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Effects of Extraction Process Conditions on Semi Refined Carrageenan Produced by using Spray Dryer

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Abstract: The objective for this work was to study the characteristics of Semi Refined Carageenan (SRC) which extracted from *Kappaphycus alvarezii*. The production of SRC was conducted by alkali treatment of seaweed and to obtain the powder used spray drying technique. This analysis based on two independent parameter which are concentration of potassium hydroxide (KOH) and extraction time. Extraction temperature during alkali treatment process was fixed as constant which was 70-80°C. The optimization of extraction process parameters and the experimental design were done based on Central Composite Design (CCD) of Response Surface Methodology (RSM). Gel viscosity, powder weight of SRC, particle size and gel strength were studied as responses. The bounds of all these factors keyed in the RSM to get the predicted number of experiments. Optimum process extractions for spray drying technique result showed at 6.70% of KOH concentration and 74.70 minute extraction time. At optimum conditions the viscosity of the SRC gel was found as 111.80 cP and the particle size of SRC powder was found as 86.88 μm. Gel strength was optimize at 85.60 g cm^{-2} and SRC powder production as 5.01 g.

Key word: *Kappaphycus alvarezii*, extraction, response surface methodology, carrageenan

INTRODUCTION

Carrageenan or known as carrageen gum is a family of polysaccharides which extracted from red seaweeds of the *Rhodophycaea* family (Gunning *et al*., 1998). It has a kaleidoscope of species of seaweeds, such as *Eucheuma*, *Iridaea*, *Chondrus* and *Gigartina* (Van de Velde and De Ruiter, 2002). There are several types of carrageenans, such as iota (i), kappa (k), lambda (λ), Mu (μ), Nu (v) and Theta (θ) (Thanh *et al*., 2002). Carrageenan compounds differ from agar in that they have sulfate groups ($-OSO_3^-$) in place of some hydroxyl groups. The fundamental structure consists of alternating (1-3)-linked β-D-galactopyranose and (1-4)-linked α-D-galactopyranose where galactose residues are partially sulphated at the positions of 2 and/or 6 and/or 3,6-anhydrided, depending or carrageenan type and resulting on a wide range of gelling properties (Thanh *et al*., 2002; Krishnaiah *et al*., 2008a, b).

Different types of carrageenans will have different conformation and composition. However, the fundamental structure of carrageenans consists of alternating 3-linked β-D-galactopyranose (G-units) and 4-linked α-D-galactopyranose (D-units) (Campo *et al*., 2009). The main differences which influence the properties of types of carrageenan are number and position of ester sulfate groups. According to Van de Velde and De Ruiter, 2002, carrageenan is a high molecular mass material with a high degree of polydispersity. Generally, different species of seaweed can produce variety types of carrageenan. It depends on the class of seaweed. Their structures are varying according to species, seasons, geographic location and age of population (Graham and Wilcox, 2000, Prasad *et al*., 2010).

Extraction process act to removes colouring matter and some proteins and makes the gum more easily extractable. 6-sulfate may also eliminate in some extraction process. Basket of seaweed immersed and cooked in alkali solution and then soaked with fresh water to naturalized most of the residual alkali (Bono *et al*., 2012). *Kappaphycus alvarezii* (*Euchema cottoni*) is used in this process because it contains mainly kappa (k) carrageenan and this is the carrageenan that forms a gel with potassium salts. Iota-containing seaweeds can also be

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processed, although the markets for iota (i) carrageenan are significantly less than those for kappa. Lambda (λ) carrageenans do not form gels with potassium and would therefore dissolve and be lost during the alkali treatment (McHugh, 2003). Spray drying is a method of producing a dry powder from a liquid or slurry by rapidly drying with a hot gas. This is the preferred method of drying of many thermally-sensitive materials such as foods and pharmaceuticals (Mujumdar, 2006). Spray drying is a process involving the conversion of feed from liquid condition into fine droplets from the exposing them to a hot drying media (Cheng, 1969). The feed will be transform into dried particulate from can be a solution, suspension, dispersion, emulsion or slip. The dried product can be in forms of powders, granules or agglomerates depending on process variable such as particle size, flow rate, viscosity and solubility or the dryer design (type of atomizer) (Tonon *et al*., 2008). Hence, it can be used to turn the carrageenan slurry into powder that has longer shelf life and is readily available. The advantages of the dried extract over conventional liquid forms are lower storage costs, higher concentration and stability of active substances (Oliveira *et al*., 2006). The spray drying has also been adopted for manufacture of powders due to its ability to generate a product with precise quality specifications in continuous operations (Bono *et al*., 2011; Souza and Oliveira, 2006).

The objective of this research is to produce SRC powder using spray drying technique and then to evaluate the effects of variables to predict targeted responses by using Response surface Methodology (RSM) software. Therefore, the effect of process parameter on the properties of SRC powder which is viscosity, weight of SRC, particle size and gel strength will be determined.

MATERIALS AND METHODS

Basically, the laboratory work was divided into three parts; the first part of the experiment was preparation of SRC with extraction and drying process. Second part was characterization of SRC and followed by optimization of the process parameters in extraction process condition using RSM.

Preparation of SRC from *Kappaphycus alvarezii*

Preparation of raw seaweed: Raw seaweed was prepared sun drying by fresh seaweed collected from farm. Sundried seaweed was washed thoroughly with distilled water for two reasons; to remove contaminants and to standardize the moisture content of the seaweed. Wet seaweed was then dried in oven at 60°C for 15-16 h to remove the excess moisture.

Extraction of seaweed: The dried seaweed was extracted in batch cooker in Potassium Hydroxide (KOH) solution. The extraction temperature was controlled to be constant, whereas the KOH concentration and extraction time were fixed to the desire value. The extracted seaweed was then cooled down to room temperature and washed with distilled water to remove the excess KOH. These processes seaweed was classified as SRC.

Powder production: After that, the sample melted with distilled water at 90°C before spray drying process. The ratio of sample to distilled water is 1:15. Feed flow rate is set up at 8 mL h^{-1} and hot air flow rate in drying chamber was 40 m^3 h^{-1}. The hot air inlet temperature is 140°C to spray dryer. Spray dryer process was performed in the laboratory scale spray dryer Lab Plant SD-05.

Characterization of SRC powder

Gel viscosity measurement: The powder of carrageenan should be mix with distilled water with 1.5% of powder carrageenan at 75°C by using impeller. The viscosity of the samples will be tested by using viscometer. The spindle is no. 6 and speed of rotation for the viscometer is 30 rpm after placed the sample in water bath for 2 h. The viscosity of carrageenan solutions should be determined under conditions where there are no tendencies for the solution to start gelling. Therefore, the viscosity measurements of carrageenan solutions should be determined at sufficiently high temperatures (75° C) to avoid the effect of gelation.

SRC powder weight measurement: After spray drying process carried out, SRC powder was collected in a plastic bag and sealed. Then, it was weighted by using electronic weighing scale and the result was recorded.

Particle size analysis: Particle size will be tested by placed the powder on the Microscope with Image Analyzer (MIA) at 4X enlargement. During MIA analysis, electrons will passes completely through the sample and image will be appear in the computer. Then taking the picture of the image in microscope and the image will transmit to the computer by image acquisition system. The system will process and analyze the image in special particle size analysis software until output the results appear through monitor.

Gel strength measurement: Sample was prepared by dissolving 1.5 g of SRC powder to 100 mL of distilled water with continuous magnetic stirring at 90°C around 20-30 min. The sample was allowed to stabilize in a water bath to eliminate bubbles forms at 80-90°C for 15 min. The viscous solution was poured into three 50 mL beakers and left for 20-30 min before sealing them. Samples were stored for 24 h and maintained at ambient temperature prior to analysis (Thrimawithana *et al*., 2010).

TA-XT plus texture analyzer was used to evaluate the compressibility of carrageenan produced. Each run was replicated three times. A Load Cell of 5 kg was used where the maximum force is ±5 kg. A fixture and an analytical probe (P 0.5/R = 12.7 mm diameter) were compressed into the sample to a depth of 15 mm at a cross-head speed of 2 mm sec^{-1}.

Experimental design and optimization: Based on previous research works (Arvizu-Higuera *et al*., 2008) showed, ranges of alkali treatment process parameters were extraction temperature (60-85°C), extraction time (30-180 min) and KOH concentration (5-12% w/w). In this study, extraction temperature was maintained in the range of 70-80°C, only KOH concentration and extraction time was manipulated. Thus the experiment was designed to use the range of process parameters as shown in Table 1.

The experimental runs and the conditions were conducted as suggested by Design-Expert® Software version 7 with Central Composite Design (CCD) of RSM (Bono *et al*. 2008; Shouqin *et al*., 2007).

The optimum condition of cooking process parameters were established using procedure included in Design-Expert® Software. The input to the procedure was the range of process parameters as used in the experimental runs, whereas the target responses was chosen as the maximize condition.

Table 1: Range of extraction process

Factor	Lower limit	Upper limit
3KOH concentration (% w/w)	6	9
Extraction time (min)	45	90

RESULTS AND DISCUSSION

Based on experimental design suggested by Design-Expert® Software version 7, 13 runs of experiments were conducted (Krishnaiah *et al*., 2012). The results presented in Table 2.

The ANOVA analysis depicted that the experimental results can be fitted into a model, as shown in the following Eq. 1-4 in terms of actual factors which A is KOH concentration (% w/w) and B is extraction time (min).

$$\text{Viscosity} = 71.21-39.13A+25.97B+23.40A^2+5.90B^2-25.25AB \tag{1}$$

$$\text{Powder weight} = 5.06+0.18A+0.20B+0.15AB \tag{2}$$

$$\text{Particle size} = 92.21-11.53A-35.87B \tag{3}$$

$$\text{Gel Strength} = 91.52-0.93A+1.29B-17.83A^2+17.51B^2+0.0015AB \tag{4}$$

Effect on viscosity of SRC: Analysis on viscosity of carrageenan showed in the range 54.6-194.8 cP. Figure 1 shows effect of extraction time and KOH concentration on viscosity for semi refined carrageenan powder. It can be seen that the viscosity clearly increased as the extraction time whereas the opposite trend showed on KOH concentration.

Effect on weight obtained for SRC powder: Figure 2 depicted that weight of SRC powder with various KOH concentration and extraction time. It could be seen powder weight slightly increased as extraction time and KOH concentration increased. Extraction time and KOH concentration was not really effected on weight of SRC powder. Analysis showed powder on weight in the range 4.801-5.670 g.

Effect on particle size of SRC: The particle size of the SRC powder showed about 42.31-163.42 μm in the range KOH concentration 6-9% and duration of extraction around 45-90 min. Figure 3 show longer extraction time at

Table 2: Experimental result

Run	KOH conc. (% w/w)	Extraction time (min)	Viscosity (cp)	Powder weight (g)	Particle size (μm)	Gel strength (g cm^{-2})
1	9.00	45.00	54.60	4.88	102.76	60.41
2	7.50	67.50	68.20	5.10	92.31	96.91
3	7.50	67.50	74.50	4.99	88.81	90.81
4	9.00	67.50	58.80	5.21	97.64	61.40
5	6.00	90.00	194.80	5.01	98.73	59.87
6	9.00	90.00	64.80	5.67	42.31	61.47
7	7.50	45.00	62.00	4.98	143.97	63.21
8	7.50	90.00	96.40	5.20	53.87	68.91
9	6.00	45.00	83.60	4.80	163.42	58.87
10	7.50	67.50	78.40	5.00	83.40	95.78
11	6.00	67.50	134.60	4.90	49.75	70.09
12	7.50	67.50	70.40	5.00	92.11	97.21
13	7.50	67.50	60.30	5.01	89.71	92.80

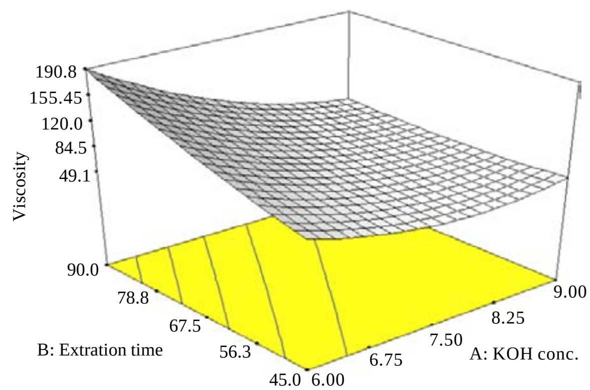

Fig. 1: Effect of extraction time (min) and KOH concentration (% w/w) upon viscosity (cP)

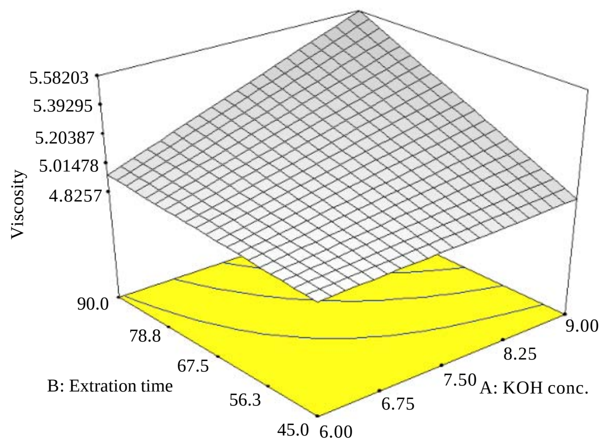

Fig. 2: Effect of extraction time (min) and KOH concentration (% w/w) upon powder weight (g)

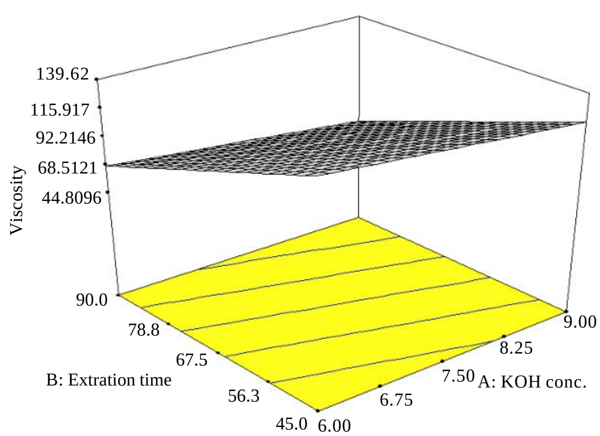

Fig. 3: Effect of extraction time (min) and KOH concentration (% w/w) upon particle size (µm)

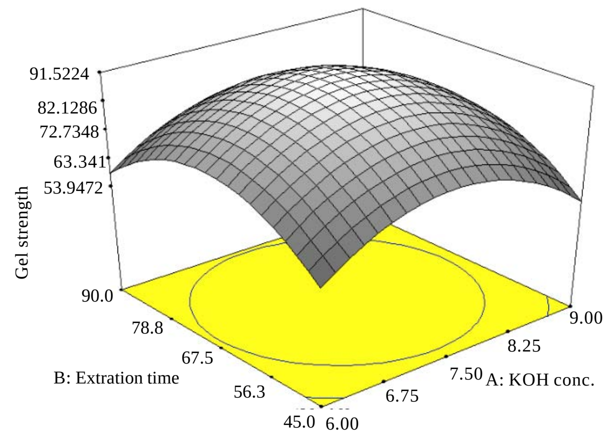

Fig. 4: Effect of extraction time (min) and KOH concentration (% w/w) upon gel strength (g cm^{-2})

Table 3: Optimization Target for the response

KOH concentration (% w/w)	Extraction time (min)	Viscosity (cP)	Powder weight (g)	Particle size (μm)	Gel strength (g cm^{-2})
6.70	74.70	111.8	5.01	86.88	85.61

90 min with higher concentration gives the smallest particle size which is about 42.31 μm. This is show smaller particle size produced by increasing the KOH concentration and extraction time.

Effect on gel strength of SRC: Figure 4 depicted that the effect of extraction time on gel strength is slightly changed with the variation of KOH concentration. However, the strength decreased significantly with the KOH concentration at low and high extraction time.

Optimum formulation for semi-refined carrageenan: Through desirability of result analysis, the parameters for process conditions and effect of SRC can be determined. SRC with sufficient amount of KOH concentration and seaweed extraction time had been developed in order to obtain maximum weight of SRC powder, viscosity and gel strength and smaller particle size. The goal for parameters can be tabulated in Table 3 for desirability solution.

CONCLUSION

In a nutshell, optimization of different extraction process conditions for production of SRC powder from *Kappaphycus alvarezii* with respect to viscosity, weight, particle size and gel strength obtained had been carried out. From RSM, viscosity of carrageenan decreased as seaweed extraction time and concentration of potassium hydroxide (KOH) increased. However, weight of semi-refined carrageenan powder increased as seaweed extraction time and concentration of potassium hydroxide (KOH) increased.

Opposite with particle size analysis, show that extraction time and KOH concentration is inversely proportional with particle size of carrageenan. The results show that smaller particle size produced when extraction time and KOH concentration increases. For gel strength of carrageenan, it show slightly changed with the variation of extraction time and KOH concentration. Hence, gel strength is directly proportional to extraction time and KOH concentration. This is because the higher concentration of KOH, the more 3,6-anhydrogalactose will form and hence increase the gel strength.

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