THE STUDY OF DRYING PARAMETER OF CARRAGEENAN USING SPRAY DRYER

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ABSTRACT

This research is conducted to study the effect of inlet temperature of spray dryer, concentration of feed solution, and the feed flow rate of spray dryer on the moisture content, outlet temperature, yield, and the existent of carrageenan compound in the seaweeds powder produced by spray dryer. Prior the spray drying, the extraction process needs to be carried out. The extraction process involves five steps including pre-rinsing carrageenan containing seaweed, cooking the seaweed with an aqueous solution containing potassium hydroxide (KOH), washing the seaweed in neutralizing bath, rinsing the seaweed in water, and last but not least is drying and chopping the seaweed. Then, the extracted carrageenan solution is fed into the spray dryer to get the powder form product of carrageenan. Analysis on the outlet temperature of the spray dryer gives the increasing reading of outlet temperature when the inlet temperature and the feed concentration value were increased while decreasing with the increasing of the pump speed. For the analysis on moisture content, the increasing of inlet temperature and pump speed resulting the decreasing of the moisture content. The highest yield was obtained when the feed concentration was high and low at the low inlet temperature and pump setting. While the moisture content was increasing when the feed concentration was increases. The study showed that the carrageenan compounds exist in all the products and not very affected by those parameters changing by undergoing the FTIR analysis. This shows that spray drying method can protect the composition of polymer contained in the carrageenan from being denatured.

KAJIAN MENGENAI KAEDAH PENGERINGAN KARAGENAN MENGGUNAKAN PENGERING SEMBURAN

ABSTRAK

Kajian ini dijalankan untuk mengkaji kesan suhu masuk pengering semburan, kepekatan larutan suapan, dan kadar aliran suapan pengering semburan terhadap kandungan kelembapan, suhu outlet, hasil, dan kewujudan karagenan dalam serbuk rumpai laut yang dihasilkan oleh pengering. Sebelum dikeringkan,proses pengekstrakan perlu dijalankan. Proses pengekstrakan melibatkan lima langkah termasuk pra-bilas karagenan yang mengandungi rumpai laut, memasak rumpai laut dengan larutan akueus yang mengandungi kalium hidroksida (KOH), membasuh rumpai laut , membilas rumpai laut dengan air, dan pengeringan serta mencincang rumpai laut. Kemudian, ekstrak karagenan dikeringkan menggunakan pengering semburan untuk mendapatkan serbuk karagenan. Analisis pada suhu alur pengering semburan memberikan bacaan peningkatan suhu alur apabila suhu masuk dan nilai suapan kepekatan meningkat manakala berkurangan dengan peningkatan kelajuan pam. Bagi analisis pada kandungan lembapan, peningkatan suhu masuk dan kelajuan pam disebabkan penurunan kandungan lembapan. Manakala kandungan lembapan meningkat apabila kepekatan suapan meningkat. Produk Karageenan terhasil paling banyak pada kadar aliran suapan pengering semburan paling tinggi dan rendah pada kelajaun pam dan suhu masuk yang rendah. Kajian telah menunjukkan bahawa sebatian karagenan wujud dalam semua produk dan tidak begitu dipengaruhi oleh mereka perubahan parameter dengan menjalani analisis FTIR. Ini menunjukkan bahawa kaedah pengeringan semburan boleh melindungi komposisi polimer yang terkandung dalam karagenan dari dinaturasi.

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α - Alpha

β - Beta

CHAPTER 1

INTRODUCTION

1.1 Introduction

Carrageenans or carrageenins are family of linear sulfated polysaccharides that are extracted from red seaweeds. Carrageenans are used to gel, thicken, or suspend, therefore they are used in emulsion stabilization, for syneresis control, and for bodying, binding and dispersion. Major uses of carrageenans are in foods, particularly dairy applications. Carrageenans products in powder form have high demand in industry than in liquid form as it has many advantages than the liquid form. The moisture removed

from the seaweed prevents it from decaying. Dried seaweed is compact so that less storage space is needed and also keeps it in good condition.

In order to produce semi-refined carrageenans, an extraction process needs to be done. The method of extracting the carrageenan is always start by cooking the seaweed with Potassium Hydroxide (KOH) (Mishra *et al.*, 2006). Then, washing the solid materials after drained the liquid, chop, dry and grind the solid materials prior the melting of the solid.

Drying is the oldest method of preserving food, and dried products which have useful attributes in terms of good storage stability, economical way of transport and unique structural qualities. Drying also is one of the most important unit operations and has probably the widest applications in the food and pharmaceutical industries. There are three common drying methods used to create particles from solutions which namely as freeze-drying, spray-drying and drum drying.

Based on Masters (1991), spray-drying is one of the well-established methods for producing dry powders and it is the direct opposite to the spray-freeze-drying technique, i.e. an atomized spray is contacted with hot gas which is used as the drying medium. Then, evaporation takes place to yield dried particles, which are subsequently separated from the gas stream by a variety of methods. Currently, spray drying is the most preferred method for producing whey proteins in powder form. Normally, it comes at the end-point of the processing line, as it is an important step to control the final product quality (Anandharamakrishnan, 2008).

Recently, many techniques have been developed to microencapsulate food ingredients, spray-drying is the most common technology used in food industry due to low cost and available equipment. Microencapsulation by spray-drying has been successfully used in the food industry for several decades (Gouin, 2004) and this process is one of the oldest encapsulation methods used since the 1930s to prepare the first encapsulated flavors using gum acacia as wall material (Shahidi and Han, 1993).

1.2 Problem Statement

Carrageenans in powder form have high demand in industries because of their wide application in industries especially in food and dairy industry as gelling and thickening agents and also in nutraceutical industry for encalpsulation purposes. Dried carrageenan has many advantages than the fresh one. Basically, the drying process preserves the carrageenan by removing enough moisture from the food to prevent it from decaying. The dried carrageenans are compact so that less storage space is needed and also can keep well.

In normal application, carrageenan will be recovered by alcohol precipitated or potassium chloride to separate it from soluble impurities. There is also other method for carrageenan recovery which is by drying. However, the cost for the alcohol precipitation method is higher than the drying method. Therefore, drying method is more prefer rather than other methods.

In this research, spray drying technique is used to produce carrageenans in powder form which is known as refined carrageenans. Spray drying is a method of producing a dry powder from a liquid or slurry by rapidly drying with a hot gas. Nowadays, dried carrageenans have high demand in industries as they are valuable as ingredients in wide varieties of prepared food. By using spray drying technique, the product particle size can be controlled and also able to retaining and protecting some of volatile compounds (Obon *et al.*, 2005).

1.3 Objectives

i. To investigate the effect of inlet temperature on outlet temperature, moisture content, and the yield of carrageenan compound in the seaweeds powder produced by spray dryer.

ii. To determine the effect of concentration of feed solution on outlet temperature, moisture content, and the yield of carrageenan compound in the seaweeds powder produced by spray dryer.

iii. To study the effect of pump speed of spray dryer on outlet temperature, moisture content, and the yield of carrageenan compound in the seaweeds powder produced by spray dryer.

1.4 Scope of Study

In order to achieve the objectives of this research, the scope of the study has been determined:

i. To extract carrageenan from seaweed using solvent extraction method.

ii. To investigate the effect of inlet temperature on outlet temperature, moisture content, and the yield of carrageenan compound in the seaweeds powder produced by spray dryer using various temperature of 140 °C, 160 °C, 180 °C, and 200 °C.

iii. To determine the effect of concentration of feed solution on outlet temperature, moisture content, and the yield of carrageenan compound in the seaweeds powder produced by spray dryer using various concentration of feed solution of 0.05 g/ml, 0.15 g/ml, 0.20 g/ml, and 0.25 g/ml.

iv. To study the effect of pump speed in spray dryer on outlet temperature, moisture content, and the yield of carrageenan compound in the seaweeds powder produced by spray dryer using various pump speed of 4, 6, 8, and 10.

1.5 Rationale and Significance of the Study

Malaysia has high potential to be a main supplier of carrageenans as in Malaysia especially Sabah and Sarawak are two states in East Malaysia which have a great potential for seaweed culture. There are several seaweed species found growing

naturally on reefs in Semporna area, south of Sabah and in Banggi Island of the South China Sea and in Kudat area, in north Sabah.

CHAPTER 2

LITERITURE REVIEW

2.1 Carrageenans

Carrageenans are sulfated linear polysaccharides extracted from certain red seaweed of the Rhodophyceae class. Usually the main species of Rhodophycae used in the commercial production of carrageenans are *Euchema cottonii* and *E. spinosum*. These are spiny bushy plants, about 50cm high, which grow on reefs and in shallow lagoons around the Philippines and Indonesia and other island coasts in the Far East including Malaysia. *E. cottonii* yields kappa carrageenan and *E.spinosum* contains iota

carrageenan. Carrageenan extracted from this species consists of both kappa and lambda types even though it has been shown that these do not occur within the same plant but in individual plants which grow together (McCandless *et al.*, 1973).

Carrageenan is found in seaweed of Rhodophyceae (red algae) class from which it can be isolated. Carrageenan does not exist as a free polymer in the red algae but exist in the part of the skeleton of the algae. The occurrence and distribution of the various carrageenan types I Rhodophyceae is dependent on the species, location and life cycle of the seaweed. Carrageenan can be found in the Gigartinaceae and Solieriaceae families and particularly in the species belonging to the Gigartina, Chondrus, Eucheuma, and Iridaea.

Carrageenans are broadly used in the most industries comprise main application in food industry, dairy industry, pharmaceutical and many more industries as thickening, gelling agent and more recently used in the food industry as excepient in pill and tablets. According to Robledo and Freile-Pelrgrin (2010), steady supply of raw material from reliable sources is requisite to meet and maintain the required volumes and qualities. The recent demand for carrageenans has required the development of cultivation as well as search for promising resources mainly in the tropics.

2.2 Extraction Carrageenan from Seaweed

The extraction method of carrageenans is always undergo same processing steps which are pre-rinsing carrageenan containing seaweed, cooking the seaweed with an aqueous solution containing potassium hydroxide (KOH), washing the seaweed in

neutralizing bath, rinsing the seaweed in water, and last but not least is drying and chopping the seaweed.

In the pre-rinsing step, Mishra *et al.* (2006) rinse the fresh dried seaweed with freshwater to remove off debris and dried it in an oven at 90ºC for 5 h. The cooking step requires the seaweed to be cooked with an aqueous alcohol solution. KOH was decided as the best solvent in this step as it gives better yield and quality gels with high viscosity of carrageenans which also proof the fact that *Kappa* carrageenan forms strong, rigid and slightly opaque gels when potassium ions are present. They link to the sulphate groups causing an anti-parallel double helix structure (Ross Kealy, 2003). The temperature used in this step are within 60℃ – 90℃ with the cooking time between the range of 45 minutes – 5 hours as mentioned (Mishra *et al.* 2006) and Distantina *et al.* (2011).

2.3 Structure of Carrageenan

Carrageenan containing galactose units which are sulfated in the 6-position can form 3,6-anhydro units by elimination of sulphate by ring formation by treatment with a base. The alkali modification treatment can provide a carrageenan material with improved gel properties, assuming the carrageenan contains 6-sulphatted galactose units.

Carrageenan is normally soluble in warm water where it wills forms high viscosity solutions, and insoluble in most organic solvents. Referring the Encyclopedia of Chemical Technology (1980), there are different types of carrageenan such as kappa,

iota, lambda, ny, and my are known. Those types of carrageenans can be differentiated according to the nature of their repeating galactose units. The most popular and important for commercial purposes are kappa, iota, and lambda carrageenan. The structure of carrageenan is shown in Figure 2.1.

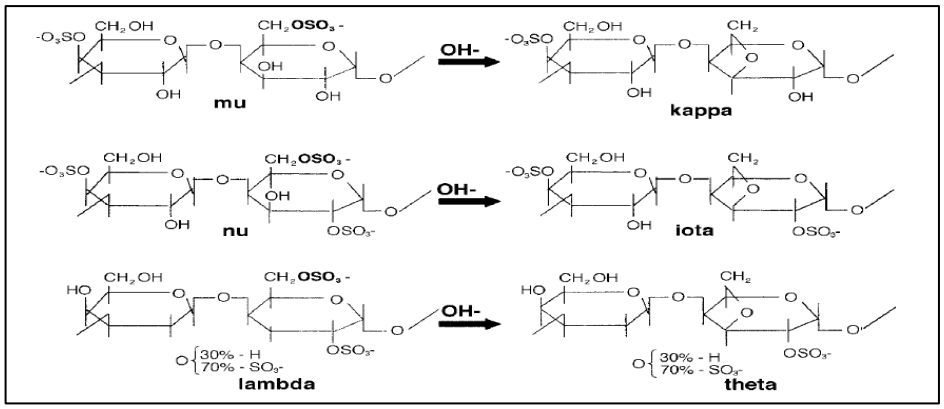

Figure 2.1: Structure of Carrageenan

2.4 Drying Method for Seaweed Processing

Carrageenan that is obtained by extraction of the seaweed into water or aqueous dilute alkali can be recovered by alcohol precipitation, by drum drying, or by precipitation in aqueous potassium chloride and subsequent freezing. Drying is one of the steps which are recovering the carrageenan. There are several methods of drying carrageenan such as drum drying, freeze drying, and spray drying. Agar production including carrageenan by modern industrial freezing techniques was initiated in 1921 in California, U.S.A. by Japanese named Matsuoka. Lemoine *et al*. (2008) used freeze-dryer in the presence of 20 mM ammonium carbonate to dry the extracted carrageenans. Drum drying also one of the methods to dry the extracted carrageenan which form the

large component particles as powder. Drum drying is one of the most energy efficient drying methods and is particularly effective for drying high viscous liquid or pureed foods.

During the Second World War, the production of agar including carrageenan commenced in Portugal and Spain as well. Until today, the carrageenan in powder form has high demand in industry compared to the fresh carrageenan. Dried carrageenan has many advantages as the drying process preserves carrageenan by removing enough its moisture content which can prevent it from decaying.

2.5 Spray Drying

2.5.1 Introduction of Spray Drying

The practice of each method is depending on the types of desired microparticles. The properties of final product such as wall structures, sizes, shape quantity and quality are included into consideration. Nevertheless, the cost of operation, practicality of the process and overall time frame are also will be considered. In industry, spray drying is more preferred due to its continuous production and easiness of industrialization (Shu *et al*., 2008).

Spray drying is a method of producing a dry powder from a liquid or slurry by rapidly drying with a hot gas. Spray drying is one of the best drying methods to convert, in a single step, fluid materials into solid or semi-solid particles. Rapid evaporation

keeps the droplets temperature relatively low will not affect the quality of product (Roustapour *et al.*, 2009). Therefore, contribution of spray drying in food and dairy industries are increasing as compared to other conventional drying methods. Obon *et al*. (2009) stated that the spray drying technology has the ability to quickly transform seaweeds into dried particulate form and has the ability to control the particle size distribution. The product in powder form from spray drying will result in much reduced volume and longer shelf life (Cano-Chauca *et al*., 2005), showing that the spray drying is cost-effective process compared to other methods.

Atomization of fluid feed, drying medium and spray contact, drying of feed, and separation of product from air are four main stages in spray drying which these stages and their operational parameters have major impact on the drying efficiency and the final product properties (Sollohub *et al*., 2010).

Spray dryer is the equipment used for spray drying. The schematic diagram of spray dryer is shown in Figure 2.2.

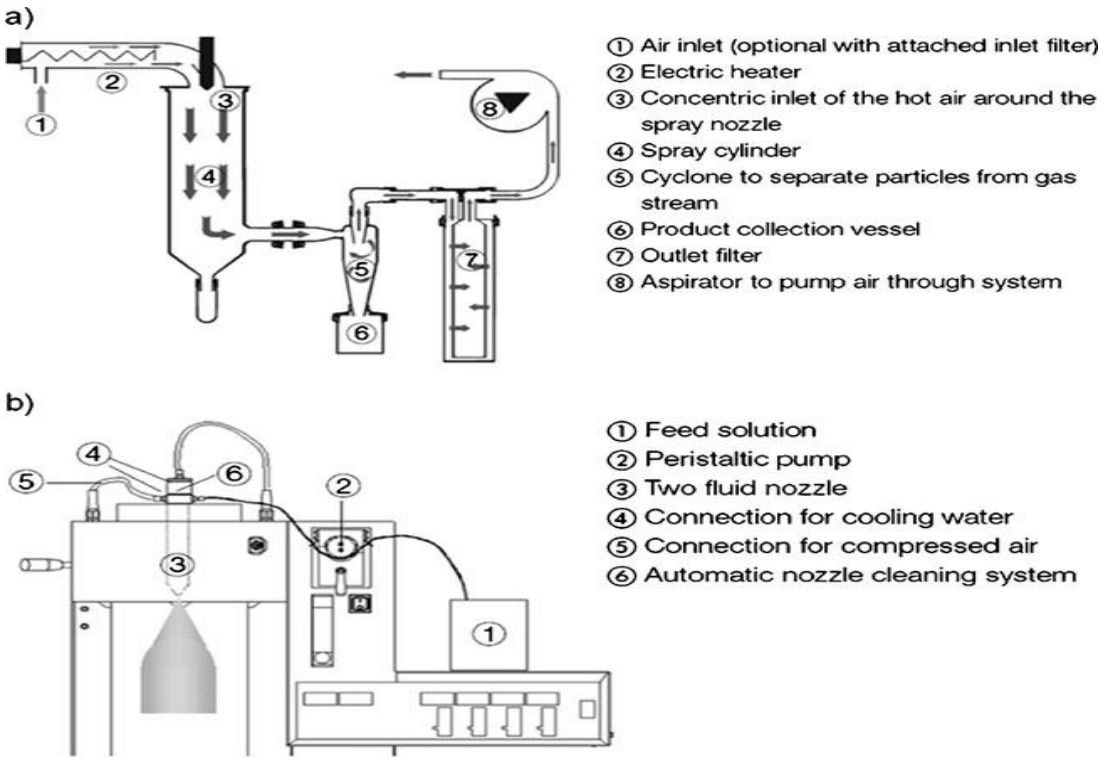

Figure 2.2: A Schematic Diagram of a Spray Dryer. (a) Air flow, (b) feed delivery (Buchi Corporation, 2002)

The continuous design and flexibility of the spray dryer allows it to deliver powder of a specific particle size and moisture content regardless of the dryer capacity or product heat sensitivity (Al-asheh *et al*., 2003). The spray dryer delivers a highly controlled powder quality in the continuous operation. In addition, the surface area produced by atomization of the liquid feed enables a short gas residence time which allows spray drying without thermal degradation. This permits the short time production of the spray drying product because there is no product hold up in the spray dryer.

However, fruit juices that were obtained using spray drying process may have some constraints in their physical and chemical properties such as stickiness and solubility, due to the present of low molecular weight sugar and acids (Tonon *et al*., 2008). The stickiness problem in spray drying is related with the low glass transition temperature (T_g). Maintaining chamber wall temperature lower than the T_g of the powder could reduce deposition of such powders. In order to dry the solution without deposition, the T_g must be controlled. Basically the sticky behavior of fruit powder depends on the sugar content and temperature of the product. Roos and Karel (1991) and Slade and Levine (1994) stated that sticky point of food powders decreases with decreasing molecular weight and products with low T_g have also sticky point at low temperatures.

2.5.2 Principles of Spray Drying

Different types of spray dryers are used for various purposes in different field and applications. In 1901, the first spray drying patents was applied by German Mr. Stauf, who sprayed the milk by nozzles into a chamber with warm air. However, in USA, in 1913, the Amarican Mr. Grey and the Dane Mr. Jensen developed a nozzle spray dryer and started to produce and sell the drying installations on a commercial scale. On the other hand, in 1912, German Mr. Kraus developed the first rotary spray dryer but not until 1933, the Danish engineer, Mr. Nyrop found the real breakthrough of atomization with his patent (Arati Parihari, 2009). Spray drying consists of the following unit operations: