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Note

Structural and Rheological Characteristics of Alginate from Sargassum cristaefolium Extracted by Twin Screw Extruder

by Sugiono Sugiono

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Structural and Rheological Characteristics of Alginate from *Sargassum cristaefolium* Extracted by Twin Screw Extruder

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ABSTRACT

This study aimed to investigate the effects of process extrusion on the characteristics of *Sargassum cristaefolium* sodium alginate (SCSA) extracted using twin-screw extruder. Box-Behnken Design (BBD) from response surface methodology (RSM) was established to understand the effects of temperature, screw speed, and pH on the multiple-responses of alginate characteristics including intrinsic viscosity, yield, and molecular weight. The results revealed that temperature, screw speed, and pH significantly affected ($P < .05$) all responses. The optimum extraction condition was found at temperature of 58.18°C, screw speed of 77.99 rpm, and pH 10.11. At this condition, the response of residence time distribution was 7.07 ± 0.029 min, yield of 34.01 ± 0.12%, intrinsic viscosity of 460.13 ± 14.75 mL/g, and molecular weight of 217.94 ± 7.14 × 10^3 g/mol. This alginate had mannuronic acid to guluronic acid (M/G) ratio of 0.28, and the L-guluronic acid block was 0.78, which was higher than the D-mannuronic acid block. Rheological characterization of SCSA in aqueous solution was shear-thinning pseudoplastic, and alginate gel in 1 M $CaCl_2$ was more elastic than liquid.

KEYWORDS

Alginate; response surface methodology; rheology; M/G ratio; *Sargassum cristaefolium*; twin-screw extruder

Introduction

Alginate is a polysaccharide present in the matrix of brown algae cell wall. The quantity and quality of this polysaccharide is affected by species, season, growing location, type and age of tissue, and extraction methods (Andriamanantoanina and Rinaudo 2010; Bertagnolli et al. 2014; Draget and Taylor 2011; Fenoradosoa et al. 2010; Larsen et al. 2003; Leal et al. 2008). Alginate is comprised of linear polymers of β- (1-4)-D-mannuronate (M) and α-L-guluronate (G) at different proportions and monomer sequences (Larsen et al. 2003; Torres et al. 2007). The alginate blocks are formed from three blocks: two blocks of homopolymer sequence of D-mannuronic acid block (MM) and L-guluronic acid block (GG) and heteropolymer from guluronic and mannuronic acids (GM). The M/G ratio and the distribution of G and M blocks in the chain remarkably affected the physiological and rheological characteristics of alginate. Alginate acts as a thickener and gel formation in the presence of calcium ions (Gomez et al. 2009).

Classic alginate extraction method with batch process is ineffective as it is time consuming and requires a lot of reactants and solvents (Fertah et al. 2014; Torres et al. 2007). Extraction of alginate using microwave, supercritical CO_2, autohydrolysis, and ultrafiltration is expensive and difficult for industrial applications (Balboa et al. 2013; Quitain et al. 2013). A more effective extraction method is extrusion using twin screw extruder, as it only requires a short time and less solvent, less waste

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produced, and the process is safe (Vauchel et al. 2008a). Extrusion is a continuous process that combines pressure, shear stress, and temperature in the moving screw (Hernandez-Carmona et al. 2013; Kartika et al. 2010). This method has been widely applied to the lignocellulose pre-treatment process for bioethanol production and extraction of sunflower oil (Dufaure et al. 1999; Evon et al. 2013, 2007; Zheng and Rehmann 2014). The use of twin screw extruder for alginate extraction process has been performed by Baron et al. (2010), who focused on the effect of screw speed on residence time distribution (RTD) of algae in the screw channel.

Extrusion conditions (temperature, screw speed, and pH) affected the movement of algae in the screw channel, specific mechanical energy (SME), and physicochemical properties of alginate (Baron et al. 2010; Fertah et al. 2014; Hernandez-Carmona et al. 1999; Kartika et al. 2010; Torres et al. 2007). This current work is to optimize extrusion conditions (screw speed, temperature, and pH) for the alginate extraction from brown algae *Sargassum cristaefolium*. The alginate is characterized for structural and rheological characteristics and gel formation, as well.

Materials and methods

Materials

Sargassum cristaefolium brown algae was obtained from Poteran Island, Sumenep, Madura, Indonesia, in April 2016. Fresh brown algae was washed using tap water, submerged in 0.1% KOH for 1 h, and re-washed to remove residue. The brown algae was sun-dried, ground, and sieved through 60 mesh (Subaryono and Apriani 2010). The brown algae was submerged in 0.1% formaldehyde overnight, washed, and dried at 50°C for 6 h in cabinet dryer (Hernandez-Carmona et al. 1999; Wedlock and Fasihuddin 1990). All chemicals including KOH, formaldehyde, hydrochloric acid (HCl) 35%, ethanol 96%, Na_2CO_3 were technical grade, and reagent for analysis ($CaCl_2$, D_2O, H_2SO_4, and NaOH) were analytical grade.

Twin-screw extruder

The alginate extraction was performed using intermeshing corotating twin-screw extruder (Berto Industry BEX-DS-2256, serial number BC-0405-054-08-004), with capacity of 7 kg/h. Three thermocouples were set to produce heat. The barrel temperature was monitored on the control panel and reduced using an air compressor; diameter of die was 8 mm. The extruder was operated at screw speed of 0–180 rpm and auger speed (feed rate) of 0–35 rpm. Barrel and screw profile of twin-screw extruder is shown in Figure 1.

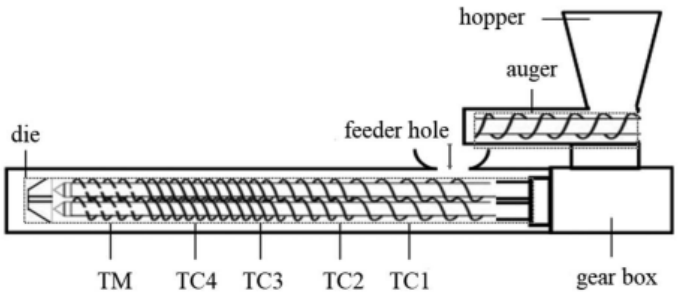

Figure 1. Schematic modular barrel and screw profile of twin screw extruder (Berto Industry BEX-DS-2256). TC = groove transfer direct pitch element (TC1 = 300 mm, TC2 = 220 mm, TC3 = 140 mm, TC4 = 120 mm), TM = groove mixing pitch element (80 mm), total long element = 800 mm.

Methods

Extraction of alginate

Pre-extraction of brown algae. Brown algae was dissolved in 0.03 M HCl with ratio of 1:20 (b/v) under strong stirring. The pH was adjusted to 2.9 by 1 M NaOH and gently stirred at speed of 500 rpm for 64 min. The brown algae was rinsed with distilled water to eliminate the excess of acid, and the remaining water was removed using a pressure machine (Hernandez-Carmona et al. 1999).

Alginate extraction using twin-screw extruder. Pretreated brown algae was gradually added to Na_2CO_3 solution (pH 8–12) with ratio of 1:3 (b/v), stirred, and transferred into the extruder's hopper. Alginate was extracted using twin-screw extruder at auger speed (feed rate) of 30 rpm, screw speed of 50–100 rpm, and temperature of 40–80°C, according to Vauchel et al. (2008b). Brown algae was moved between rotating screws and then released in the open die. The extrudate was then dissolved in Na_2CO_3 pH 8–10 with ratio of 1:10 (b/v) and stirred at 1000 rpm. The mixture was centrifuged at 5000 rpm for 10 min to obtain supernatant. The filtrate was added to ethanol 96% with ratio of 1:2 (v/v), kept for 1 h, and filtered. Alginate was washed twice with ethanol 70% and 96%, respectively, filtered, and pressed. Alginate extract was oven-dried at 45°C for 24 h, crushed, and sieved with 60 mesh.

RSM experimental design

A Box-Behnken Design (BBD) was used to evaluate effect of extrusion parameters, including temperature (x_1), screw speed (x_2), and pH (x_3) on the multiple-response alginate. The design had 15 combinations and was randomly ordered with 3 replications at the center point (run 13–15) (Montgomery 2005). The three evaluated variables were temperature (40–80°C), screw speed (50–100 rpm), and pH (8–12). Coding and actual variables of independent parameters are presented in Table 1. The selection of the levels are based on the result of our previous experiment in preliminary study. The experimental data of multiple-responses of alginate (RTD, yield, intrinsic viscosity, and molecular weight) from the Box-Behnken Design was analyzed using response surface regression (Table 2) and fitted the second order polynomial model:

$$y = + + \quad (1)$$

where Y is response of intrinsic viscosity, yield, and molecular weight; β_0 is intercept coefficient; β_i, β_{ii}, β_{ij} are regression coefficients for linear, quadratic, and interaction; x_i, x_j, are independent variables of temperature, screw speed, and pH. The accuracy of polynomial model was analyzed using Design-Expert (Ver. 7.0) software to obtain correlation coefficient (R) and determination coefficient (R^2) of observed variables (RTD, yield, intrinsic viscosity, and molecular weight). The significance of R and R^2 was statistically evaluated using F-test ($P < .05$). The validity of optimum condition between the validation experiment and predictions was evaluated by paired t-test ($P < .05$) using Minitab (Ver. 16.0) software.

Characterization of alginate

Determination of yield. Yield was determined as ratio of the weight of extracted alginate and initial weight of dry brown seaweed, then multiplied by 100% (Torres et al. 2007).

Table 1. Independent variables, coding and actual level of Box-Behnken design.

		Coding and actual level		
Independent variables	Symbol	+1	0	−1
Temperature (°C)	(X1)	80	60	40
Screw speed (rpm)	(X2)	100	75	50
pH	(X3)	12	10	8

Table 2. Variable experiment and multiple responses of alginate extraction by twin screw extruder in Box-Behnken design.

	Variable of extraction				Response		
No.	Temperature (°C)	Screw speed (rpm)	pH	RTD (min)	Yield (%)	Intrinsic viscosity (mL/g)	Molecular weight (kDa)
1	60	100	8	12.00	16.06	180.15	87.72
2	80	75	8	14.00	14.68	143.78	66.86
3	40	100	10	5.60	30.54	255.20	119.79
4	80	75	12	6.40	35.20	288.22	135.55
5	40	75	8	15.00	13.68	249.60	117.12
6	60	50	12	6.00	27.07	100.44	46.42
7	80	50	10	6.51	31.12	171.11	79.79
8	40	50	10	8.00	30.16	264.00	123.98
9	40	75	12	6.71	31.35	219.95	102.99
10	60	100	12	5.25	33.80	220.00	103.02
11	80	100	10	5.45	36.00	194.00	90.66
1	60	50	8	15.8	15.37	280.00	131.62
1	60	75	10	7.08	34.60	403.11	190.57
5	60	75	10	6.95	34.99	529.07	251.29
11	60	75	10	7.04	35.38	410.00	193.94
Pred	58.18	77.99	10.11	6.72 ± 0.240[a]	35.46 ± 1.28[b]	447.58 ± 64.32[c]	212.01 ± 30.89[d]
Valid	58.18	77.99	10.11	7.07 ± 0.029[a]	34.01 ± 0.12[b]	460.13 ± 14.75[c]	217.94 ± 7.14[d]

Determination of intrinsic viscosity. Alginate viscosity was measured at 25°C using viscometer glass (Canon, USA) with capillary diameter of 0.56 mm. To prepare alginate solution, alginate (30 mg) was dissolved in 10 mL distilled water and stirred for 3 h at 25°C (Chee et al. 2011). A series of different concentrations (0.05–0.3 g/dL) of alginate were then made. The flow time of alginate solution (t) was relatively measured to distilled water (t_0) and temperature was set 25°C. Intrinsic viscosity was determined by extrapolating η_{sp}/c concentration to zero.

$$\text{Relative viscosity, } \eta = \frac{t}{t_0} \tag{2}$$

$$\text{Specific viscosity, } \eta_{sp} = \eta - 1 \tag{3}$$

$$\text{Reduction viscosity, } = \frac{\eta_{sp}}{c} = \frac{\eta_{-1}}{c} \tag{4}$$

$$\text{Intrinsic viscosity, } [\eta] = \lim_{c \to 0} \frac{\eta_{sp}}{c} \tag{5}$$

Determination of molecular weight. Determination of alginate molecular weight was based on correlation of average intrinsic viscosity and molecular weight using formulation of Mark-Houwink (Eq. 6), where k = 0.023 dL/g and a = 0.984, which was proposed by Clementi et al. (1998) and quoted by Fertah et al. (2014), Chee et al. (2011), and Torres et al. (2007) to empirically relate intrinsic viscosity [η] to the average molecular weight (M_w).

$$[\eta] = kM_w^a \tag{6}$$

The [η] is intrinsic viscosity (dL/g), and M_w is molecular weight (kDa).

Residence time distribution. Residence time distribution of algae was the time at which the material was injected until the material was released in the opening die (Baron et al. 2010).

Structural analysis. Preparation of alginate samples for ^{1}H-NMR analysis with partial hydrolysis was performed according to Davis et al. (2004). Twenty milligrams of alginate was dissolved in 60 mL aquabidest and stirred for 3 h, and the pH was adjusted to 5.6 with 0.1 M HCl. The mixture

was hydrolyzed in a water bath at 95°C for 1 h, pH was set to 3.8 with 0.1 M HCl. Then, the mixture was heated again in a water bath for 45 min. pH was neutralized with 0.001–0.1 M NaOH and freeze dried. Analysis of ^{1}H-NMR spectroscopy was performed according to Grasdalen et al. (1979). Sodium alginate (5 mg) was diluted in 0.6 mL D_2O, heated at 80°C, and then injected to NMR (JEOL 400 spectrometer).

The method to calculate M/G ratio and determination of block structure alginate was according to Grasdalen (1979). The quantitative analysis of individual guluronic acid (F_G) and doublet GG fraction used the signal and relative area anomeric protons from the peaks I–III using the following equation:

$$F_G = A_I/(A_{II}+A_{III}) \tag{7}$$

$$F_{GG} = A_{III}/(A_{II}+A_{III}) \tag{8}$$

Fraction M was determined from:

$$F_M = 1-F_G \tag{9}$$

The M/G ratio was calculated using the equation:

$$M/G = (1-F_G)/F_G) \tag{10}$$

Doublet fraction (F_{GG}, F_{MM}) was calculated by the following equation:

$$F_{GG}+F_{GM}=F_G \tag{11}$$

$$F_{MM}+F_{MG}=F_M \tag{12}$$

Rheology of the sodium alginate solution and calcium alginate gel. The rheological behavior of sodium alginate solution and gel was determined using a Haake Mars III rotational rheometer (Karlsruhe, Germany). Plate and plate type geometry with 35 mm diameter plate was used for gels, and a cone and plate geometry (cone diameter 35 mm, angel 4°) was used for alginate solution. Dynamic viscosity measurement for sodium alginate solution in concentrations range of 2–3% (w/v) was carried out at a temperature of 25°C. Dynamic viscosity measurement of alginate solution of 2.5% (w/v) at different temperatures (range 25–35°C) used cone and plate geometry (35 mm diameter plate, angel 4°), in the range of shear rate of 0–1000 s^{-1}. Power law model was used to calculate flow behavior index (n) and consistency coefficient (K) using the following equation:

$$\eta_a = K\,\gamma^{n-1} \tag{13}$$

Where η_a is the apparent viscosity (Pa.s), K is the consistency coefficient (Pa.s^n), γ is the shear rate (s^{-1}), and n is the flow behavior index (dimensionless).

Rheological measurement of alginate gel was analyzed by oscillatory dynamic test. Plate and plate type geometry with 35 mm diameter plate was used. The angular frequency (ω) was set at range of 0–100 rad/s. Dynamic moduli storage modulus (G'), loss modulus (G''), loss tangent (tan δ = G''/G'), and complex viscosity [η*] were determined as a function of angular frequency (ω). Alginate gel was prepared from an aqueous solution of sodium alginate (20 g/L). The solution (30 mL) was put in a dialysis tube using membrane cellulose with molecular weight cut off 2000 (MWCO) against 1 M $CaCl_2$ solution and stirred for 48 h to assure equilibrium (Gomez et al. 2009; Rahelivao et al. 2013). All rheological measurements were performed in triplicate, and the average values with overlapping trace were reported.

Specific mechanical energy. Specific mechanical energy was calculated using power motor P (W), current I (57.64 A), feed rate Qs (kg/h), power factor of electrical motor cos Q (0.95), screw speed Ss

(50–100 rpm), voltage V (380 volt), and maximum screw speed Ssmax (180 rpm), based on the following formula:

$$\mathrm{SME} = \frac{\mathrm{P}}{\mathrm{Qs}}, \text{and } \mathrm{P} = \frac{\mathrm{VxIx}\cos Q \text{ x Ss}}{\mathrm{Ss}max} \tag{14}$$

Results and discussion

Residence time distribution

The treatment of temperature, pH, and screw speed gives a quadratic effect on brown algae residence time in the screw channel (Figure 2a). The algae residence time distribution was accelerated with increasing temperature, pH, and screw speed. This condition can be explained by several phenomena. First, the increase of screw speed causes compressive force, and friction of the algae with the screw walls is stronger and causes the algae cell wall to break apart. Alginate then quickly interacts with the solvent to form viscoelastic fluid, making the RTD faster. Kartika et al. (2010) stated that the increasing screw speed causes the SME effect to be stronger as a result of the cell wall becoming porous. Second, rising barrel temperature causes brown algae to inflate the cell wall and become porous due to thermal expansion. This condition causes more interaction of alginate with solvents. The algae becomes slippery so that the movement in the screw becomes faster, and there is low die pressure. Third, the increased pH level causes the solubility of alginate in the sodium carbonate solution to become larger. The surface of the algae becomes slippery, so that it is easy to move in the screw channel, and the

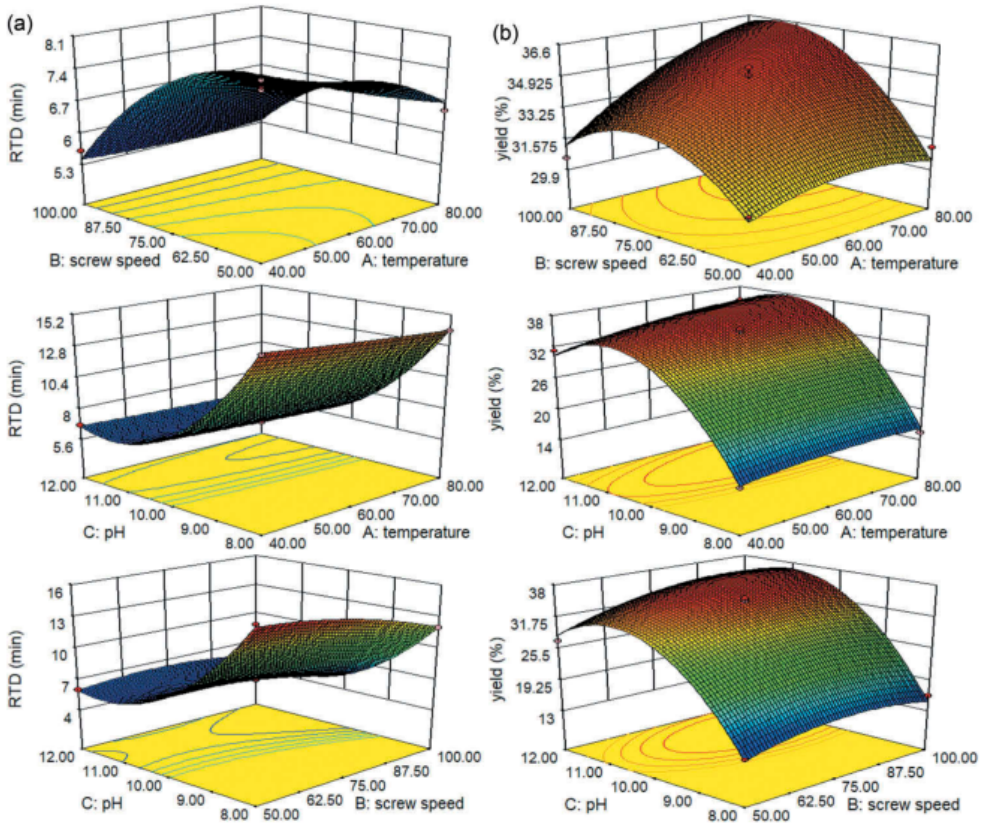

Figure 2. Response surface plot of the effect of extrusion parameters on residence time distribution (RTD) (a) and yield of alginate (b).

extractability of alginate becomes higher. Baron et al. (2010) reported increased screw speeds caused deeper RTD algae in the screw channel. Sugiono et al. (2018) suggested that increased pH caused greater alginate solubility and that the solubility rate of alginates was logarithmically proportional to the time and alkali concentration of the extraction process. Wang et al. (2016) reported that increased screw speed and temperature barrel increased the friction level of the material to form viscoelastic fluid as a result of lower die pressure and torque motor, and the material moved faster in the screw extruder.

Yield

The yield of alginate was 13–36% (Table 2). Temperature showed linear correlation to the yield, while pH and screw speed showed quadratic correlation to the yield (Figure 2b). The yield of alginate increased with the increasing temperature, screw speed, and pH. This may have resulted from formation of porous structure of brown seaweed cell walls, thus increasing the extractability of alginate and its solubility in sodium carbonate solution. Fertah et al. (2014) found that alginate yield increased as higher temperature was used and then decreased because of alginate chain degradation. In higher temperature, increased alginate yield was associated with more porous structure formation in the cell walls of brown algae (Hernandez-Carmona et al. 1999; Torres et al. 2007). Baron et al. (2010) reported that various screw speeds of twin-screw extruder in extracting brown seaweed alginate resulted in stronger effects on specific mechanical energy of the materials. Kartika et al. (2005) showed that positive correlation between SME and screw speed was found due to disintegration of brown seaweed cell walls, leading to enhancement of extractability and solubility of alginate (Figure 3).

Intrinsic viscosity

Extraction conditions demonstrated quadratic effect on intrinsic viscosity of alginate. Intrinsic viscosity increased at temperature 60°C, screw speed 75 rpm, and pH 10 and decreased at temperature 80°C, screw speed 100 rpm, pH 12 (Figure 4a). This reflects the increasing extractability of high molecular weight alginate at temperature 60°C, screw speed 75 rpm, and pH 10. Meanwhile, alginate viscosity decreased at temperature 80°C, screw speed 100 rpm, and pH 12 due to degradation of polymer chains. This is in accordance with a previous study by Torres et al. (2007), which found that specific viscosity of brown seaweed alginate increased at temperature 60°C but then decreased at temperature 80°C. Sellimi et al.

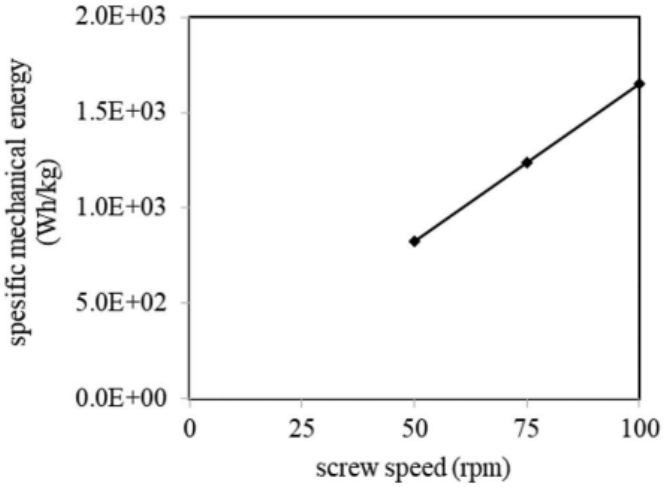

Figure 3. Effect of screw speed on the specific mechanical energy.

(2015) found that alginate viscosity was highly susceptible to heating, and the viscosity decreased with the increasing temperature. Lowered alginate viscosity at high temperature was associated with disruption of alginate polymer chains (Fertah et al. 2014). Furthermore, alginate viscosity was stable at pH 10, and alginate was degraded at pH 12 due to reaction of β-elimination; while at low pH, alginate was hydrolyzed (Haug et al. 1963; Smidsrod et al. 1969; Vauchel et al. 2008b). Extrusion temperature at screw speed 75 rpm raised extractability of high molecular weight alginate, while alginate polymer chain was degraded at screw speed 100 rpm. Kartika et al. (2010) showed that increasing screw speed caused stronger SME, leading to disruption of alginate polymer chain.

Molecular weight

Extraction conditions showed quadratic effect on molecular weight, which increased at temperature 60°C, screw speed 75 rpm, and pH 10 and decreased at temperature 80°C, screw speed 100 rpm, and pH 12 (Figure 4b). Increased extraction temperature caused pores in the structure of cell walls, thus increasing extractability of alginate (Lorbeer et al. 2015). Lower molecular weight resulted from degradation of alginate polymer chains due to enhanced temperature and pH of extraction (Hernandez-Carmona et al. 1999; Silva et al. 2015; Torres et al. 2007). A decline in alginate molecular weight at around neutral pH was affected by hydrolysis catalyzed by protons (Haug et al. 1963; Smidsrod et al. 1969). Lorbeer et al. (2015) reported conversion of non-comprehensive Ca/H in pre-extraction process, in which high molecular weight alginate possessed greater amount of calcium and magnesium ions, thus making it more resistant to extract than low molecular weight alginate.

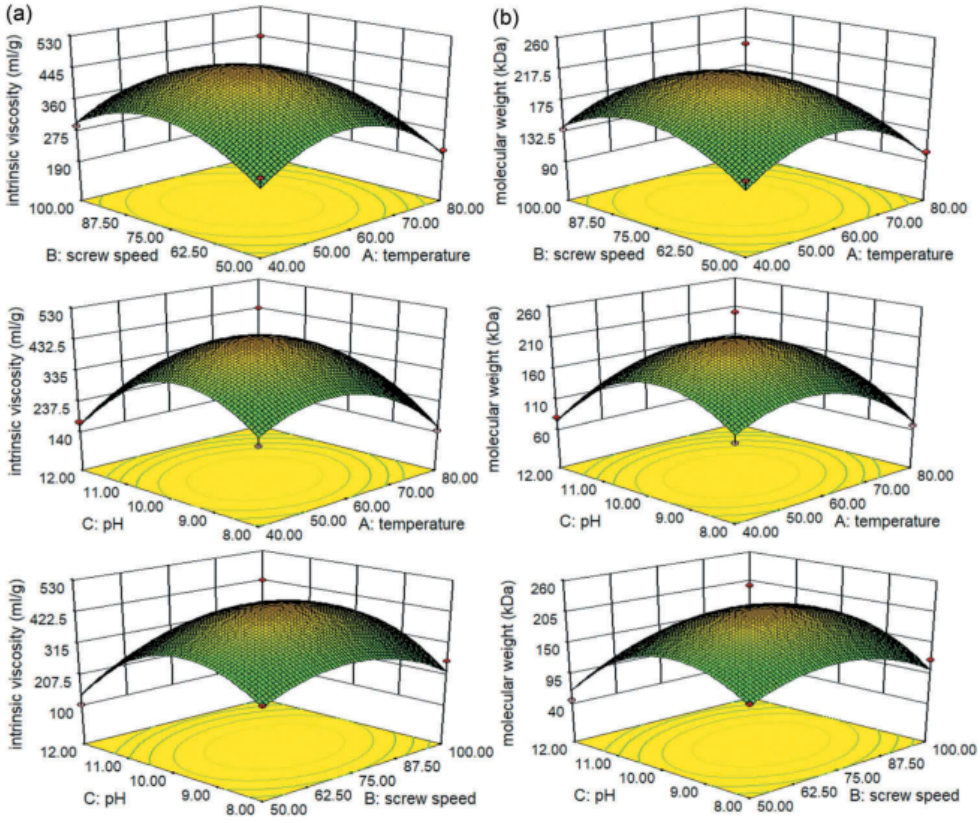

Figure 4. Response surface plot of the effect of extrusion parameters on the intrinsic viscosity (a) and molecular weight of alginate (b).

Increasing SME was associated with an increase in shear rate due to high screw speed, resulting in degradation of alginate chain polymer (Baron et al. 2010; Huang and Ma 2016).

Fitting model

The experiment was designed to understand the effects of temperature, screw speed, and pH on alginate quality and determine the optimum condition using response surface methodology based on RTD, yield, intrinsic viscosity, and molecular weight. Three evaluated variables of temperature (40, 60, and 80°C), screw speed (50, 75, and 100 rpm), and pH (8, 10, and 12) in Box-Behnken Design were performed at 3-replications in the center point (Table 2). The center point was selected based on the results of our previous experiment in preliminary study. Response surface regression analysis was based on the experimental data. Prediction of second order polynomial of the responses is presented in Table 3.

The established quadratic models of RTD, yield, intrinsic viscosity, and molecular weight were evaluated using the following indicators: model significance, lack of fit, and correlation coefficient, as presented in Table 4. The model fit possessed significance at $P < .05$, $R^2 \geq 0.8$, and lack of fit >0.1 (Montgomery 2005). Based on these parameters, polynomial models of the multiresponses in second equation order model were entirely fitted and examined for the goodness of fit. The results showed that the P value of all response variables was $<P = .05$, with no significant lack of fit, and R^2 values were >80%. Therefore, the models that were designed using BBD were highly adequate.

Optimization and verification

The optimum conditions for alginate extraction were found at temperature 58.18°C, screw speed 77.99 rpm, and pH 10.11. At these conditions, maximum results were predicted at RTD 6.72 min, yield 35.46%, intrinsic viscosity 447.58 mL/g, and molecular weight 212.01 kDa, resulting in desirability value 0.852 (Figure 5). Desirability value of almost 1.00 means that prediction of optimal condition by Design Expert reflects high validity (Ale et al. 2012; Lorbeer et al. 2015; Qiao et al. 2009; Sugiono et al. 2014).

The prediction of optimum conditions (temperature 58.18°C, screw speed 77.99 rpm, and pH 10.11) was verified using 3-replicate experiment, which resulted in RTD 7.07 ± 0.029 min, yield 34.01 ± 0.12%, intrinsic viscosity 460.13 ± 14.75 mL/gm and molecular weight 217.94 ± 7.14 kDa. The verification demonstrated a 95% prediction interval (PI) low and a 95% PI high, indicating that

Table 3. Model quadratic and significant code of alginate extraction by twin screw extruder.

Y	a	b_1	b_2	b_3	b_{12}	b_{13}	b_{23}	b_1^2	b_2^2	b_3^2
RTD (min)	7.02	−0.37*	−1.00*	−4.06*	0.34*	0.17	0.76*	0.066	−0.70*	3.44*
Yield (%)	34.99	1.37*	1.80*	8.45*	1.13	0.79	1.09	−0.77	−2.27*	−10.07*
Intrinsic viscosity (mL/g)	447.39	−31.38	11.63	−30.11	−8.54	28.72	32.09	−112.8*	−89.05*	−148.43*
Molecular weight (kDa)	211.93	−14.86	5.50	−14.23	−4.07	13.8	15.2	−53.95*	−42.77*	−70.86*

Type equation $Y = a + b_1x_1 + b_2x_2 + b_3x_3 + b_{12}x_1.x_2 + b_{13}x_1.x_3 + b_{23}x_2.x_3 + b_{11}x_1^2 + b_{22}x_2^2 + b_{33}x_3^2$
a = intercept; b_i = linear (b_1 = temperature, b_2 = screw speed, b_3 = pH); b_{ij} = interaction (b_{12} = temperature and screw speed, b_{13} = temperature and pH, b_{23} = screw speed and pH); b_{ii} = quadratic (b_1^2 = temperature^2, b_2^2 = screw speed^2, b_3^2 = pH^2).
Significant code * $P < .05$

Table 4. The quality of fit of the second-order functions.

Function	Model significance (Ps)	Lack of fit (P_L)	Correlation coefficient (R^2)
RTD	< 0.0001	0.0584	0.9984
Yield	< 0.0001	0.7917	0.9939
Intrinsic viscosity	< 0.0001	0.7876	0.9123
Molecular weight	< 0.0001	0.3367	0.9114

the verification results for attaining maximum yield, intrinsic viscosity, and molecular weight were consistent and valid. The result of paired t test showed that the differences in multiple responses of alginate between verification and prediction of the optimal point were not significant ($P > .05$). It was an evident that the predicted values were in great agreement with the actual extraction conditions. Verification experiments supported the optimal point of predictions and validity.

Chemical structure

The ^{1}H-NMR spectra of *Sargassum cristaefolium* sodium alginate (SCSA) showed three different structures (Figure 6). The signal chemical shift at 4.92 ppm corresponds to H-1 of guluronic units (peak 1), mannuronic units (M-1), and alternative blocks GM (GM-5) overlapping at chemical shift 4.83 ppm and signal 4.34 ppm H-5 guluronic blocks GG-5. The result of this research was in accordance with previous results reported by Heyraud et al. (1996) and Gomez et al. (2009). From the integration of these signals and spectra calculation of ^{1}H NMR, it was found that M/G ratio was 0.28, and GG blocks were 0.78, which was higher than MM blocks of 0.22.

This M/G ratio of alginate from *S. cristaefolium* was relatively similar to *S. filipendulla* (0.19), *S. muticum* (0.31), *S. polycystum* (0.21), and *S. turbinaroides* (Davis et al. 2003; Fenoradosoa et al. 2010), but the M/G ratio was very low when compared to *S. vulgare* (1.27/1.56) and *S. latifoilum* (0.82) reported by Torres et al. (2007) and Larsen et al. (2003). Variation of the M/G ratio of alginate from brown algae was affected by species, seasons, growing location, type and age of tissue, and extraction methods (Andriamanantoanina and Rinaudo 2010; Bertagnolli et al. 2014; Davis et al. 2004; Fenoradosoa et al. 2010; Larsen et al. 2003; Leal et al. 2008). The comparison of the M/G ratio and doublet fraction data for *S. cristaefolium* with other *Sargassum* species is shown in Table 5.

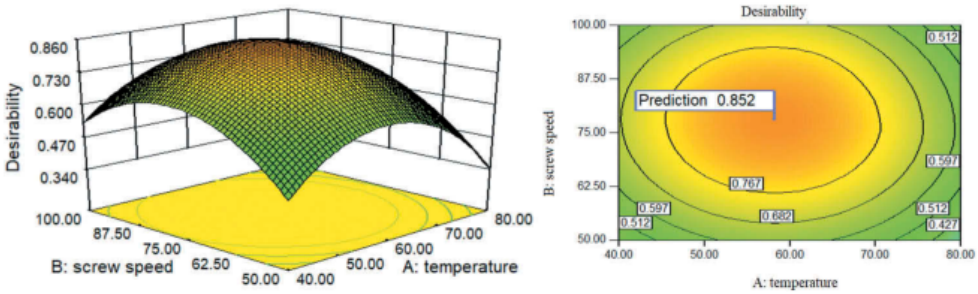

Figure 5. Desirability in optimization of screw speed and temperature.

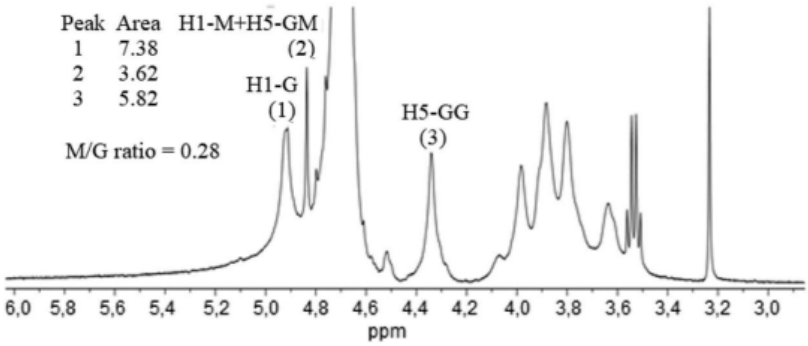

Figure 6. ^{1}H-NMR spectra of the sodium alginate from *Sargassum cristaefolium* in deuterium water and its characteristic signals.

Rheological properties

Effect of concentration on the flow behavior

Rheological properties of *Sargassum cristaefolium* sodium alginate in aqueous solution at different ranges of concentration (2–3%) and ranges of shear rate (0–1000 s^{-1}) are shown in Figure 7a. Flow behavior of SCSA in aqueous solution was shown as curve shear rate versus apparent viscosity and analyzed using power law model. SCSA in aqueous solution exhibited the property of shear thinning pseudo plastic with flow behavior index n less than 1 ($n < 1$) (Table 6). Rao et al. (2003) reported that based on the value of flow behavior index (n), a liquid is swelling plastic fluid if $n > 1$. Liquid is Newtonian if flow behavior index $n = 1$ and pseudo plastic if $n < 1$.

The influence of concentration on the parameters in power law model showed that the flow behavior index (n) and consistency coefficient (K) decreased with increasing concentration in the range of 2–3%. Pseudo plasticity of fluid seemed to increase with increasing alginate concentration. The result of this study was in accordance with reports by Ma et al. (2014) and Sugiono et al. (2019). Rao et al. (2003) found that pseudo plastic fluid behavior represented a characteristic of polysaccharide solution with high polymer structure and molecular weight.

Regarding the effect of concentration on the flow curve of SCSA in aqueous solution, it was found that the apparent viscosity increased with increasing concentration in the range 2–3%. Apparent viscosity was affected by shear rate, meanwhile apparent viscosity decreased with the increasing angular frequency (Figure 7a). Disruption and formation of molecular bindings in angular frequency caused structural changes that affected rheological properties (Tunick 2011). In high frequency, there is no sufficient time to damage inter and intra-molecular fractions for reassociation. This phenomenon caused permanent molecular integration or long chain polymer separation, leading to reduction of viscosity (Ma et al. 2014). Properties of this fluid were in accordance with previous results of Sellimi et al. (2015) and Gomez et al. (2009) but were in contrast to that reported by Torres et al. (2007).

Mancini et al. (1996) stated that apparent viscosity was affected by shear rate, temperature, and concentration. Apparent viscosity increased with the increasing alginate concentration (Fenoradosoa et al. 2010; Torres et al. 2007). At high concentration, overlapping molecules occurred that induced formation of intermolecular junctions. This limited the regulation and stretching of alginate molecules, thereby enhancing apparent viscosity of solution (Bae et al. 2008; Sellimi et al. 2015).

Effect of temperature on the flow behavior

The effect of temperature on the flow behavior of SCSA in aqueous solution was also studied. Flow behavior of SCSA in aqueous solution at different temperatures was plotted as a curve of shear rate versus

Table 5. Compositional data of the sodium alginate from species of *Sargassum*.

Sargassum species	Origin	F_G	F_M	M/G ratio	F_{GG}	F_{MM}	F_{MG}	F_{GM}	Reference
S. filipendulla	Brazil	0.6	0.4	0.67	0.42	0.22	0.18	0.18	Bertagnolli et al. (2014)
S. vulgare	Brazil	0.44	0.56	1.27	0.43	0.55	0.01	0.01	Torres et al. (2007)
		0.39	0.61	1.56	0.36	0.58	0.03	0.03	Torres et al. (2007)
S. fluitans	Cuba	0.66	0.34	0.52	0.57	0.25	0.09	0.09	Davis et al. (2004)
S. oligocictum	Australia	0.62	0.38	0.62	0.55	0.31	0.07	0.07	Davis et al. (2004)
S. filipendulla	_[a]	0.84	0.16	0.19	0.76	0.07	0.08	0.08	Davis et al. (2003)
S. muticum	England	0.76	0.24	0.31	0.59	0.07	0.17	0.17	Davis et al. (2003)
S. polycystum	_[a]	0.82	0.18	0.21	0.77	0.12	0.05	0.05	Davis et al. (2003)
S. thumbergii	Korea	0.66	0.34	0.53	0.48	0.17	0.17	0.17	Davis et al. (2003)
S. dentifolium	Egypt	0.66	0.34	0.52	0.55	0.23	0.11	0.11	Larsen et al. (2003)
S. asperifolium	Egypt	0.59	0.41	0.69	0.48	0.30	0.11	0.11	Larsen et al. (2003)
S. latifolium	Egypt	0.55	0.45	0.82	0.51	0.41	0.04	0.04	Larsen et al. (2003)
S. cristaefolium	Indonesia	0.78	0.22	0.28	0.62	0.05	0.16	0.16	This study

[a]Unknown origin.

F_G = Guluronate fraction; F_M = Manuronate fraction; F_{MM} = Homopolymer manuronate fraction; F_{GG} = Homopolymer guluronate fraction; M/G ratio = Ratio of manuronate to guluronate; F_{MG} = Heteropolymer manuronate guluronate fraction; F_{GM} = Heteropolymer guluronate manuronate fraction.

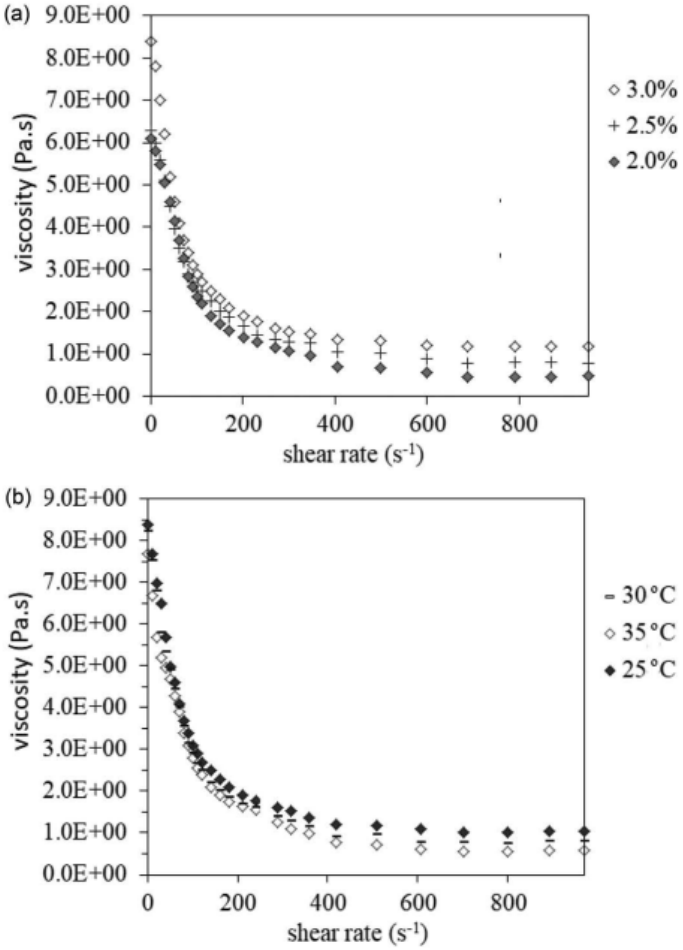

Figure 7. Dynamic viscosity *Sargassum cristaefolium* sodium alginate (SCSA) on concentration 2–3% in aqueous solution at temperature 25°C (a), and the dynamic viscosity of 2.5% SCSA in aqueous solution during heating 25–35°C with increasing the shear rate 0–1000 s^{-1} (b).

Table 6. Power law parameters of SCSA in aqueous solution at concentration 2–3% and temperature 25–35°C.

Concentration(%)	Temperature (°C)	K (Pa.s^n)	n(-)	R^2
2.0	25	12.1363 ± 0.4190	0.5950 ± 0.0200	0.9706 ± 0.0098
2.5	25	11.8206 ± 0.7670	0.6126 ± 0.0117	0.9662 ± 0.0106
3.0	25	11.1726 ± 0.8501	0.7030 ± 0.00160	0.9706 ± 0.0098
3.0	25	11.5776 ± 0.1622	0.6250 ± 0.14080	0.9688 ± 0.0053
3.0	30	12.3480 ± 0.0970	0.6210 ± 0.0040	0.9703 ± 0.0016
3.0	35	12.1005 ± 0.1956	0.5710 ± 0.0226	0.9633 ± 0.0034

apparent viscosity (Figure 7b). Figure 7b shows that the flow behavior of SCSA in aqueous solution exhibits a shear thinning pseudo plastic because flow behavior index (n) was less than 1. Pseudo plastic behavior of SCSA in aqueous solution in this result was in accordance with previous results reported by Xiao et al. (2012) and Ma et al. (2014). The influence of temperature on the parameters in power law model showed that the flow behavior index increased with increasing of temperature, whereas the consistency coefficient decreased. An increase in temperature caused a decrease of pseudo plasticity, indicated by a decrease in flow behavior index, but power law model was stable (Table 6). The flow

behavior of the pseudo plastic could be maintained at higher test temperature. This indicated that SCSA in aqueous solution was less sensitive to the temperature in the range test.

Regarding the effect of temperature on the flow curve of SCSA in aqueous solution, it was found that the apparent viscosity decreased with increasing temperature in the range 25–35°C (Figure 7b). This finding was in accordance with results from Sellimi et al. (2015). Higher temperature caused the distance between molecules of alginate to increase, which was a result of thermal expansion. Therefore, the apparent viscosity of alginate decreased (Ma et al. 2014; Rao et al. 2003; Sugiono et al. 2019).

Dynamic viscoelastic properties

Gelation ability of SCSA showed that the storage modulus and loss modulus increased with increasing frequency sweep test at range 0.1–100 rad/s (Figure 8a). The storage modulus was always higher than loss modulus in all the frequency ranges of sweep test. This condition had correlation with the higher component of GG block and high molecular weight of SCSA. The result of this research was in accordance with Gomez et al. (2009) and Rahelivao et al. (2013). In comparison, *G'* and *G"* of SCSA was higher than commercial sodium alginate (CSA) in all ranges of frequency in sweep test (Figure 8a). This condition was in accordance with the molecular weight and composition of GG block in SCSA that was higher than CSA. Viscoelastic moduli of CSA increased continuously at increasing frequency sweep. This is an indication that the viscoelastic behavior of CSA was more liquid-like because the majority of energy was dissipated or lost by viscous flow (Ma et al. 2014). *G"* of CSA was higher than *G'* by about 10 rad/s; however, *G'* was higher than *G"* at high frequency (Figure 8b). This was an implication of no

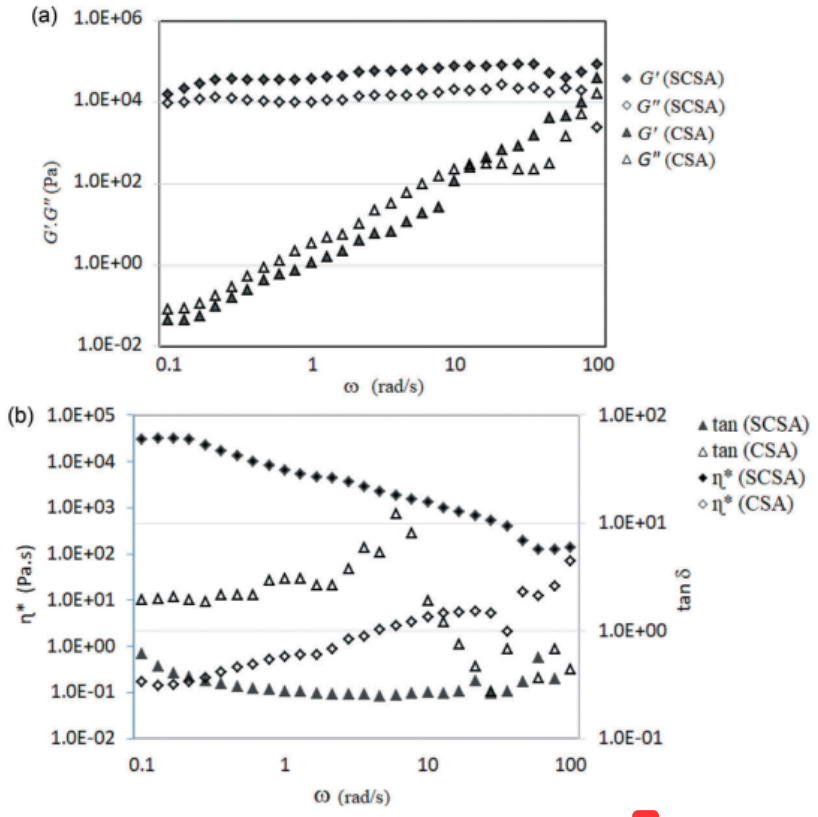

Figure 8. Dynamic rheological behavior of alginate gel in 1 M $CaCl_2$ at temperature 25°C. Storage modulus (*G'* = filled symbols) and loss modulus (*G"* = open symbols) (a), and complex viscosity (η*) and loss tangent (tan δ) (b). SCSA = *Sargassum cristaefolium* sodium alginate, CSA = commercial sodium alginate.

structural network and links between macro-molecules of weak structure soft gel or dispersion that affected low molecular weight of CSA, as well as limited cross-linked polymer between GG block in the presence of Ca^{2+} ion (Tunick 2011).

Complex viscosity of SCSA decreased with the increase of frequency of sweep test, and loss tangent (tanδ) was low (δ < 1) (Figure 8b). This condition was correlated with strong interaction GG block with Ca^{2+} ion in the polymer of SCSA (Funami et al. 2009; Grant et al. 1973). Value of loss tangent was low (δ < 1) and $G'>G''$, which means that the polymer was more viscous-like, cross linked polymer, gel stiff strong, and stable because the building 3D-network structure. SCSA gel was more elastic than liquid. CSA gel at low frequency $G'<G''$ was more liquid; while at high frequency, $G'>G''$ was more elastic, cross over at frequency 12.91 rad/s, and loss tangent δ > 1 (Figure 8b) (Vicini et al. 2015). This condition showed that the gel alginate of CSA was more semiliquid (Funami et al. 2009).

Conclusion

Extraction conditions (temperature, screw speed, and pH) significantly affected multiresponses of alginate. The optimum conditions were found at temperature 58.18°C, screw speed 77.99 rpm, and pH 10.11, resulting in RTD 7.07 ± 0.029 min, yield 34.01 ± 0.12%, intrinsic viscosity 460.13 ± 14.75 mL/g, and molecular weight 217.94 ± 7.14 kDa. The M/G ratio of *Sargassum cristaefolium* sodium alginate was 0.28 and GG blocks of 0.78 were higher than MM blocks of 0.22. *Sargassum cristaefolium* sodium alginate in aqueous solution showed a shear-thinning pseudoplastic liquid, and its gel in 1 M $CaCl_2$ was more elastic than liquid.

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Structural and Rheological Characteristics of Alginate from Sargassum cristaefolium Extracted by Twin Screw Extruder

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