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Original research

Effect of edible organic acids (ascorbic, citric, malic, and tartaric) on the rheological behavior of xanthan gum dispersion

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A B S T R A C T

The present study aims to determine the influence of edible organic acids (ascorbic, citric, malic, and tartaric) at two concentrations (0.5, and 1 %) on the viscosity and rheological behavior of xanthan gum dispersion (0.2%, w/v). The results of this study showed that the apparent viscosity of xanthan gum dispersion reduced as the shear rate (SR) increased (shear-thinning behavior). Furthermore, the apparent viscosity of the xanthan gum dispersion decreased as the organic acid concentration increased. The highest decrease in viscosity was related to 1% citric acid and the lowest was related to 0.5% ascorbic acid. The rheological behavior of xanthan gum dispersion was successfully modeled using Power law, Bingham, Herschel-Bulkley, and Casson models, and the Power law model was the best one for describing the behavior of xanthan gum dispersion containing edible organic acids. The Power law model showed good performance with the maximum r-value (mean r-value=0.993) and least sum of squared error (SSE) values (mean SSE value=0.115) and root mean square error (RMSE) values (mean RMSE value=0.046) for all samples. The consistency coefficient values of the samples (Power law and Herschel-Bulkley models) reduced as the acid percent was increased. The sample containing 1% citric acid had the lowest consistency coefficient and the sample containing 0.5% ascorbic acid had the highest consistency coefficient. Based on the results of this research, the use of xanthan gum in food products containing high concentrations of citric acid is not recommended.

Keywords: Consistency coefficient; Herschel-Bulkley; Power law; Xanthan gum

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1. Introduction

Gums are hydrophilic non-starchy carbohydrate polymers with high molecular weight (Bak and Yoo, 2023). By having various functional properties, gums (hydrocolloids) can be exploited for many applications in different food industries (Salehi, 2020; Yousefi et al., 2022). Nowadays, one of the most versatile hydrocolloids (also widely use in the food industry) is xanthan gum (Martins et al., 2023). Xanthan gum is an anionic gum and a microbial (obtained from *Xanthomonas campestris*) heteropolysaccharide usually utilized in emulsions such as mayonnaise and salad dressings to provide an enhanced gel-like texture (Nor Hayati et al., 2016; Nsengiyumva and Alexandridis, 2022). Xanthan gum is used to achieve high viscosity and pseudoplasticity, and has excellent suspending and stabilizing properties (Martins et al., 2023; Salehi and Inanloodoghouz, 2024). Xanthan gum is often used in food emulsions because of its high acid, alkali and heat resistance and unique rheological properties, which allow it to have high viscosity at low shear rates, resist the Brownian motion of droplets and maintain the static stability of the emulsion (Cancella et al., 2024; Liang et al., 2024).

The functional properties of food hydrocolloids are remarkably affected by the quality of solvent/cosolutes in a food system (Yousefi et al., 2022). For example, the effect of acidification on a typical commercial xanthan and on pyruvate-free xanthan, alone and in gelling mixtures with konjac glucomannan, has been studied by Agoub et al. (2007). Their results showed that in both xanthan samples, progressive reduction in pH caused a progressive increase in temperature of the disorder–order transition in differential scanning calorimetry, and a progressive reduction in gelation temperature with konjac glucomannan. Dogsa et al. (2014) reported that the rheological properties of carboxymethyl cellulose at

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different pH values were directly related to the carboxymethyl cellulose supramolecular structure in the aqueous environment. The effect of organic acids on the viscosity and rheological behavior of guar gum solutions was studied by Salehi et al. (2024). Their results showed that the apparent viscosity of the guar gum dispersions decreased as the concentration of organic acids increased from 0% to 1%. The maximum decrease in viscosity was due to 1% ascorbic acid, and the minimum decrease was due to 0.5% malic acid.

The study of rheological properties is essential for the application of polysaccharide gums as a food additive, especially in the production of foods which involves certain rheological properties, for instance as stabilizer and thickener (Safdar et al., 2023). Rheology is fundamental for the characterization of the materials submitted to different flow conditions (Mota and Pereira, 2022). Previous studies have reported that the conformational structure of xanthan gum depended on the pH of the media (Bak and Yoo, 2023). As the edible organic acids (malic acid, citric acid, ascorbic acid, and tartaric acid) used in this study are of the most abundant food acids, so study on the solution of xanthan gum in the presence of these organic acids can also shed light on its behavior in real food systems.

2. Material and Methods

2.1. Materials

Xanthan gum powder (food grade) was purchased from FuFeng Co. (China). In this research, organic acid including ascorbic, citric, malic, and tartaric were purchased in powder form (China) and dissolved in distilled water.

2.2. Preparation of xanthan gum containing acids

Two concentrations of each organic acid (malic acid, citric acid, ascorbic acid, and tartaric acid), 0.5% and 1%, were prepared, and the distilled water sample was considered as the control sample (0% acid). The xanthan gum solutions were provided by solving the gum powder (0.20%, w/v) in distilled water and different concentrations of ascorbic, citric, malic, and tartaric acids solutions using a magnetic stirrer. The provided dispersions were stored at 20°C for 1 h to complete the xanthan gum hydration process.

2.3. Flow behavior

Rheological data are important indicators of quality and success in industrial applications of xanthan gum (Palaniraj and Jayaraman, 2011). Flow behavior of the samples was evaluated using a viscometer (Brookfield, DV2T, RV, USA) at 20°C with an UL Adapter Kit. The apparent viscosity and shear stress (SS) of xanthan gum dispersions at various shear rates (12.2-171.2 s^{-1}) were measured (Salehi et al., 2023).

2.4. Mathematical modeling

The data obtained from previous stage (viscometer) were fitted to Power law, Bingham, Herschel-Bulkley, and Casson equations (Salehi and Inanloodoghouz, 2023). In this research, these models were used to match the SS and shear rates (SR) results of the xanthan gum dispersions containing edible organic acids. The experimental results were correlated for ease of use in rheological studies while maintaining appropriate accuracy using the function cftool (Curve Fitting Tool) in Matlab software (version R2012a).

2.5. Statistical analysis

Each experiment was carried out at least three times. The results were expressed as the mean ± standard deviation. SPSS software (version 21, Endicott, NY, USA) was used to run one-way ANOVAs at a probability level of 0.05 ($p < 0.05$). Duncan's multiple range test was used to compare mean values of various parameters.

3. Results and Discussion

3.1. Flow behavior

The industrial applications of xanthan gum depend on its rheological behavior in aqueous solutions. Xanthan gum is widely used as a thickening, stabilizing, gelling, and suspending and flocculating agent in the food and pharmaceutical industries. The rheological properties of this polymer impact industrial operations including mixing, transporting, pumping, and other processes involving fluid flow (Cevoli et al., 2013; Cancella et al., 2024). Figure 1 displays how the viscosity of xanthan gum dispersion change when the shear is applied at different speeds. It can be seen that the apparent viscosity of xanthan gum dispersion become less when it is stirred faster (shear-thinning behavior). In this study, the apparent viscosity of the sample containing 0.5% ascorbic acid reduced from 64.6 mPa.s to 20.2 mPa.s with the SR increased from 12.2 s^{-1} to 171.2 s^{-1}.

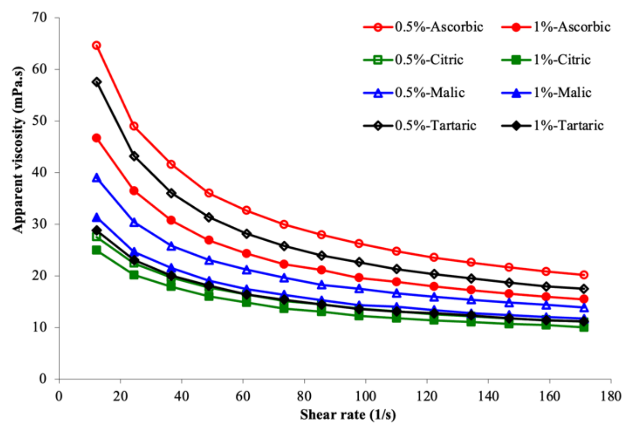


Fig. 1. Flow curves of xanthan gum in different dispersion media containing edible organic acids at 20 °C.

The rheological behavior and structural strength of xanthan gum dispersions were highly dependent on the pH value (Hayta et al., 2020). Also, the solution viscosity of xanthan gum decreases more at low concentrations (about 0.2%) than at high concentrations (about 0.5%) as the pH changes (Nsengiyumva and Alexandridis, 2022). The influence of organic acids on the apparent viscosity of xanthan gum dispersion is shown in Figure 2. The addition of organic acids to xanthan gum dispersion reduces its viscosity. This behavior was observed for all organic acids. The apparent viscosity of the xanthan gum dispersions reduced as the organic acids concentration increased from 0 to 1 %. The highest decrease in viscosity was related to 1% citric acid and the lowest was related to 0.5% ascorbic acid. The results show that when the citric acid concentration increased to 1%, the apparent viscosity of the xanthan gum dispersion reduced from 67.13 mPa.s to 15.02 mPa.s (SR=61 s^{-1}). Martins et al. (2023) reported similar result that the flow behavior of xanthan gum and carboxymethyl cellulose decreased under strongly acidic condition. Agoub et al. (2007) demonstrated that the degradation of pyruvate group in xanthan gum under strongly acidic condition, resulting in stabilizing helical conformation.

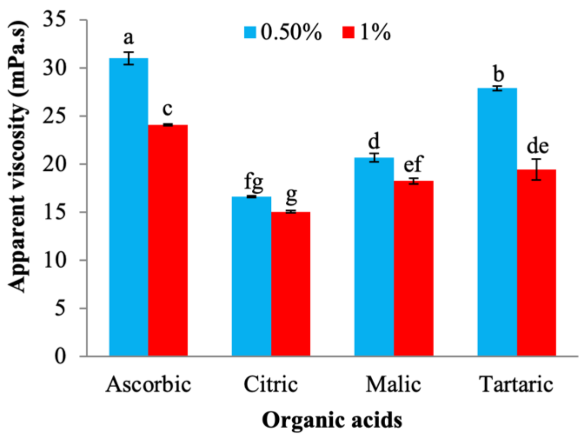


Fig. 2. Impact of organic acids on the apparent viscosity of xanthan gum dispersion (shear rate=61). Different lowercase letters denote significant differences ($p<0.05$) between different acids.

3.2. Power law model

As described by Mota and Pereira (2022), the Power law model is a suitable model to describe the behavior of viscosity for Non-Newtonian fluids and can be described in terms of SS or apparent viscosity. The Power law model fits very well ($r > 0.916$, SSE<1.589, and RMSE<0.364) the pseudoplastic behavior of aqueous solutions of xanthan gum containing edible organic acids in the range of SR from 12.2 to 171.2 s^{-1} (Table 1). The rheological behavior of xanthan gum and diutan gum aqueous solutions was studied by Mota and Pereira (2022). Their results showed that the Power Law model fits very well the pseudoplastic behavior of aqueous solutions of xanthan and diutan gums.

The consistency index and the flow behavior index were determined by fitting the flow curves by Ostwald-De-Waele model (Power law) to each gum solution (Cancella et al., 2024). The presence of organic acids in the solution containing xanthan gum had a significant effect on the consistency coefficient and flow behavior index changes of the gum dispersion (Figure 3). The impact of organic acids on the consistency coefficient of xanthan gum dispersion is reported in Figure 3. It was observed that the addition of organic acids had a significant influence on the rheological parameters of solutions. The consistency coefficient of the samples reduced when the acid percent was increased. The sample containing 1% citric acid had the lowest consistency coefficient and the sample containing 0.5% ascorbic acid had the highest consistency coefficient. The results show that when the citric acid concentration increased from 0 (control sample) to 1%, the consistency coefficient of the xanthan gum dispersion reduced from 1.126 Pa.s^n to 0.064 Pa.s^n. Hayta et al. (2020) and Dogsa et al. (2014) revealed that consistency coefficient of xanthan gum-galactomannan mixture and apparent viscosity of carboxymethyl cellulose decreased at low pH level.

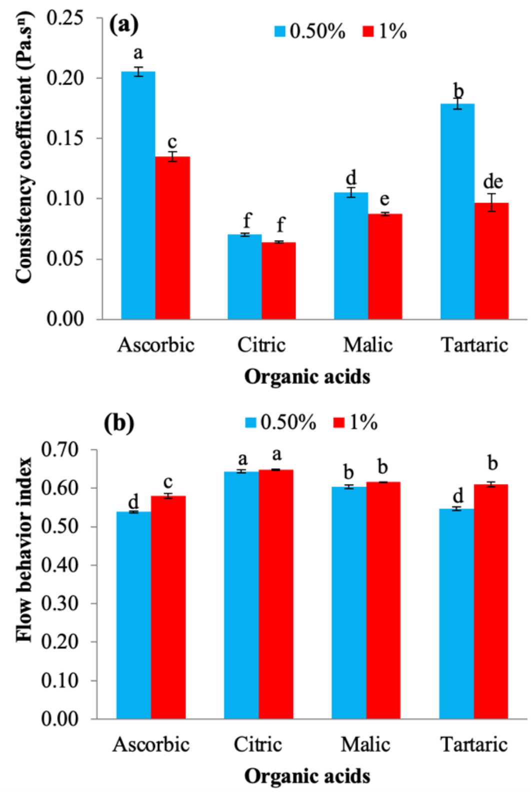


Fig. 3. Impact of organic acids on the consistency coefficient (a) and flow behavior index (b) of xanthan gum dispersion (Power law model). Different lowercase letters denote significant differences ($p<0.05$) between different acids.

The study of rheology of aqueous solution of xanthan gum can contribute to better understanding the flow behavior of this biopolymer that has many industrial and scientific applications. The impact of organic acids on the flow behavior index of xanthan gum dispersion is reported in Figure 3. The flow behavior index of the samples increased when the acid concentration was increased (decreases in shear-thinning behavior). The sample containing 1% citric acid had the highest flow behavior index and the sample containing 0.5% ascorbic acid had the lowest flow behavior index. The results show that when the citric acid concentration increased to 1%, the flow behavior index of the xanthan gum dispersion increased from 0.302 to 0.648. The alteration within the consistency coefficient and flow behavior index of the xanthan gum dispersion may be due to the structural changes of the gum in the presence of different acids.

3.3. Bingham model

The experimental values of SS versus SR for xanthan gum dispersion containing edible organic acids were fitted to the Bingham model and the constant coefficients of this equation were calculated. The values of SSE, r, and RMSE for xanthan gum dispersions were between 0.038 and 3.341, 0.814 and 0.993, and 0.056 and 0.528, respectively Table 2.

Table 1. Values of statistical parameters of the Power law model for estimating shear stress data.

Organic acid	Acid concentration	Sum of squared error (SSE)	Correlation coefficient (r)	Root mean square error (RMSE)
Ascorbic	0.5%	0.0020	0.9998	0.0127
Citric		0.0052	0.9984	0.0191
Malic		0.0015	0.9997	0.0105
Tartaric		0.0021	0.9997	0.0130
Ascorbic	1%	0.0020	0.9996	0.0129
Citric		0.1084	0.9662	0.0616
Malic		0.0013	0.9996	0.0104
Tartaric		0.0012	0.9997	0.0099

Table 2. Values of statistical parameters of the Bingham model for estimating shear stress data.

Organic acid	Acid concentration	Sum of squared error (SSE)	Correlation coefficient (r)	Root mean square error (RMSE)
Ascorbic	0.5%	0.1864	0.9763	0.1243
Citric		0.0495	0.9839	0.0645
Malic		0.0794	0.9821	0.0810
Tartaric		0.1539	0.9764	0.1131
Ascorbic	1%	0.1084	0.9803	0.0950
Citric		0.1532	0.9505	0.0974
Malic		0.0563	0.9835	0.0684
Tartaric		0.0665	0.9837	0.0740

The impact of organic acids on the Bingham yield stress parameter (τ_{0B}) of xanthan gum dispersion is reported in Figure 4. The Bingham yield stress of the samples reduced when acids percent was increased. The dispersion containing 1% citric acid had the lowest yield stress (0.326 Pa) and the sample containing 0.5% ascorbic acid had the highest yield stress (0.867 Pa). The results show that when the citric acid percent increased from 0 (control sample) to 1%, the Bingham yield stress of the xanthan gum dispersion significantly decreased from 2.047 Pa to 0.326 Pa ($p<0.05$).

At pH values below 4, the carboxylate groups are converted from ionized to non-ionized groups, which affects the electrostatic repulsion among xanthan gum side chains and reduces the viscosity of the dispersion (Nsengiyumva and Alexandridis, 2022). In addition, the impact of organic acids on the Bingham plastic viscosity (η_B) of xanthan gum dispersion is reported in Figure 4. The plastic viscosity of the samples reduced when acids percent was increased. The dispersion containing 1% citric acid had the lowest plastic viscosity (0.009 Pa.s) and the sample containing 0.5% ascorbic acid had the highest plastic viscosity (0.015 Pa.s). The results show that when the citric acid concentration increased to 1%, the Bingham plastic viscosity of the xanthan gum dispersion significantly reduced from 0.016 Pa.s to 0.009 Pa.s ($p<0.05$).

3.4. Herschel-Bulkley model

The experimental values of SS versus SR for xanthan gum dispersion were fitted to the Herschel-Bulkley model and the constant coefficients of this equation were calculated. The values of SSE, r, and RMSE for xanthan gum dispersion ranged from 0.001-1.597, 0.900-0.999, and 0.007-0.960, respectively Table 3.

Many nutritionists and researchers believe that thickened foods used to treat dysphagia need to be more accurately characterized and described based on their rheological properties. It is becoming increasingly common, for example, for thickened foods to be mentioned to and discussed in many articles explaining concepts such as viscosity, yield stress, and non-Newtonian fluids (Wen et al., 2024). Based on the Herschel-Bulkley model, all xanthan gum dispersions demonstrated shear-thinning behavior, described by the flow behavior index lower than 0.654 (Figure 5). The results of the Herschel-Bulkley model showed that the values of the yield stress were between 1.31×10^{-11} Pa and 2.58×10^{-5} Pa.

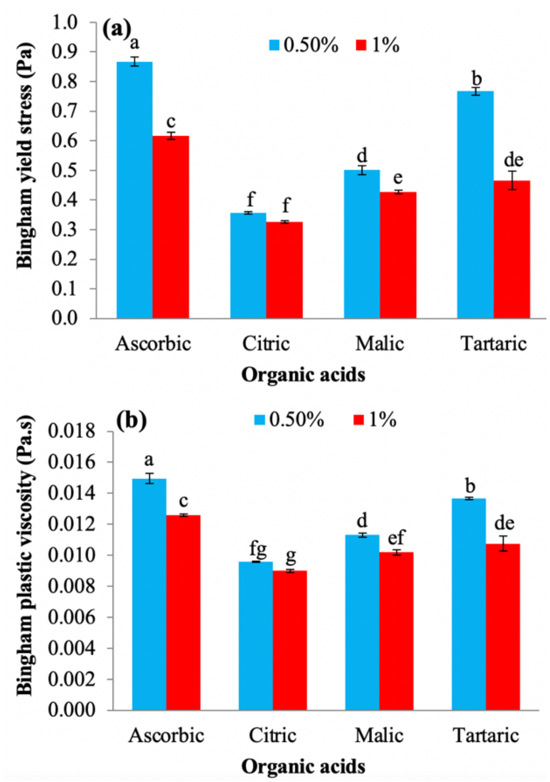


Fig. 4. Impact of organic acids on the Bingham yield stress (a) and Bingham plastic viscosity (b) parameters of xanthan gum dispersion (Bingham model). Different lowercase letters denote significant differences ($p<0.05$) between different acids.

Table 3. Values of statistical parameters of the Herschel-Bulkley model for estimating shear stress data.

Organic acid	Acid concentration	Sum of squared error (SSE)	Correlation coefficient (r)	Root mean square error (RMSE)
Ascorbic	0.5%	0.0020	0.9998	0.0129
Citric		0.0052	0.9984	0.0191
Malic		0.0015	0.9997	0.0105
Tartaric		0.0021	0.9997	0.0130
Ascorbic	1%	0.0020	0.9996	0.0132
Citric		0.1084	0.9662	0.0642
Malic		0.0013	0.9996	0.0105
Tartaric		0.0012	0.9997	0.3266

Table 4. Values of statistical parameters of the Casson model for estimating shear stress data.

Organic acid	Acid concentration	Sum of squared error (SSE)	Correlation coefficient (r)	Root mean square error (RMSE)
Ascorbic	0.5%	0.0552	0.9930	0.0675
Citric		0.0177	0.9944	0.0374
Malic		0.0239	0.9946	0.1969
Tartaric		0.0466	0.9928	0.0621
Ascorbic	1%	0.0322	0.9942	0.0518
Citric		0.1229	0.9613	0.0775
Malic		0.0162	0.9956	0.0367
Tartaric		0.0190	0.9953	0.0396

Xanthan gum is compatible with high salt concentrations and is stable over range of temperatures and pH. However, these parameters are known to influence the molecular structure of the polyelectrolyte, causing structural transitions that affect their rheological behavior (Martins et al., 2023). The impact of organic acids on the consistency coefficient (Herschel-Bulkley model) of xanthan gum dispersion is reported in Figure 5. The consistency coefficient of the samples reduced when acids percent was increased. The results show that when the tartaric acid concentration increased from 0.5% to 1%, the consistency coefficient of the xanthan gum dispersion reduced from 0.179 Pa.s^n to 0.097 Pa.s^n. The sample containing 1% citric acid had the lowest consistency coefficient and the sample containing 0.5% ascorbic acid had the highest consistency coefficient.

The consistency coefficient represents the viscosity whereas and flow behavior index reveals the shear-shinning characteristics of the materials (Safdar et al., 2023). The impact of organic acids on the flow behavior index (Herschel-Bulkley model) of xanthan gum dispersion is reported in Figure 5. The flow behavior index of the samples increased when acid concentration was increased (decreases in shear-thinning behavior). The sample containing 1% citric acid had the highest flow behavior index (0.648) and the sample containing 0.5% ascorbic acid had the lowest flow behavior index (0.539). The results show that when the tartaric acid percent increased from 0.5% to 1%, the flow behavior index of the xanthan gum dispersion increased significantly from 0.547 to 0.610 ($p<0.05$) (decreases in shear-thinning behavior).

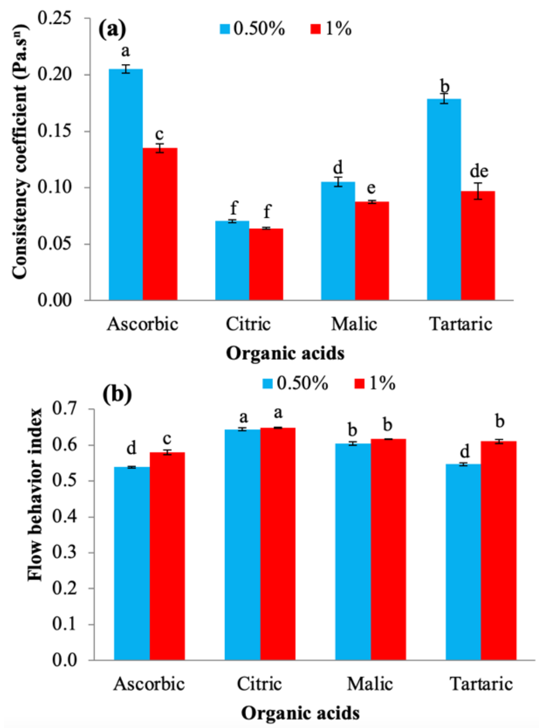


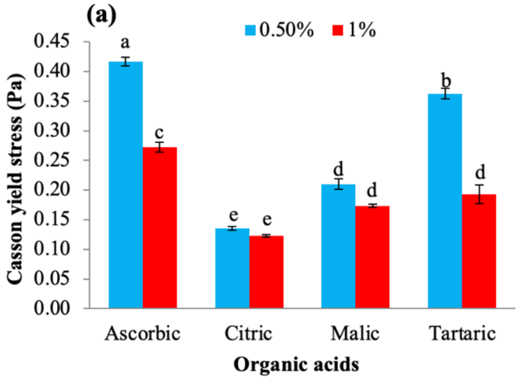
Fig. 5. Impact of organic acids on the consistency coefficient (a), and flow behavior index (b) parameters of xanthan gum dispersion (Herschel-Bulkley model). Different lowercase letters denote significant differences ($p<0.05$) between different acids.

3.5. Casson model

The experimental values of SS versus SR for xanthan gum dispersion containing edible organic acids were fitted to the Casson model and the constant coefficients of this equation were calculated. The values of SSE, r, and RMSE for xanthan gum dispersion were between 0.012 and 2.420, 0.869 and 0.998, and 0.031 and 0.508, respectively (Table 4). The impact of organic acids on the Casson yield stress (τ_{0C}) of xanthan gum dispersion is reported in Figure 6.

The Casson yield stress of the samples reduced when acids percent was increased. The dispersion containing 1% citric acid had the lowest yield stress (0.122 Pa) and the sample containing 0.5% ascorbic acid had the highest yield stress (0.416 Pa). The results show that when the tartaric acid concentration increased from 0.5%

to 1%, the Casson yield stress of the xanthan gum dispersion significantly reduced from 0.363 Pa to 0.193 Pa ($p<0.05$).



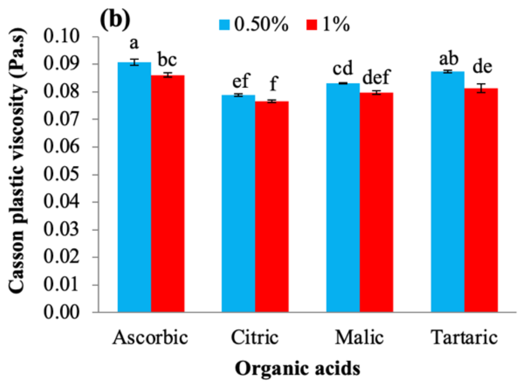


Fig. 6. Impact of organic acids on the Casson yield stress (a) and Casson plastic viscosity (b) parameters of xanthan gum dispersion (Casson model). Different lowercase letters denote significant differences ($p<0.05$) between different acids.

In addition, the impact of organic acids on the Casson plastic viscosity (ηC) of xanthan gum dispersion is reported in Figure 6. The plastic viscosity of the samples decreased when acids concentration was increased. The dispersion containing 1% citric acid had the lowest plastic viscosity (0.077 Pa.s) and the sample containing 0.5% ascorbic acid had the highest plastic viscosity (0.091 Pa.s). The results show that when the tartaric acid concentration increased from 0.5% to 1%, the Casson plastic viscosity of the xanthan gum dispersion significantly reduced from 0.087 Pa.s to 0.081 Pa.s ($p<0.05$).

4. Conclusion

In the current work, we examined the impact of edible organic acids (ascorbic, citric, malic, and tartaric) at two concentrations (0.5, and 1 %) on the viscosity and rheological parameters of xanthan gum dispersion. Xanthan gum dispersion showed the shear-thinning flow behavior. As the selected organic acids concentration increased, the viscosity of solution decreased, and the highest decrease in viscosity was related to citric acid. The apparent viscosity values of the xanthan gum dispersions (0.2% w/v) prepared in organic acids model system (0.5 %) were determined to be 31.0, 16.6, 20.7 and 27.9 mPa.s for ascorbic, citric, malic, and tartaric acids, respectively, at the shear rate of 61 s^{-1} and 20°C. Various equations were used to fit rheological data of xanthan gum dispersions, and it was observed that the Power law model was the best because the correlation coefficient (r) values were higher than 0.916 and RMSE values between 0.007 and 0.364. The consistency coefficient (Power law and Herschel-Bulkley models) of the samples reduced when acids percent was increased. But, the flow behavior index values (Power law and Herschel-Bulkley models) of the samples increased when selected organic acids percent was increased.

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Conflict of interest

The authors declare that there is no conflict of interest.

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