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Research Article

Impact of Ultrasound Intensity and Duration on the Rheological Behaviour of Sodium Alginate Solution

Fakhreddin Salehi[1*], Moein Inanloodoghouz[2]

1. Associate Professor, Department of Food Science and Technology, Faculty of Food Industry, Bu-Ali Sina University, Hamedan, Iran

2. MSc Student, Department of Food Science and Technology, Faculty of Food Industry, Bu-Ali Sina University, Hamedan, Iran

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Abstract

One of the recent uses of sonication is to change the composition and structural properties of hydrocolloids. This study aimed to examine the impacts of sonication at different intensities (0, 75, and 150 W) and time (0, 5, 10, 15, and 20 min) on the viscosity and rheological properties of sodium alginate solution. The results showed that the apparent viscosity of sodium alginate solution (control sample) decreased from 0.075 to 0.032 Pa.s with increasing shear rate from 12.2 to 134.5 s^{-1}. Also, the apparent viscosity of sodium alginate solution decreased from 0.044 to 0.019 Pa.s with enhancing the sonication time from 0 to 20 min (shear rate=61 s^{-1}, 150 W). Various rheological equations (Power law, Bingham, Herschel-Bulkley, Casson, and Vocadlo) were employed to fit the empirical values, and the results confirmed that the Power law model was the best fit to describe the flow behaviour of sodium alginate solution. The consistency coefficient of sodium alginate solution significantly decreased from 0.216 Pa.s^n to 0.151 Pa.s^n ($p<0.05$) with enhancing sonication time from 0 to 20 min. Furthermore, the consistency coefficient of sodium alginate solution decreased significantly ($p<0.05$) while the ultrasonic power enhanced. Flow behaviour index of sodium alginate solution enhanced significantly ($p<0.05$) while the intensity and duration of ultrasound treatment enhanced.

Keywords: Consistency coefficient, Flow behaviour index, Rheological models, Ultrasound.

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* Corresponding author: F.Salehi@Basu.ac.ir

1. Introduction

Hydrocolloids (gums) are used alone, or quite often in combination as thickeners, stabilizers and gelling agents for food applications. They absorb large quantities of water and can form polymeric networks, imparting various textural and mouthfeel effects to foods [1]. Sodium alginate is one of the widely used hydrocolloids as an enzyme carrier and food additives, which is made up of mannuronic acid and guluronic acid as two kinds of structure units, forming a kind of no linear block copolymer of branched chain [2]. This gum is non-toxic, eco-friendly and widely available, which has spurred the food industry to utilize sodium alginate as an edible coating or a thickening, stabilizing, suspending, emulsifying, and gelling agent. The aqueous solution of this gum produces high viscosity and pseudoplastic behavior [3-5]. For example, Jansrimanee and Lertworasirikul [3] and Matuska et al. [6] used sodium alginate solution for the edible coating of osmotic dehydrated pumpkin and strawberries, respectively.

The past few decades have seen a dramatic rise in the applications of high-intensity, or power, ultrasound in chemistry, with a range of synthetic procedures and process methods having found to benefit from sonication [7]. The main mechanism responsible for the effect of ultrasound on liquid systems is the physical forces generated by acoustic cavitation such as microjets, shear, shockwaves, and acoustic streaming [8, 9]. One of the beneficial applications of ultrasound is for the polymers such as hydrocolloids (gums) degradation [10-14]. The results of Salehi et al. [15] showed that the flow behavior index of carboxymethyl cellulose (CMC) solution significantly increased with increasing sonication time. In addition, the consistency coefficient of CMC solution significantly decreased with increasing sonication time. The results of Li et al. [10] suggest that sonication is an effective means of polysaccharide (konjac glucomannan) degradation without considerable structural destruction. The effects of ultrasound with different ultrasonic frequencies on the properties of sodium alginate were investigated by Feng et al. [16]. Their results showed that the molecular weight of the ultrasound treated sodium alginate gradually increased while the molecular number of sodium alginate increased and then decreased with the increase of the ultrasonic frequency.

Prolonged exposure of solutions of macromolecules to high-energy ultrasonic waves produces a permanent reduction in viscosity [17]. Besides, the application of ultrasound to modify biopolymers is increasingly studied. Therefore, the aim of this study was to examine the influence of ultrasonic treatments at different intensities (at 3 levels of 0, 75, and 150 W) and time (at 5 levels of 0, 5, 10, 15, and 20 min) on the rheological properties, viscosity, consistency coefficient, and flow behavior index of sodium alginate solution.

2. Materials and methods

2.1. Preparation of gum solutions

Sodium alginate powder (food grade) was purchased from Qingdao bright moon seaweed group Co., Ltd. (China). The sodium alginate solutions (0.30%, w/v) were provided by solving powder of gum in distilled water using a magnetic stirrer. Provided sodium alginate solution was stored for 1 h at room temperature to complete hydration of the gum.

2.2. Sonication process

To apply the sonication treatments on the sodium alginate solution, the tank of the ultrasonic bath (40 kHz, vCLEAN1-L6, Backer, Iran) was filled with 2 L of sodium alginate solution at 25°C (Figure 1). Ultrasonic waves were applied to the solution inside the device at three power levels (0, 75, and 150 watts) and at five different times (0, 5, 10, 15, and 20 min).

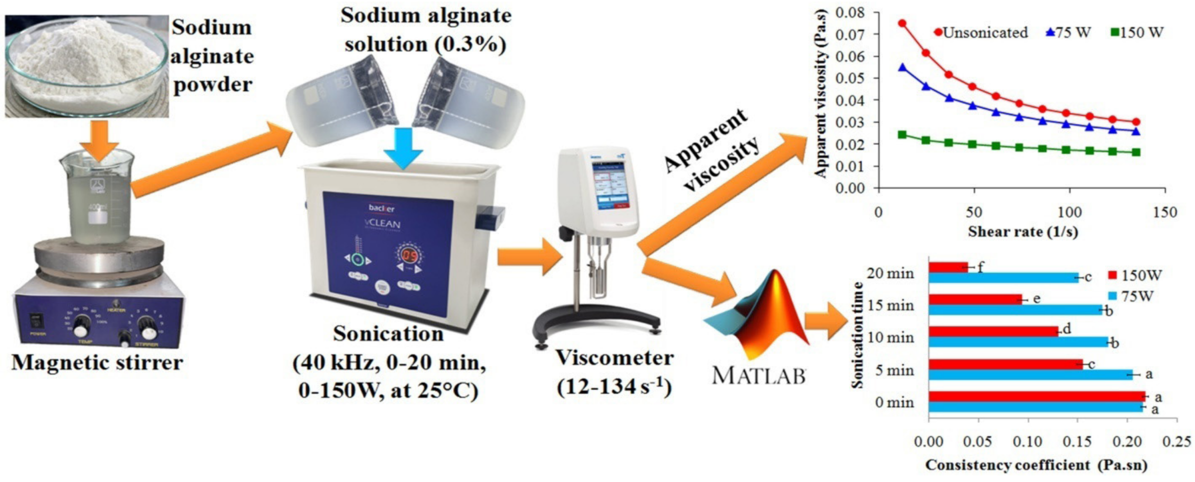

Fig. 1. Schematic diagram of ultrasonic treatment of sodium alginate solution

2.3. Apparent viscosity

The apparent viscosity of unsonicated and sonicated sodium alginate solutions was calculated using a rotational viscometer (Brookfield, DV2T, RV, USA) after each treatment. The viscosity and shear stress of sodium alginate solutions at different shear rates (12-134.5 s^{-1}) were studied using UL Adapter Kit at 25°C.

2.4. Rheological behavior

Various viscous flow models, including Power law, Bingham, Herschel-Bulkley, Casson, and Vocadlo, were used to match the empirical shear stress and shear rate data of the sonicated and unsonicated sodium alginate solutions [15]. The fit of the experimental data to the rheological models was performed using Matlab software (version R2012a).

2.5. Statistical analysis

The evaluation of viscosity and rheological parameters of sodium alginate solutions were conducted in a 3×5×3 (ultrasonic power, ultrasound treatment time, and three replicates) factorial design in a completely randomized design. Besides, the means were compared using Duncan's test at p-value<0.05 (using SPSS software version 21).

3. Results and discussion

3.1. Apparent viscosity

Unlike chemical or thermal decomposition, it is believed that ultrasonic degradation is a non-random process, with cleavage taking place roughly at the center of the molecule and with larger molecules degrading the fastest [17]. The impacts of sonication power on the apparent viscosity of sodium alginate solutions as a function of shear rate are demonstrated in Figure 2. The application of ultrasound to the sodium alginate solution reduces its viscosity. This behavior was observed at all conditions and 150 W power leading to a greater reduction in gum viscosity. The results demonstrated that the average apparent viscosity of sodium alginate solution decreased from 0.045 to 0.018 Pa.s with increasing ultrasonic power from 0 to 150 W (20 min). The decrease in the viscosity of gums when exposed to ultrasonic waves can be attributed to the breakdown of their large molecular structures into smaller shapes due to the cavitation effect [11, 18]. Li et al. [10] reported that when konjac glucomannan solution (1%, w/v) was exposed to ultrasonic waves, the apparent viscosity reduced quickly from about 50 Pa.s to a trivial level within 10-20 minutes.

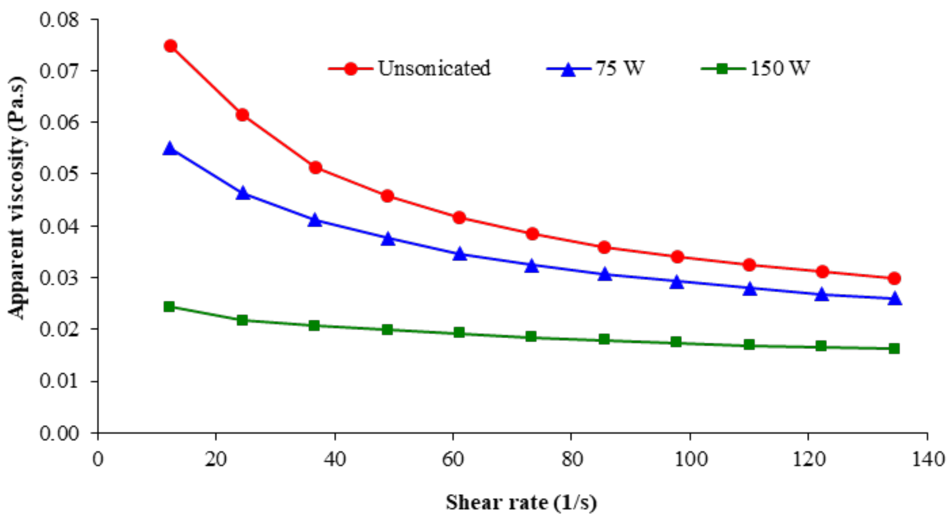

Fig. 2. Impact of sonication power on the apparent viscosity of sodium alginate solution (sonication time=20 min).

In the case of pseudoplastic fluids, high-shear microcurrents lead to reduced viscosity. As reported by Cui [19], the viscosity of the gum dispersion reduced with enhancing the shear rate as the number of entangled chains decreased at high shear rates. The shear rate dependence of the apparent viscosity of sodium alginate solutions under different conditions is shown in Figure 2. It is seen that the apparent viscosity of sodium alginate solution decreases as the shear rate increases (pseudoplastic behavior). The apparent viscosity decreased markedly from 0.064 Pa.s to 0.030 Pa.s with the shear rate increased from 12.2 to 134.5 s^{-1} (sonication power=75W and time=10 min). Consistent with the results of this study, Feng et al. [16] confirmed that the sodium alginate solution treated by ultrasound at 50 kHz tended to be closer to a Newtonian behavior, while the untreated and treated sodium alginate solution exhibited pseudoplastic behavior. In addition, Sarraf et al. [20] confirmed that the viscosity of xanthan gum solution reduced with enhancing shear rate, indicating pseudoplastic behavior. Farizadeh and Abbasi [21] studied the flow behavior of sonicated and unsonicated locust bean gum solutions. Their results demonstrated that the apparent viscosity of all samples reduced as the shear rate enhanced, showing their pseudoplastic behavior.

Sonochemical degradation of polymers has proved to be an attractive process especially considering the fact that there are no changes in the chemical nature of the polymer and the reduction in molecular weight (also the intrinsic viscosity) is simply by splitting the most susceptible chemical bond [7]. The impact of sonication time on the apparent viscosity of sodium alginate solution as a function of shear rate and sonication power (75 and 150 W) is shown in Figure 3. As can be seen in this figure, the viscosity of the sodium alginate solutions decreased with increasing ultrasonic treatment time. At all shear rates, the viscosity of the unsonicated sodium alginate solution was higher than that of the sonicated samples. Compared with the sonicated samples, the shear rate has a greater influence on the unsonicated sample. The highest apparent viscosity involved the control sample and the lowest apparent viscosity related to the samples sonicated for 20 min. The effect of ultrasonic waves on the rheological properties of locust gum solution was investigated by Farizadeh and Abbasi [21]. Their results demonstrated that with increasing ultrasonic treatment time, the viscosity of the locust bean gum solution decreased.

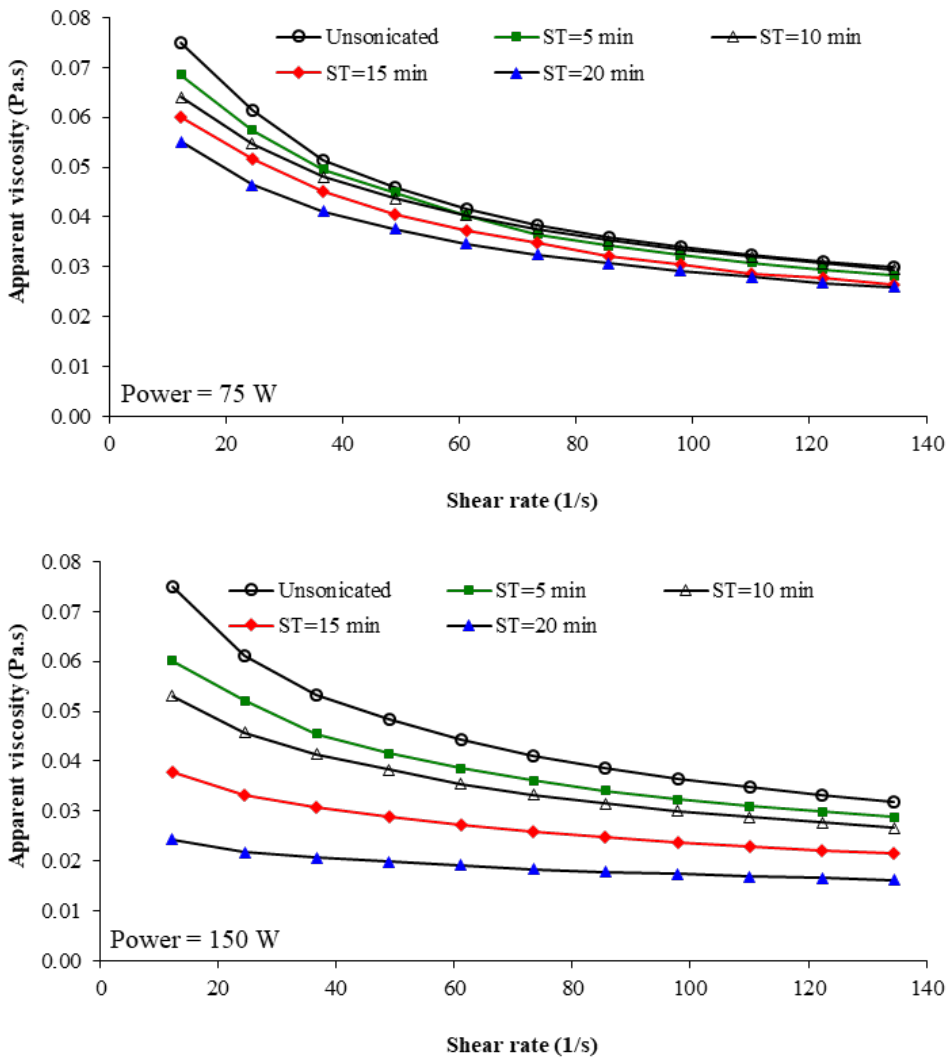

Fig. 3. Impact of sonication time (St) on the apparent viscosity of sodium alginate solutions.

3.2. Rheological model parameters

Rheological properties play an important role in process design. In several recent researches, ultrasound has been used for controlled degradation of natural gums resulting in changes in viscosity and rheological properties [10, 11, 21]. Table 1 demonstrates the predicted values and statistical parameters obtained to verify the fit of each rheological model to the observed shear rate/shear stress data of sodium alginate solution. The statistical parameters reported in this table show that Power law model (equation 1) is appropriate to describe the rheological behavior of untreated and treated sodium alginate solution.

$$\tau = k\dot{\gamma}^{n} \tag{1}$$

In Power law model, τ is the measured shear stress (Pa), k is the Power law consistency coefficient (Pa.s^n), $\dot{\gamma}$ is the shear rate (1/s) and n is the flow behavior index (dimensionless). In this study, Power law model showed a good fit with the maximum r-value and the minimum SSE (sum of squared error) and RMSE (root mean squared error) values for all conditions compared to that of the other models. The Power law was considered to be the appropriate model for characterizing the rheological behavior of many gum solutions [15].

Table 1. The predicted values and statistical parameters obtained to verify the fit of each rheological model.

Model name	Model constants	r	SSE	RMSE	Predicted apparent viscosity (Pa.s)										
Power law	k=0.2139 n=0.5966	0.9997	0.0049	0.0233	0.078	0.059	0.050	0.045	0.041	0.038	0.036	0.034	0.032	0.031	0.030
Bingham	τ_{0B}=0.8953 η_B=0.0241	0.9912	0.1695	0.1372	0.097	0.061	0.048	0.042	0.039	0.036	0.035	0.033	0.032	0.031	0.031
Herschel-Bulkley	τ_{0H}=3.2E-9 k_H=0.2139 n_H=0.5966	0.9997	0.0049	0.0234	0.078	0.059	0.050	0.045	0.041	0.038	0.036	0.034	0.032	0.031	0.030
Casson	τ_{0C}=0.3787 η_C=0.1206	0.9972	0.0532	0.0769	0.088	0.060	0.049	0.044	0.040	0.037	0.035	0.033	0.032	0.031	0.030
Vocadlo	τ_{0V}=7.9E-9 n_V=0.5966 k_V=0.0753	0.9997	0.0049	0.0234	0.078	0.059	0.050	0.045	0.041	0.038	0.036	0.034	0.032	0.031	0.030
Experimental apparent viscosity (Pa.s)					0.074	0.060	0.051	0.045	0.041	0.038	0.035	0.034	0.032	0.031	0.030
Shear rate (1/s)					12.2	24.5	36.7	48.9	61.2	73.4	85.6	97.8	110.1	122.3	134.5

Where, r is correlation coefficient; SSE is sum of squared error; RMSE is root mean squared error.

The rheological behavior of untreated and treated sodium alginate solutions was fitted with the Power law model. The calculated constant coefficients of the Power law equation include k and n are reported in Table 2 together with the corresponding statistical error values (r and RMSE) for all conditions. Mean values of SSE, RMSE and r for all sodium alginate solutions ranged from 0.0004-0.0230, 0.0065-0.0505, and 0.9986-0.9999, respectively. Based on the Power law model, all sodium alginate solutions demonstrated marked pseudoplastic behavior, described by the flow behavior index values lower than 0.87 under all conditions. The results confirmed that the consistency coefficient values were between 0.024 and 0.225 $Pa.s^n$ and the n ranged from 0.58 to 0.87 (indicating the pseudoplastic nature of sodium alginate solution).

Table 2. Impact of sonication power and treatment time on the Power law model parameters.

Sample	Sonication power (W)	Sonication time (min)	k ($Pa.s^n$)	n	SSE	r	RMSE
1	75	0	0.2158	0.5977	0.0064	0.9997	0.0266
2	75	5	0.2059	0.5993	0.0113	0.9994	0.0328
3	75	10	0.1814	0.6301	0.0130	0.9994	0.0378
4	75	15	0.1752	0.6256	0.0138	0.9992	0.0377
5	75	20	0.1509	0.6496	0.0081	0.9995	0.0294
6	150	0	0.2181	0.6049	0.0078	0.9996	0.0290
7	150	5	0.1551	0.6529	0.0083	0.9996	0.0303
8	150	10	0.1309	0.6710	0.0074	0.9995	0.0278
9	150	15	0.0946	0.7169	0.0047	0.9997	0.0214
10	150	20	0.0396	0.8206	0.0012	0.9998	0.0110

Where, SSE is sum of squared error; r is correlation coefficient; RMSE is root mean squared error.

The consistency coefficient and flow behavior index are typical rheological parameters used in the Power law model. Different authors have concluded that consistency coefficient is a strong function of the solution and the temperature concentration, whereas flow behavior index does not have a strong dependence on the concentration and temperature of the polymeric solution [22]. The impact of sonication power on the consistency coefficient of sodium alginate solutions is shown in Figure 4. From the results, sonication considerably decreased the consistency coefficient of sodium alginate solutions. The consistency coefficient of the sodium alginate solutions was found to be greater than 0.024 $Pa.s^n$ and was significantly affected by sonication treatment. This behavior was observed at all conditions and 150 W power leading to a greater reduction in the consistency coefficient. The results demonstrated that the consistency coefficient value of sodium alginate solution decreased from 0.218 to 0.040 $Pa.s^n$ with enhancing ultrasonic power from 0 to 150 W (sonication period=20 min).

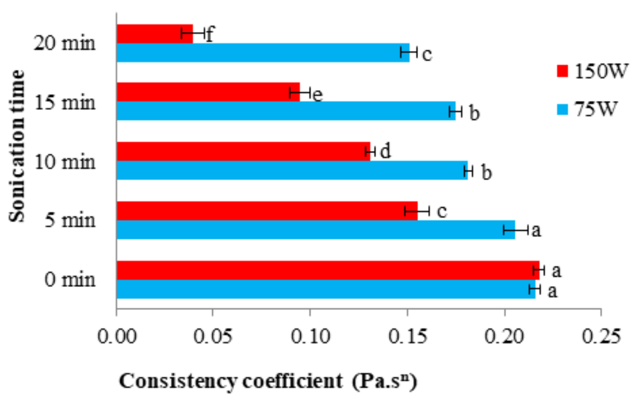

Fig. 4. Impact of sonication power and treatment time on the consistency coefficient of sodium alginate solutions.

Various letters near the bars denote for statistically significant differences at $p<0.05$.

As can be seen in Figure 4, the consistency coefficient value of the samples significantly decreased with increasing ultrasonic treatment time ($p<0.05$). The highest k value involved the unsonicated sample and the lowest k value related to the samples sonicated for 20 min. Effect of sonication on the rheological behavior of sugar beet pectin was examined by Yang et al. [23]. Their results showed that the values of viscosity and consistency coefficient of the samples reduced after sonication. In addition, the apparent viscosity decreases as the shear rate increases over the test range and shows a pseudoplastic fluid characteristic.

Ultrasound modification is proposed as a superior strategy to obtain good hydrocolloids with useful physicochemical characteristics and molecular structure [12]. It is clear from the Power law model that a non-Newtonian fluid ($n \neq 1$) with pseudoplastic behavior has a value of n lower than 1 [24]. The impact of sonication power on the flow behavior index of sodium alginate solutions is shown in Figure 5. The application of ultrasound to the sodium alginate solution increases its n value (decreases in pseudoplastic behavior). This behavior was observed at all condition and 150 W power leading to greater increase in the n values. The results demonstrated that the n value of sodium alginate solution enhanced from 0.605 to 0.821 with increasing ultrasonic power from 0 to 150 W (sonication period=20 min).

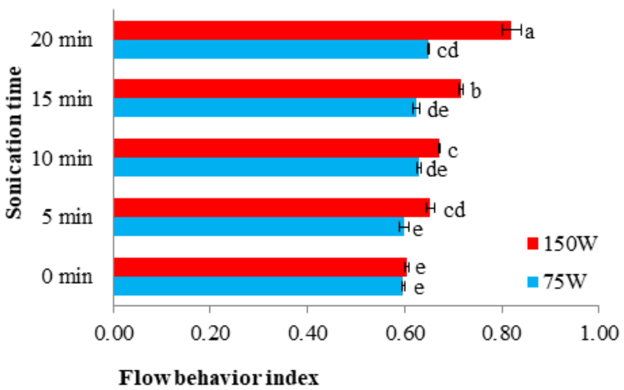

Fig. 5. Impact of sonication power and treatment time on the flow behavior index of sodium alginate solutions.

Various letters near the bars denote for statistically significant differences at $p<0.05$.

The impact of sonication time on the flow behavior index of sodium alginate solution is shown in Figure 5. As can be seen in this figure, the n values of the samples significantly increased (less shear thinning solution) with increasing ultrasonic treatment time ($p<0.05$). The lowest n value involved the unsonicated sample and the highest n value related to the samples sonicated for 20 min. In this study, the n value increased significantly from 0.598 to 0.650 with the sonication time increased from 0 to 20 min ($p<0.05$) (power=75W). The change in the k and n indexes of the sodium alginate solution may be due to the structural change of the sodium alginate during ultrasound treatment. Ultrasound energy is known to drive a number of physicochemical reactions that lead to altered functional characteristics of hydrocolloids in liquid food systems [18]. The influence of sonication on the rheological behavior and interactions of chitosan-sodium alginate solution was studied by Xu et al. [25]. Their results revealed that the sonication considerably reduces the viscosity and consistency coefficient and enhances the flow behavior index of the mixture, leading to a modification in the flow behavior from pseudoplastic to near-Newtonian behavior.

4. Conclusion

The sonication process has been commonly employed in the food industry due to its many physicochemical impacts. In the present study, the impact of ultrasonic intensities and durations on the rheological characteristics of sodium alginate solution was examined. This gum demonstrated the pseudoplastic flow behavior. The application of ultrasound to the sodium alginate solution reduces its viscosity. In addition, the viscosity of the sodium alginate solutions reduced with increasing ultrasonic treatment time. The Power law model was proved to be the most appropriate equation to explain the rheological behavior of sodium alginate solutions over the entire empirical range. The consistency coefficient value of the samples significantly decreased with increasing ultrasonic treatment time ($p<0.05$). The lowest n value involved the unsonicated sample and the highest n value related to the samples sonicated for 20 min. The findings of this study suggest that sonication offers a good opportunity to alter the physicochemical characteristics of sodium alginate solution and can significantly improve the rheological properties of this gum by sonication.

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مقاله پژوهشی

تاثیر شدت و مدت فراصوت بر رفتار رئولوژیکی محلول آلژینات سدیم

فخرالدین صالحی[۱*]، معین ایناللودوقوز[۲]

۱. دانشیار گروه علوم و صنایع غذایی، دانشکده صنایع غذایی، دانشگاه بوعلی سینا، همدان، ایران.

۲. دانشجوی کارشناسی ارشد گروه علوم و صنایع غذایی، دانشکده صنایع غذایی، دانشگاه بوعلی سینا، همدان، ایران.

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چکیده

یکی از کاربردهای اخیر فراصوت، تغییر ترکیب و خواص ساختاری هیدروکلوئیدها است. این مطالعه با هدف بررسی اثرات فراصوت در شدت‌های مختلف (۰، ۷۵ و ۱۵۰ وات) و زمان (۰، ۵، ۱۰، ۱۵ و ۲۰ دقیقه) بر ویسکوزیته و خواص رئولوژیکی محلول آلژینات سدیم انجام شد. نتایج نشان داد که با افزایش سرعت برشی از ۱۲/۲ به ۱۳۴/۵ بر ثانیه، ویسکوزیته ظاهری محلول آلژینات سدیم (نمونه شاهد) از ۰/۰۷۵ به ۰/۰۳۲ پاسکال ثانیه کاهش می‌یابد. همچنین، ویسکوزیته ظاهری محلول آلژینات سدیم با افزایش زمان اعمال فراصوت از صفر به ۲۰ دقیقه، از ۰/۰۴۴ به ۰/۰۱۹ پاسکال ثانیه کاهش یافت (سرعت برشی ۶۱ بر ثانیه، ۱۵۰ وات). معادلات رئولوژیکی مختلف (قانون توان، بینگهام، هرشل بالکلی، کاسون و وکادلو) برای برازش مقادیر تجربی استفاده شدند و نتایج تأیید کرد که مدل قانون توان بهترین برازش را برای توصیف رفتار جریان محلول آلژینات سدیم دارد. ضریب قوام محلول آلژینات سدیم با افزایش زمان فراصوت از ۰ تا ۲۰ دقیقه به طور معنی‌داری از $0/216\ Pa.s^n$ به $0/151\ Pa.s^n$ کاهش یافت ($p<0/05$). علاوه بر این، ضریب قوام محلول آلژینات سدیم با افزایش توان فراصوت به طور معنی‌داری کاهش یافت ($p<0/05$). شاخص رفتار جریان محلول آلژینات سدیم با افزایش شدت و مدت تیماردهی با فراصوت به طور معنی‌داری افزایش یافت ($p<0/05$).

واژگان کلیدی: **شاخص رفتار جریان، ضریب قوام، فراصوت، مدل‌های رئولوژیکی.**

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* نویسنده مسئول: F.Salehi@Basu.ac.ir