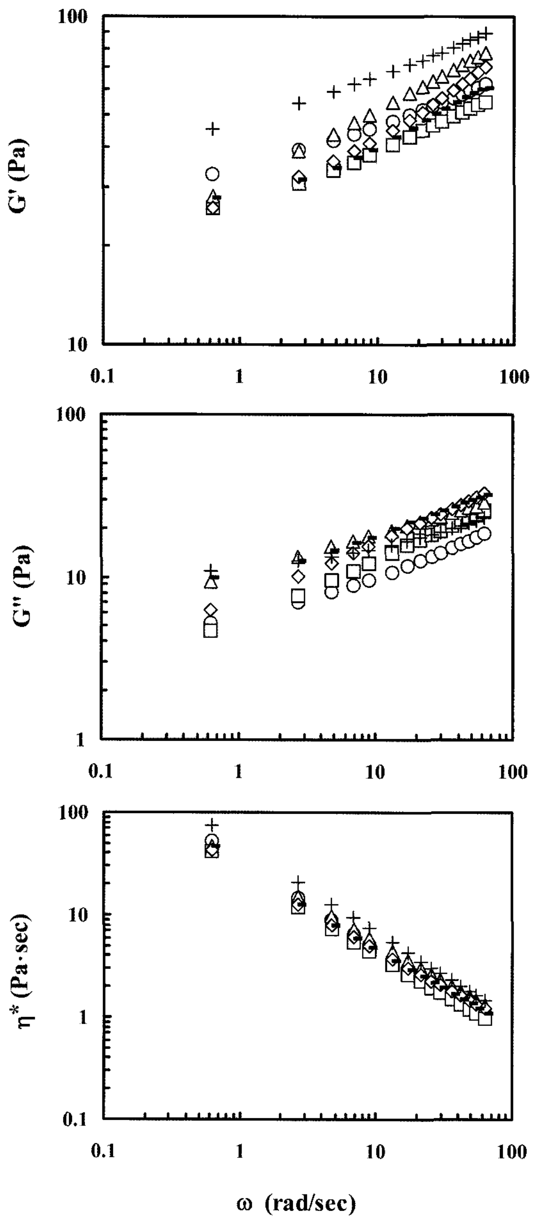

Fig. 2. Plots of log ω (frequency) versus log G' (storage modulus), log G" (loss modulus), and log η^* (complex viscosity) for rice flour-gum mixtures at 25°C: (○) control, (□) CMC, (△) GG, (◇) MHPC, (–) LBG, (+) xanthan gum.

Table 3. Storage (G') and loss (G") moduli, complex viscosity (η^*), and tan δ at 30 rad/sec of rice flour-gum mixtures at 25°C[1]

Gum type	G' (Pa)	G" (Pa)	η^* (Pa·sec)	tan δ
Control (no gum)	53.7±1.54	13.8±0.60	8.80±0.23	0.26±0.01
CMC	47.7±0.22	19.5±0.39	8.18±0.01	0.41±0.01
GG	66.1±0.12	23.7±0.14	11.1±0.02	0.36±0.01
HPMC	54.9±2.80	24.4±0.20	9.54±0.42	0.44±0.02
LBG	53.8±1.65	25.5±0.61	9.33±0.25	0.47±0.01
Xanthan	79.5±1.66	19.5±0.39	13.0±0.27	0.25±0.01

[1] Values are given as mean±standard deviation of triplicate measurements.

Table 4. Slope values from the curves of log G' and log G" versus log ω for rice flour-gum mixtures at 25°C[1]

Gum type	Slope of G' (Pa·sec)	R^2	Slope of G" (Pa·sec)	R^2
Control (no gum)	0.15±0.02	0.99	0.32±0.01	0.99
CMC	0.17±0.01	0.99	0.37±0.01	0.99
GG gum	0.22±0.01	0.99	0.24±0.01	0.99
HPMC	0.23±0.01	0.99	0.37±0.01	0.99
LBG	0.26±0.01	0.99	0.36±0.01	0.99
Xanthan	0.15±0.01	0.99	0.21±0.01	0.98

[1] Values are mean±standard deviation for triplicate measurements.

concentrations were very effective in increasing elastic property. Such dynamic behavior of RF dispersions mixed with GG and xanthan gum may be attributed to an increase in the viscoelasticity of the continuous phase in starch-gum composite systems due to the thickening properties of GG and xanthan gum as described by Alloncle and Doublier (22). Doublier and Cuvelier (26) also reported that the viscoelastic properties of xanthan gum can be related to its relatively higher stiffness 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. However, the RF-CMC mixture showed that elastic properties were decreased by the addition of CMC as previously indicated. Although G' values of the control were slightly higher than those of the RF-CMC mixture, the network structure of the control can be less stable in the light of the lower slope value of G' compared to the RF-CMC mixture, as shown in Table 4. In addition, this result suggests that the dynamic moduli in the RF-gum mixture systems can be influenced by other components of RF composition such as lipids, proteins, carbohydrates, and other non-starch molecules, as noted by Rojas et al. (15). From the above observations, it can be concluded that changes in the dynamic moduli of RF-gum mixtures were influenced by gum addition and the type of gum.

In order to illustrate the differences in viscoelastic behavior, the loss tangent 'tan δ', i.e. the G"/G' ratio, can be described as a characteristic parameter. A tan δ value less than one indicates predominantly elastic behavior, while that greater than one indicate predominantly viscous behavior. The tan δ values (0.25-0.47) of RF-gum mixtures were all less than one, indicating that the mixtures were more elastic than viscous, which is in good agreement with results reported in other starch or flour-gum mixtures (2, 16-18, 27). The tan δ values (0.36-0.47) of RF-gum mixtures, except for xanthan gum, were also higher than that of the control (0.26), showing that tan δ increased with gum addition. This means that G" increased much more than G' after gum addition to RF dispersion. In particular, the higher growth of G" than G' was more pronounced in the RF-LBG mixture (tan δ = 0.47) than in the other gum mixtures (tan δ = 0.25-0.44), demonstrating a more pronounced effect of LBG on the viscous properties of RF dispersion. However, the tan δ value of RF dispersion did not change much with the addition of xanthan gum, indicating that the viscous properties of RF dispersion were not affected by the addition of xanthan gum. Choi *et al.* (28) also reported a more pronounced effect of xanthan gum on elastic properties based on a comparison of the dynamic moduli of xanthan gum and other gums in the gum solution system. From these observations, it is obvious that the viscous properties of RF-gum mixtures were affected by the addition of gums, except for xanthan gum, and depended on the type of gum. In general, the dependence of viscous properties on gum type can be explained by thermodynamic incompatibility between the amylose component and the gums, as explained by Alloncle and Doublier (22) and Eidam and Kulicke (29). Therefore, the rheological behavior in RF-gum mixture is determined by the characteristics of the rice starch (permanent junction zones in the network) and of the gums (temporary entanglements in the network) (2, 29). According to Eidam and Kulicke (29), the gum molecules in the three-dimensional network function as disruptive sites, resulting in network defects. The number of permanent junction zones in the starch is reduced and the overall elasticity decreases. Therefore, the decreased number of junction zones of rice starch molecules increases the viscous properties. Based on the above statement, it can be concluded that the synergistic effect of gum on the rheological properties of RF dispersion may be attributed to the increased viscoelasticity of the continuous phase, in which gum is concentrated in starch-gum composite systems because of the thickening properties and greater hydration capacity of gum as explained by Alloncle and Doublier (22). Finally, these observations indicate that the addition of gum to RF dispersion has a pronounced effect on the dynamic rheological properties in RF-gum mixture systems, and that the extent of variation greatly depends on the type of gum, as also noted by Yoo *et al.* (16).

The dynamic rheological data of log (G', G") versus log ω were also subjected to linear regression. The magnitudes of the slopes and determination coefficients (R^2) are summarized in Table 4. The slopes of G' (0.15-0.26) and G" (0.21-0.37) were positive with high R^2 values (0.98-0.99). The slopes of G" in the RF-gum mixture systems were relatively higher than those of G', while the slope of G' increased with gum addition, except for xanthan gum. There were also differences between slope values for the control and RF-gum mixtures, except for xanthan gum. These observed results show that the elastic properties of RF dispersion can be decreased by gum addition, except for xanthan gum. The slopes (0.21-0.24) of G" for RF dispersions mixed with GG and xanthan gum were relatively lower than those (0.36-0.37) for CMC, HPMC, and LBG, demonstrating that their G" was less dependent on frequency than that of the RF dispersion containing CMC, HPMC, or LBG. Xanthan gum also showed a relatively low frequency dependence of G' and G" in comparison to those of the other gums as observed by Choi *et al.* (28) who studied the dynamic rheological properties of xanthan gum in aqueous solution.

From a structural point of view, log (G', G") versus log ω plots for true gels have zero slope, while for weak gels they have positive slopes and G' is greater than G" with a frequency dependency (30). These dynamic rheological data (Table 3 and 4) suggested that the dynamic rheological behavior of the RF-gum mixtures is similar to that of weak gel because the slopes of G' (0.15-0.26) and G" (0.21-0.37) were positive and the magnitudes of G' (47.7-79.5 Pa) were much higher than those of G" (13.8-25.5 Pa) at 30 rad/sec, giving tan δ values in the range of 0.25-0.47 as described by Ross-Murphy (30). Such behavior is in good agreement with the results found in most starch or flour dispersions mixed with different gums, such as GG (2, 16, 22, 27, 31), LBG (2, 17, 22, 31), and xanthan gum (2, 3, 18, 22, 31).

Acknowledgments

This work was supported by the Dongguk University research fund.

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