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Optimal Operating Conditions of Spray Dried Noni Fruit Extract using κ-carrageenan as Adjuvant

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Abstract: A detailed study was conducted using lab scale spray dryer to produce micro particles using κ-carrageenan (1 wt.%) as the encapsulation or binding agent by different ratio, M_{core}/M_{wall} (1:1, 1:2, 1:4 and 1:6) at different temperature (90, 100, 120 and 140°C). The concentrated noni extract and spray dried noni micro particles were analyzed for encapsulation yield, DPPH scavenging activity, total phenolic content and particle size analysis. From the results it was clear that percentage of DPPH scavenging activity and total phenolic content was slightly higher for 1:6 at 90°C than 1:2 at 90°C. However, 1:2 at 90°C was concluded as optimal operating conditions. By particle size analyzer it was found that at optimal operating spray drying condition, the mean diameter of the particle was varied from 2.53 to 2.27 μm, which is found to be less when compared to all the other ratios at different temperatures.

Key words: Noni extracts, spray drying, dried extract, antioxidant activity

INTRODUCTION

Morinda citrifolia L., the noni has been used in traditional Polynesian medicine for more than 2000 years. They belong to the family Rubiaceae and commonly found in the Hawaiian and Tahitian islands. They usually grow in open coastal regions at sea level and in forest areas upto about 1300 feet above sea level. The bark, stem, root, leaf and fruit have been used traditionally as a folk remedy for many diseases including diabetes, hypertension and cancer (Hirazumi *et al*., 1994, 1996). These diseases are due to production of more Reactive Oxygen Species (ROS) such as superoxide anion radical, hydroxyl radical and hydrogen peroxide than enzymatic antioxidant such as superoxide dismutase (SOD), glutathione peroxidase (GPx), catalase etc. and non-enzymatic antioxidant such as ascorbic acid (vitamin C), α-tocopherol (vitamin E), glutathione, carotenoids, flavonoids etc. This normally occurs when our body is in under stress. Since, *Morinda citrifolia* L. claims to have high antioxidant potential, it can supplement as antioxidant compounds with food items and as preventive medicine.

The polynesians utilized the whole noni plant in various combinations for herbal remedies. Ripe fruit of this evergreen shrub has a strong butyric acid smell, flavor and high phenolic content. The fruit can grow to a size of 12 cm. The fruit juice is in high demand in alternative medicine for different kinds of illness such as arthritis, diabetes, high blood pressure, muscle aches and pains, menstrual difficulties, head aches, heart disease, AIDS, cancer, gastric ulcers, sprains, mental depression, senility, poor digestion, atherosclerosis, blood vessel problems and drug addiction. Scientific evidence of the benefits of noni fruit juice is limited but there is some evidence for successful treatment of colds and influenza. The juice of noni fruits has also been shown to prolong the life span of mice implanted with Lewis lung carcinoma. It was also proposed that the fruits of noni might suppress the growth of tumors by stimulating the immune system (Hirazumi *et al*., 1994). The fruits of this plant were also used as foods in time of famine, however very few reports are available on their chemical composition.

Levand and Larson (1979) identified several compounds including acetyl derivatives of asperuloside, glucose, caproic acid and caprylic acid in fruits. Wang *et al.* (1999) isolated two glycosides such as rutin and asperulosidic acid and novel trisaccharide fatty acid ester from the fruits of noni in one of the other study. Carboxylic acids especially octanic acid and hexaonic acid

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have also been identified from the fruits. About 51 volatile compounds have also been found in the ripe fruit (Sang *et al*., 2001), including organic acids (mainly octanic and hexanoic acids), alcohols (3-methyl-3-butene-1-ol), esters (methyl octanoate, methyl decanoate), ketones (2-heptanone) and lactones (E-6-dodeceno-γ-lactone) (Farine *et al*., 1996). Noni juice is also recommended by 50 medical doctors in USA and Hawaii to help relieves their patients from about 23 different conditions, such as allergy, anti-aging, arthritis, cancer, diabetes (type I and II), digestion, muscle pain, high blood pressure etc. and to increase the feelings of well being (Solomon, 2000). From the literature review it was clear that the fruit has antibiotic and antioxidant properties, but still there is a lack of scientific evidence by *in vivo* study.

Spray drying is the most popular drying technology used within the foods, chemical and pharmaceutical industries for production of dry particles from liquids. Hence, it can be used to turn the noni extract 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. (Souza and Oliveira, 2006). The aim of this study is the evaluation of the effects of processing parameters of spray drying on free radical scavenging activity, total phenolic content and particle size of spray dried noni extract.

MATERIALS AND METHODS

Chemicals: Methanol, ethyl acetate (HPLC grade) were obtained from J.T. Baker (USA). Folin Ciocalteau reagent and Tannic Acid (TA) were purchased from Merck, Germany. 2,2-Diphenyl-l-picrylhydrazyl (DPPH) and sodium carbonate were from Sigma chemicals (St. Louis, USA). 5 kg of *Morinda citrifolia* L. fruit before ripening stage were picked from local area in Kota Kinablu, Sabah, Malaysia. All other chemicals of reagent grade and were used without further purification.

Preparation of plant extracts: The *Morinda citrifolia* L. fruit was tap washed followed by washing with distilled water. The fruit was peeled first and core (pulp and seed) was cut into small species. The skin, pulp and seed were sun dried for 2 days. Then, the sample was kept at 60°C at hot air oven for 1 day to remove all the moisture content. The dried fruit was then finely powdered by using mixer. Two hundred and fifty milliliter of ethyl acetate was added to 25 g of powdered sample (10 wt.%) and extraction was done at water shaker bath at 35°C for 3 days. The supernatant was then separated from the residue by filtration using Whatmann No. 1 filter paper. The extracted solution was stored in a closed container and kept at 4°C before being analyzed.

Selection of the drying excipient and spray drying of the noni fruit extract: In the pharmaceutical and food industries, the correct selection of the drying excipient is an important step to guarantee the stability and the quality of finished product (Georgettia *et al*., 2008). Hence, literature survey was made in order to select an adequate drying excipient to be added to the noni fruit extract before spray drying. Drying adjuvants generally added in order to improve both the dryer performance and product properties. In this study, optimal operating condition of spray dryer was obtained for the selected drying adjuvant κ-carrageenan, with respect to antioxidant activity and total phenolic content of the spray dried powder.

κ-carrageenan (1 wt.%) and noni fruit extract were mixed with different volume ratio of 1:1, 1:2, 1:4 and 1:6 and stirred to form an aqueous solution. The resulting emulsion was then spray dried on a lab plant spray dryer SD-05 (pilot scale), with co-current flow regime (the spray-dried product and the drying air flow are in the same direction). The drying chamber has diameter of 215 mm and height of 500 mm. The main components of the system were the feed system of the noni fruit extract, constituted by peristaltic pump, a fluid atomizer (inlet orifice diameter of 0.5 mm) and an air compressor; a feed system of the drying gas, constituted by a blower and a air filter; a temperature control system of the drying gas and a product control system (cyclone). The feed flow rate was kept at 315 mL h^{-1}. The flow rate of the drying air was fixed at 60 m^3 h^{-1} and the atomizing air at a pressure of 1.1 bar. After spray drying, the powders were collected through a high efficiency cyclone in a glass container, transferred in a glass vial and stored in the desiccators at ambient temperature.

Several spray drying runs were carried out in order to investigate the effects of the inlet drying gas temperature for different volume ratio of noni fruit extract to excipients. Further, analyses were made to find out the antioxidant activity and total phenolic content of the spray dried powder.

Encapsulation Yield (EY): The Encapsulation yield was calculated as the ratio of the mass of microcapsules obtained at the end of the process to the mass of initial substances added including adjuvant and noni spray dried powder.

$$EY\ (\%) = \frac{\text{Weight of microcapsules after spray drying}}{\text{Total weight of adjuvants and noni spray dried powder}} \times 100$$

DPPH radical scavenging activity: 2,2-Diphenyl-1-picrylhydrazyl is a stable free radical that reacts with compounds, which are able to donate a hydrogen atom. Thus, the hydrogen donating ability of concentrated noni fruit extract and spray dried noni fruit extract was determined from the change in absorbance at 515 nm as per Blois (1958) method with slight modification. For radical scavenging measurements, sample in methanol solution were prepared by dissolving 10 mg of spray dried powder in 30 mL of methanol and centrifuge for 10 min using Sartorius Sigma 3-18 K centrifuge. Aliquots of supernatant were added in 3 mL of 0.025 g L^{-1} DPPH in methanol. The change in absorbance was measured after 40 min at room temperature using 4802 UV-VIS double beam spectrophotometer. Methanol was used as reference. DPPH (0-100 mg L^{-1}) was used to obtain standard calibration curve. All measurements were made in triplicate.

The DPPH radical scavenging activity in terms of percentage was calculated according to the following equation.

$$\text{DPPH scavenging activity (\%)} = \frac{1 - \text{Abs}_{515}\ \text{sample}}{\text{Abs}_{515}\ \text{DPPH solution}} \times 100$$

Total phenolic content: Total Phenolic Content (TPC) was determined according to the FC method (Slinkard and Singleton, 1977) with slight modification. Briefly, sample in methanol solution was prepared by dissolving 10 mg of spray dried powder in 30 mL of methanol and centrifuge for 10 min using Sartorius Sigma 3-18 K centrifuge. The supernatant of sample extract (0.5 mL) was added to 2.5 mL of 0.2 N FC reagent and allowed to react for 5 min. Later 2 mL of 75 g L^{-1} of sodium carbonate was added to the reaction mixture and made to 25 mL using distilled water. Finally, the reaction mixture was incubated for 2 h at room temperature and absorbance was measured at 760 nm using 4802 UV-VIS double beam spectrophotometer. Methanol was used as reference. Tannic acid (0-100 mg L^{-1}) was used to produce standard calibration curve. The total phenolic content was expressed in mg of Tannic acid equivalents (TAE/g of spray dried powder).

Particle size analyzer: The particle size of the spray dried powder was determined using laser scattering particle size distribution analyzer. The measurements were made in triplicate.

RESULTS AND DISCUSSION

Spray dried microparticles of noni fruit extract: Several drying techniques can be utilized but spray drying is the mostly commonly used technique in the herbal processing industries. During the spray-drying process, the adjustable parameters include inlet temperature (°C), air flow rate (m^3 h^{-1}), liquid flow rate (mL h^{-1}) and compressed air pressure. In addition the ratio of M_{core}/M_{wall} can also be changed during the experiment. In this study, inlet temperature and the ratio of M_{core}/M_{wall} were changed to find out the optimum operating condition of spray dryer to obtain noni micro particles (Fig. 1).

The EE is one of the major parameters measuring the spray-dried microcapsules, higher EE is expected to be

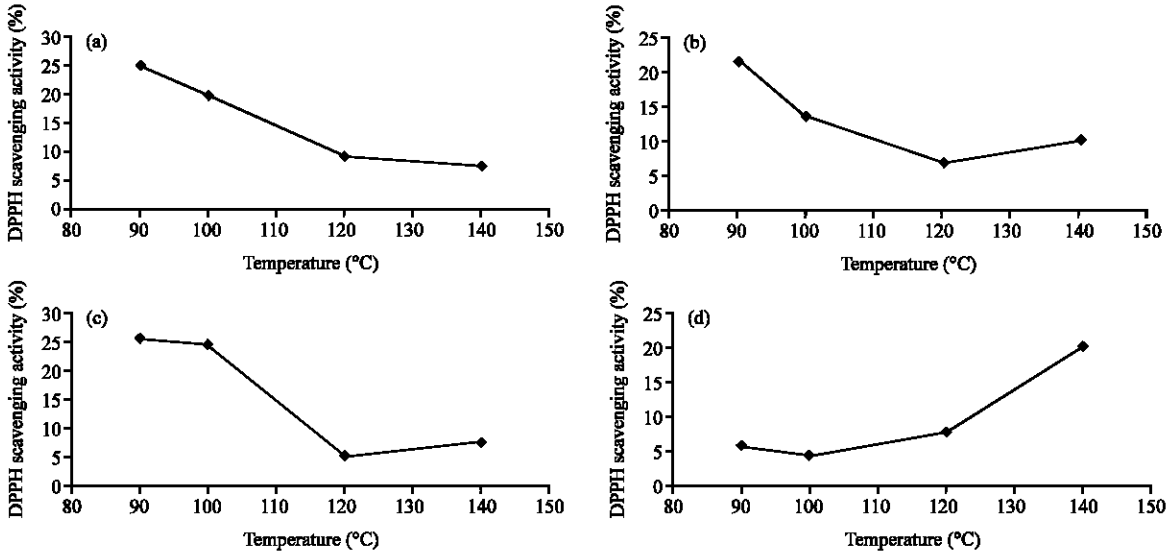

Fig. 1: Percentage of DPPH scavenging activity of spray dried noni fruit extract with respect to temperature for, (a) 1:2 M_{core}/M_{wall}, (b) 1:4 M_{core}/M_{wall}, (c) 1:6 M_{core}/M_{wall} and (d) 1:1 sM_{core}/M_{wall}

obtained. During the spray-drying process, temperature and the ratio of M_{core}/M_{wall} are the two key factors affecting on the EE. Outlet temperature is mainly determined by inlet temperature, higher inlet temperature will lead to higher outlet temperature. When the drying time was longer, it may cause accumulation of some under-dried larger particles on the chamber wall and lead to low EY and hence optimum pump flow rate was fixed. When inlet temperature was high, thus will lead to high outlet temperature, it can also be found that lots of particles accumulated on the chamber wall and thus also lead to a low EY. During spray-drying process, spray gas flow rate has little effect on the products properties. If the feed solution rate is fast, the water will not vaporize fully in short time and the spray dried powder can't dry enough and hence the pump rate is fixed as mentioned in methods and materials. If inlet temperature is low, there will not enough quantity of heat to dry the product and thus a quantity of water exist in the product and the wet powders are easily to be stuck on the chamber wall (generally, only those powders collected in the container are regarded as effective), so it will decrease the EY. Therefore, lowest inlet temperature maintained as 90°C, this is also due to the boiling point of the extracted solvent (ethyl acetate) is around 80°C, below which ethyl acetate will not get vaporize.

Encapsulation yield: By the experimental study, it was found that at 1:2 ratio of M_{core}/M_{wall} the EY was higher at lower temperature (90°C) than at higher temperature (100, 120 and 140°C). This may due to at higher temperature all the active components may get volatilized. Even at 90°C, EY was obtained only around 32%. At 1:2 ratio of M_{core}/M_{wall} at 140°C, it was observed that most of the particles obtained by spray drying get accumulated on the chamber wall and hence EY is only around 8%. This may be due to wall stickiness temperature. At 1:1 ratio of M_{core}/M_{wall} the EY was higher at 140°C, when compared to other temperature. At 90 and 100°C, for the same ratio, the EY was minimum and moreover little amount of ethyl acetate was observed in the liquid collector, this may be due to 1:1 ratio may not be appropriate proportion due to insufficient amount of binding material. At 1:1 ratio of 120°C even though the EY is minimum, there is no observance of ethyl acetate extract in the liquid collector. This may be due to at higher temperature either the active components getting attached to the binding material or else getting volatilized. Similarly, at 1:1 ratio of 140°C there is no observance of ethyl acetate extract in the liquid collector. At 1:6 and 1:4 ratio of M_{core}/M_{wall} the more EY was obtained at 90 and 100°C, respectively. In general, when compared to all other ratio, EY was found to be higher at 1:1 ratio of M_{core}/M_{wall} at 140°C for the spray dried Mengkudu fruit extract with κ-carrageenan as binding material. However it was observed that at this ratio (1:1 at 140°C), the DPPH scavenging activity is less.

DPPH and TP of noni microparticles: As the temperature increases at 1:2 ratio of M_{core}/M_{wall}, the percentage of DPPH scavenging activity decreases (Fig. 1a). This may due to at higher temperature the active components may get volatilized. At 1:4 and 1:6 ratio, as the temperature increases percentage of DPPH scavenging activity decreases upto 120°C and increases slightly further when the temperature raise to 140°C (Fig. 1b, c). This may due to at this temperature, some of the active components which are ready to volatilized may attached to the binding material. At 1:1 ratio, as the temperature increases the percentage of DPPH activity increases (Fig. 1d). At lower temperature (90 and 100°C) ethyl acetate was observed in the liquid collector which is also substantiated by lower DPPH scavenging activity. This study found that at 1:6 ratio of M_{core}/M_{wall} at 90°C, percentage of DPPH scavenging activity is 25.32%, which is found to be higher when compared to all other ratio. But 1:2 and 1:6 ratio of M_{core}/M_{wall} at 90 and 100°C, respectively exhibit 24.91 and 24.26% of DPPH scavenging activity. Since there is no much difference in percentage of DPPH scavenging activity between the ratio 1:2 at 90°C and 1:6 at 90°C, it is suggested to select the optimum ratio as 1:2 ratio at 90°C because at this ratio the non active component (excipient) is less and also EY is also maximum. However, the EY for 1:2 at 90°C is only 31.56% (Table 1).

At 1:2 ratio of M_{core}/M_{wall} at 90 and 100°C, 1:4 ratio of M_{core}/M_{wall} at 90°C, 1:6 ratio of M_{core}/M_{wall} at 90 and 100°C

Table 1: Encapsulation yield, percentage of DPPH scavenging activity, total phenolic content and particle size distribution of spray dried microparticles

Ratio M_{core}/M_{wall}	Temperature (°C)	EY (%)	Percentage of DPPH scavenging activity	TPC (mg of TAE/g of SDP)	Mean particle size diameter (μm)
1:2	90	31.56	24.91	30	2.27
1:2	100	26.04	19.57	27	3.96
1:2	120	20.73	9.24	ND	ND
1:2	140	7.71	7.64	ND	ND
1:4	90	11.20	21.66	21	3.48
1:4	100	31.42	13.76	ND	ND
1:4	120	5.15	6.94	ND	ND
1:4	140	27.96	10.24	ND	ND
1:6	90	19.33	25.32	36	3.20
1:6	100	16.07	24.26	36	2.65
1:6	120	9.96	5.30	ND	ND
1:6	140	6.44	7.52	ND	ND
1:1	90	13.07	5.85	ND	ND
1:1	100	16.80	4.38	ND	ND
1:1	120	11.86	7.80	ND	ND
1:1	140	48.13	20.07	27	4.82

SDP : Spray dried particle, ND: Not determined

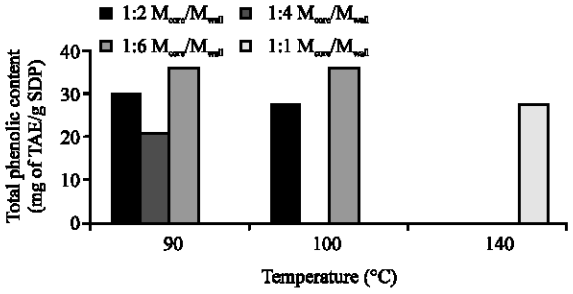

Fig. 2: Total phenolic content of spray dried powder at different temperature for different ratio of M_{core}/M_{wall}

and 1:1 ratio of M_{core}/M_{wall} at 140°C microcapsules were selected for TPC analysis since they exhibit higher percentage of DPPH scavenging activity. Although, FC procedure does not give a full picture of the quantity and quality of the phenolic constituents of the extracts, this widely used method provides a rapid and useful overall evaluation of the phenolic content of extracts. As that of percentage of DPPH scavenging activity, TPC also higher at 1:6 ratio at 90°C and found to be 36 mg of TAE/g of spray dried powder. However, at 1:2 ratio of M_{core}/M_{wall} at 90°C TPC was found to be 30 mg of TAE/g of spray dried powder (Fig. 2).

Correlation: The experimental values of percentage of DPPH scavenging activity of noni micro particles were fitted as a function of the total phenolic contents, in order to detect any relationship between the antioxidant activities of phenolic compounds. The correlation coefficient between the DPPH scavenging activity and total phenolic content of noni micro particles is 0.700. These results suggests that approximately 70% of the antioxidant capacity of the spray dried noni fruit extract may be attributed partly due to the contribution of the phenolic compounds. In fact, the total phenolic content does not incorporated all antioxidant present in the extract (Djeridane *et al*., 2006). This is the reason why samples with similar concentration of the total phenolic content, may vary in their scavenging activity.

Particle size of spray dried mengkudu fruit extract micro particles at 1:2 ratio of M_{core}/M_{wall} at 90°C varied from 2.532 to 2.270 μm. In general all tested samples exhibited a similar particle size distribution, suggesting that the manufacturing method, generated particles within a narrow size distribution regardless of different inlet temperature and different ratio of M_{core}/M_{wall}. However, analysis of particle size distribution revealed that at optimum operating condition, the particle size is found to be less when compared to all other ratios.

CONCLUSION

For the extracts containing volatile components the temperature may be one of the most important problem during food processing, as well as in sample handling and storage. The effect of the temperature on water activity of the dried product is also very important, but insufficiently understood yet. However, the results of this study showed that the highest antioxidant activity and total phenolic content were found at 1:6 ratio of M_{core}/M_{wall} at 90°C. But 1:2 ratio M_{core}/M_{wall} at 90°C is found to be optimum ratio due to presence of less inactive components and also which posses similar antioxidant activity and total phenolic content as that of above. Nevertheless, noni micro particles obtained by this study showed valuable antioxidant activity, opening perspectives for its use as a natural and multifunctional dietary food additive or supplements. This study also would be helpful to the pharmaceutical application of noni fruit. Since, pharmaceutical value of this fruit is very high, hence it is necessary to identify the nutritional and functional compounds it contains and explain their mechanisms of action in order to determine the real potential of this fruit and the technological processes that preserve these properties.

The antioxidant activities of spray dried noni fruit extract is affected by the spray drying temperature, which was clearly understood by this study. κ-carrageenan may be an effective drying aid for spray drying of noni fruit extract. Addition of one or more adjuvants can also be added to find out the synergetic effect on powder activity and properties.

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