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Effect of Guar Gum on Rheological Properties of Acorn Flour Dispersions

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Abstract Rheological properties of acorn flour-guar gum mixtures (4% w/w) at different guar gum concentrations (0, 0.2, 0.4, 0.6, and 0.8% w/w) were evaluated in steady and dynamic shear. The acorn flour-guar gum mixtures at 25°C showed high shear-thinning flow behavior (n= 0.20-0.27). Consistency index (K), apparent viscosity ($\eta_{a,100}$), and Casson yield stress (σ_{oc}) increased with the increase in guar gum concentration. Within the temperature range of 25-70°C, the $\eta_{a,100}$ of mixtures obeyed the Arrhenius relationship with high determination coefficient (R^2= 0.974-0.994). Activation energy values (5.37-6.77 kJ/mole) of acorn flour dispersions in the mixtures with guar gum (0.2-0.8%) were much lower than that (12.5 kJ/mole) of acorn flour dispersion (0% guar gum). Storage modulus (G'), loss modulus (G"), and complex viscosity (η^*) increased with the increase in guar gum concentration. Dynamic rheological data of ln (G', G") versus ln frequency (w) of guar gum-acorn flour mixtures had positive slopes with G' greater than G" over most of the frequency range, indicating that they exhibited weak gel-like behavior.

Key words: acorn flour, guar gum, rheological property, apparent viscosity, storage modulus

Introduction

The addition of gum to starch or flour in food system is widely used to modify and control the rheological properties of starches or starch-based foods. The specific adjustment of the rheological properties of starch is of significance in order to regulate production processes and to optimize applicability, stability, and sensory properties of food products (1). In particular, since both ingredients exist together in many food systems, knowledge about the rheological properties of starch-gum mixtures is important for understanding molecular interactions between starches and gums (2). Generally, it is well known that the addition of gum may influence the viscosity and retorgradation of starch dispersions, as well as the syneresis of starch gels (3-9). According to Alloncle *et al.* (4), the observed enhancements in overall viscosity of starch could be attributed to the increase in gum concentration produced by swelling of the starch granules during gelatinization in starch-galactomannan mixtures (guar gum or locust bean gum). Therefore, understanding rheological properties of starch-gum mixtures will lead to improvements in the formulation of starch-based foods.

Guar gum is a water soluble polysaccharide which consists of a chain of $(1 \rightarrow 4)$-linked β-D-mannopyranosyl units with single branching α-D-galactopyranosyl units connected via $(1 \rightarrow 6)$ linkage, on average, to every second main chain unit (10). The most extensive branching of guar has been known to be responsible for its easier hydration properties as well as its greater hydrogen-bonding activity which may also take place within and/or between polysaccharides, proteins, and starches and could enhance complex formation (11). The most important property of guar gum is its ability to produce high viscosity with pseudoplastic rheology at low concentration, because it is composed of long, soluble and rigid chains, which have a large hydrodynamic volume (12).

Acorn is an edible oval nut, which mainly contains starch, obtained from oak tree. It is known that there are many different kinds of commercially available processed acorn products, such as bread, cake, confectionary, and gels, which are made from acorn flour (13). Among them, the acorn gel, which is called mook, is a potential starch source in Korea and is the most popular traditional food. Several researchers have studied flour and starch obtained from acorn in order to characterize rheological properties of acorn gels (13-15).

Recently, many researchers have studied the effect of gums on rheological properties of various cereal starches. Rojas *et al.* (16) also studied the effect of guar gum on the pasting properties of wheat flour dispersions. However, a study on the rheological properties of acorn flour-guar gum mixtures has not yet been carried out. In particular, there are no studies the effects of guar gum on dynamic rheological properties of acorn flour dispersions in the starch-gum mixture systems. Therefore, the main objectives of the present study were to determine the rheological properties of acorn flour-guar gum mixtures in steady and dynamic shear, and to evaluate the effect of guar gum concentration and temperature on rheological properties.

Materials and Methods

Materials Acorn flour and guar gum were purchased from Nongmin Food Co. (Daejeon, Korea) and Sigma Co. (St. Louis, MO, USA), respectively. The proximate composition of acorn flour was determined, in triplicate, according to AOAC (17).

Preparation of acorn flour dispersions Acorn flour-

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guar gum mixtures (4% w/w) were prepared by mixing acorn flour with distilled water, and guar gum to obtain 0.2, 0.4, 0.6, and 0.8% (weight basis) gum levels. A flour dispersion with no added gum (0% guar gum) was also prepared as a control. The mixture was moderately stirred for 1 hr at room temperature, and then was 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 mixture was immediately transferred to the rheometer for the measurements of rheological properties.

Rheological measurements The steady rheological properties were measured with a concentric cylinder viscometer (Haake VT550, Haake Inc., Germany) at 25°C using the MV2 system. A constant temperature circulator (Model DS50-K10, Haake GmbH, Karlsruhe, Germany) was used to provide the working temperature in the range of 0 to 90 °C (±0.1°C). In order to describe the variation in the rheological properties of acorn flour-guar gum mixtures under steady shear, the data were fitted to the well-known power law (Eq. 1) and Casson (Eq. 2) models:

$$\sigma = K\dot{\gamma}^{n} \tag{1}$$

$$\sigma^{0.5} = K_{oc} + K_c\dot{\gamma}^{0.5} \tag{2}$$

Where σ is the shear stress (Pa), $\dot{\gamma}$ is the shear rate (s^{-1}), K is consistency index ($Pa.s^n$) 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 s^{-1} was calculated. Furthermore, the effect of increasing temperature (25-70 °C) on $\eta_{a,100}$ was also studied on acorn flour-guar gum mixtures.

The dynamic rheological properties were obtained with a Carri-Med CSL^2 100 rheometer (TA Instruments Inc., New Cattle, DE) at 25°C, using a parallel plate system (4 cm dia.) at gap 500 mm. Dynamic rheological data were obtained from frequency sweeps over the range of 0.63-63 rad/s 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. Carri-Med software (version 3.1) was used to obtain the experimental data and to calculate storage modulus (G′), loss modulus (G″) and complex viscosity (η^*). All rheological experiments were conducted in triplicate. Results reported are an average of the three measurements.

Results and Discussion

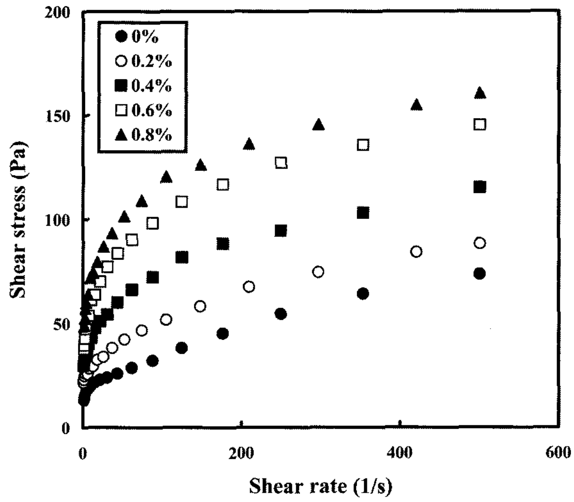

Fig. 1. Shear stress vs. shear rate plots of acorn flour dispersions with different guar gum concentrations at 25°C.

Proximate compositions The tested acorn flour had the composition: 11.56% moisture, 1.22% protein (N x 6.25), 0.25% fat, 0.44% ash, and 86.53% carbohydrate (by difference).

Steady shear properties The shear stress (σ) versus shear rate ($\dot{\gamma}$) data for acorn flour-guar gum mixtures at different guar gum concentrations at 25°C are shown in Fig. 1. Experimental data of σ and $\dot{\gamma}$ were fitted well to two models, power law model and Casson model, with high determination coefficients (R^2= 0.922-0.998). All the samples had high shear-thinning behavior with values of flow behavior indexes (n) as low as 0.20-0.27 (Table 1). The n values (0.20-0.23) of acorn flour-guar gum mixtures did not change much with the increase in guar gum concentration from 0.2 to 0.8%. In general, the observed shear-thinning behavior can be explained by disruption of an entangled polysaccharide molecule network during shearing, as described by Morris (18). The rate of disruption of existing intermolecular entanglements becomes greater than the rate of reformation of intermolecular entanglement with increasing shear rate, with a resultant decrease in apparent viscosity.

The magnitudes of apparent viscosity ($\eta_{a,100}$), consistency index (K), and Casson yield stress (σ_{oc}) obtained from the power law and Casson models increased with the increase in guar gum concentration. Compared to acorn flour dispersion (i.e. without guar gum), acorn flour dispersions at different guar concentrations also showed much higher magnitudes of σ_{oc}, in the range of 22.3-61.7. From these

Table 1. Effect of guar gum concentration on steady shear rheological properties of acorn flour dispersions at 25°C

Guar gum concentration (%)	Apparent viscosity, $\eta_{a,100}$ (Pa.s)	Consistency index, K ($Pa.s^n$)	Flow behavior index, n (-)	Yield stress σ_{oc} (Pa)
0	0.38±0.01	10.7±0.05	0.27±0.01	12.6±0.09
0.2	0.51±0.02	19.7±0.23	0.20±0.01	22.3±0.72
0.4	0.74±0.01	25.6±0.01	0.23±0.01	30.9±1.30
0.6	1.02±0.01	35.8±2.17	0.22±0.01	44.9±0.90
0.8	1.29±0.03	50.8±2.18	0.20±0.01	61.7±2.44

observations, it could be concluded that the acorn flour-guar gum mixtures were highly shear-thinning fluids with high magnitudes of yield stresses. The magnitudes of $\eta_{a,100}$ and K of acorn flour-guar gum mixtures (0.2-0.8% guar gum) were also much higher than those of acorn flour dispersion (0% guar gum), indicating that there was a higher synergism with guar gum, because of its greater hydration capacity. According to Alloncle and Doublier (19) and Morris (20), starch paste may be regarded as a composite material consisting of swollen granules (amylopectin) dispersed in a continuous biopolymer matrix (amylose). Alloncle *et al.* (4) indicated that guar gum is located within the continuous phase (amylose), and thus the volume of this phase is reduced, which causes a dramatic increase in guar gum concentration in the continuous phase, thereby resulting in a very high viscosity. These results suggested a strong influence of guar gum on steady shear properties of acorn flour dispersions on guar gum concentration. The observed behavior is in good agreements with those found for the mixtures of guar gum with wheat flour (16) and other starches, such as corn starch (2, 11, 21), wheat starch (2, 11), triticale starch (22), rice starch (2, 23) and yam starch (24). Therefore, in our study the observed effect of guar gum on changes in viscosity of acorn flour dispersions can be explained by the guar gum concentration located within the continuous phase of the medium.

Effect of temperature on apparent viscosity The viscosity flow curves of the mixture of acorn flour with 0.4% guar gum at different temperatures (25, 40, 55, and 70°C) are shown in Fig. 2 to describe the relationship between shear rate and viscosity. The apparent viscosity values decreased with the increase in temperature from 25 to 70°C. The effect of temperature on apparent viscosity of fluid foods at a specified shear rate can be described by the Arrhenius relationship (Eq. 3), in which the apparent viscosity decreases in an exponential function with temperature. The Arrhenius temperature relationship has been confirmed experimentally in previous studies of rice

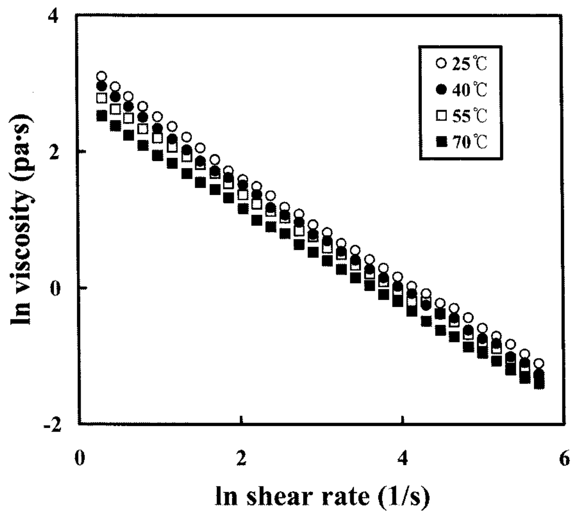

Fig. 2. Viscosity flow curves of acorn flour-guar gum mixture (0.4% guar gum) at different temperatures.

Table 2. Activation energies of acorn flour dispersions with different guar concentrations

Guar gum concentration (%)	A(Pa.s)	Activation energy (kJ mol^{-1})	R^2
0	0.002	12.5	0.990
0.2	0.042	6.18	0.985
0.4	0.082	5.46	0.974
0.6	0.117	5.37	0.983
0.8	0.083	6.77	0.994

starch-xanthan gum mixture (9), rice flour dispersion (25), modified corn starch (26), pectin gel (27) and mustard suspension (28).

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

where $\eta_{a,100}$ is apparent viscosity (Pa.s) at 100 s^{-1}, A is a constant (Pa.s), 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 concentration from regression analysis of 1/T vs. ln $\eta_{a,100}$. The calculated values of Ea and constant A of acorn flour-guar gum mixtures were in the range of 5.37-12.5 kJ/mole and 0.002-0.117 Pa.s, respectively, with high determination coefficients (R^2= 0.96-0.99) (Table 2). The observed low Ea values of mixtures mean that they are not greatly influenced by changes in temperature. Ea values (5.37-6.77 kJ/mol) of acorn flour-guar gum mixtures (0.2-0.8% guar gum) were also much lower than that (12.5 kJ/mol) of acorn flour dispersion (0% guar gum), indicating that the decrease in viscosity with temperature is more pronounced at 0% guar gum concentration. The low Ea value also means that the rheological properties are less temperature-dependent in the concentration range studied, as also noted by Holdsworth (29). Therefore, it can be concluded that the Ea of acorn flour dispersions with different guar concentrations (0.2-0.8%) in the temperature range of 25-70°C are not significantly affected by the guar concentrations. This also is in good agreement with the results obtained by Shi and BeMiller (2) who found the similar trend with starch-guar gum mixtures.

Dynamic shear properties Figure 3 shows changes in storage modulus (G'), loss modulus (G"), and complex viscosity of acorn flour-guar gum mixtures (η^*) as a function of the frequency (w) for a typical acorn flour-guar gum mixture (i.e. 0.4% guar gum) at 25°C. G' is a measure of the energy that is stored in the material or recoverable per cycle of deformation. G" is a measure of the energy that is lost as viscous dissipation per cycle of deformation. η^* is a measure of the overall resistance to flow. G' and G" values were found to increase with the increase in w, and G' was much higher than G" at all values of with the small frequency dependency. Ln η^* vs. ln w plots also shows shear-thinning behavior following the power law model. As shown in Fig. 4, the dynamic moduli (G', G", and η^*) of acorn flour-guar gum mixtures increased with the increase in guar gum concentration and showed much higher values than that of acorn flour dispersion (0% guar gum), indicating that the synergistic effect of guar gum on

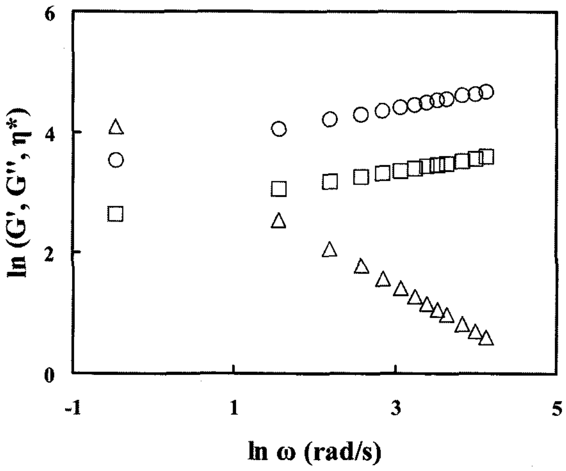

Fig. 3. Plot of ln w vs. ln G′ (○), ln G″ (□), and ln η^* (△) for acorn flour-guar gum mixture (0.4% guar gum) at 25°C.

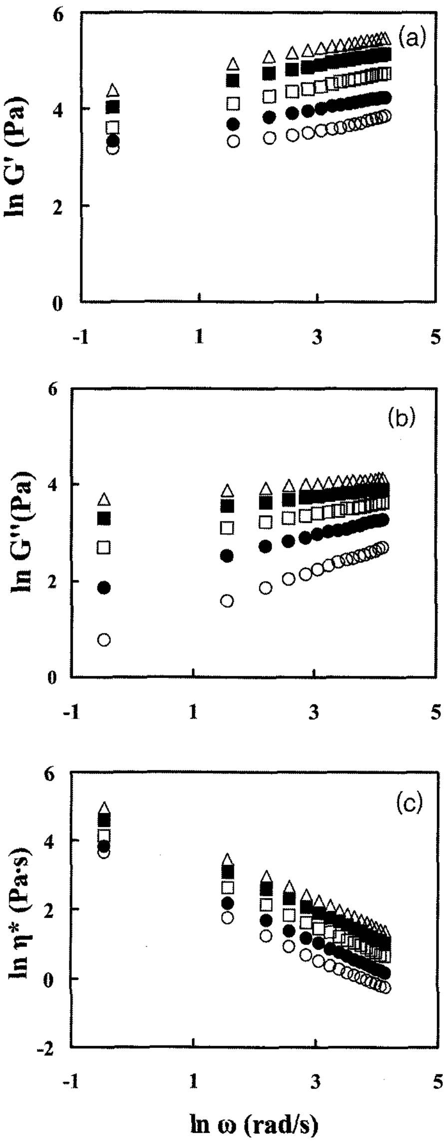

Fig. 4. Plots of ln ω vs. ln G′, ln G″ and ln η^* for acorn flour dispersions with different guar gum concentrations at 25°C. Guar gum concentrations were : (○) 0%, (●) 0.2%, (□) 0.4%, (■) 0.6%, and (△) 0.8%. (a) G′, (b) G″, and (c) η^*.

rheological properties of acorn flour dispersions was more pronounced when guar gum concentration was increased. Similar trend was reported with other starches containing guar gum (21, 30).

From a structure point of view, for weak gels ln (G', G") versus ln w plots have positive slopes and G' is greater than G" over a wide range of w, as indicated by Ross-Murphy (31). As shown in Fig. 3 and Table 3, it was found that all samples displayed weak gel-like behavior because the slopes of G' (0.24-0.68) and G" (0.08-0.41) were positive and the magnitudes of G' (37.3-207) were much higher than those of G" (10.7-57.4) at 30 rad/s, showing that the G"/G' ratio (tan d) is in the range of 0.28-0.36, as also described by Ross-Murphy (31). These results suggest that all the samples were more elastic than being viscous. The magnitudes of G', G", and η^* also increased with the increase in guar gum concentration from 0 to 0.8%. Similar trend was also observed for triticale starch-guar gum mixtures in the guar gum concentration range of 0.25-0.75% (22). From these observations, it was concluded that the dynamic rheological properties of acorn flour dispersions were influenced by the addition of guar gum and were dependant on guar gum concentration.

Many researchers have studied the interaction between hydrocolloids and starches. In general, the synergistic effect of hydrocolloids on rheological properties of starch pastes has been described by various mechanisms, including interaction between starch exudates and gum, an increase of gum concentration in the continuous phase of the medium, and the influence of gum on the physical properties of starch granules, such as size, shape, and granule integrity, as well as the amount of exudates from starch granules (32). Based on a similar mechanism, the synergistic effect of guar gum on rheological properties of acorn flour dispersions may be attributed to an increase in the viscoelasticity of the continuous phase, in which guar gum is concentrated in starch-gum composite systems because of the thickening properties and greater hydration capacity of guar gum, as described by Alloncle and Doublier (19). They also explained that the continuous phase plays an important role in maintaining the viscoelastic behaviour of the mixtures, when the gum concentration is increased. Hence, based on the above statement, it can be predicted that the increase in dynamic modulus values in acorn flour-guar gum mixture systems may be due to an increase of the guar gum concentration within the continuous phase. Finally, the above results indicate that the addition of guar gum to acorn flour dispersions has a pronounced effect on the rheological

Table 3. Storage (G') and loss (G") moduli, and complex viscosity (η^*) at 30 rad/s of acorn flour dispersions with different guar gum concentrations

Guar gum Concentration (%)	G' (Pa)	G" (Pa)	η^* (Pa.s)
0	37.3±0.26	10.7±0.61	1.30±0.01
0.2	59.7±2.30	21.2±0.56	2.13±0.08
0.4	92.0±3.60	31.4±0.70	3.28±0.12
0.6	143±3.05	44.1±0.88	5.07±0.11
0.8	207±3.21	57.4±0.80	7.27±0.12

properties in the acorn flour-guar gum mixture systems, and the extent of variation greatly depends on the guar gum concentration. However, extensive research is still required to further understand the role of guar gum in changing the rheological properties of starch-guar gum mixtures in steady and dynamic shear.

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