Research Paper

Rambutan (*Nephelium lappaceum* L.) Fruit Peel: Possibility of Extraction and Incorporation of the Microencapsulated Crude Extract in Product Development

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Abstract

'Malwane' rambutan cultivar is popular due to its attractive reddish color peel and easily separable thick flesh. High consumption of rambutan has led to accumulation of a considerable amount of the peel, which contains anthocyanin and other bioactive compounds. Thus, the objective of the present study was to explore the possibility of extracting and using microencapsulated extract obtained from rambutan peel in product development. The frozen peels were extracted with water at 50 °C, microencapsulated using maltodextrin, and spray-dried to a powder (MESP). Physicochemical parameters of water extract of frozen peels (WEFP) and MESP were studied. Pasta and Jelly were carefully made using the MESP and assess the effectiveness of incorporating MESP. The Total Polyphenolic Content of the WEFP was 24.52 ± 0.47 mg mL^{-1}, amounting to 12.3% (g g^{-1}) of the frozen peel. Anthocyanin Content of 24.76 ± 0.04 µg g^{-1}, TPC of 373 ± 7 mg g^{-1}, Antioxidant Activity of 81%, Moisture Content of 7.8 ± 0.0%, and water activity of 0.41 ± 0.004 were observed for the MESP. The incorporation of the MESP in pasta imparted a reddish color, not changed during boiling of the finished product. This indicates the ability of MESP to impart color in starch-based products processed at high temperatures and re-constituted in boiling water. However, coagulation and darkening of color were evident with jelly. The results of this study revealed the potential of utilizing discarded rambutan peels.

Keywords: Bioactive compounds, Microencapsulation, Rambutan peel extract

Introduction

Rambutan (*Nephelium lappaceum* L.) is a tropical and seasonal fruit grown originally in Southeast Asian countries and also at present in Central American countries (Maran *et al*., 2017). This fruit is popular due to its refreshing flavor and unusual appearance (Khoo *et al*., 2017), and is mostly consumed as fresh or canned forms (Maran *et al*., 2017). Moreover, presence of bioactive compounds possessing anti-oxidant, anti-microbial, and anti-hyperglycemic activities is also attributable to increased consumption of the fruit (Hernandez´ *et al*., 2017). Therefore, a considerable amount of peels is thrown away, causing environmental problems, particularly in urban areas. A number of studies have reported the value of discarded rambutan peels due to presence of bio-actives possessing antioxidant, antibacterial, and other biological properties (Thitilertdecha *et al*., 2010; Palanisamy *et al*., 2008). Moreover, some compounds in the peel is reported to possess anti-carcinogenic activity and also to be good for cell health (Hernández *et al*., 2017). Furthermore, various *in vitro* and *in vivo* tests have revealed antioxidant, antimicrobial, antidiabetic, antiviral, anti-inflammatory, anti-hypoglycemic, and anti-proliferative effects of phytochemicals in the extracts of rambutan flesh and mainly from the peel (Hernández-Hernández *et al*., 2019).

Among many rambutan cultivars grown in Sri Lanka, '*Malwana*' is the most popular due to large fruit size, attractive red color peel, and easily separable thick flesh. The attractive red color rambutan peel contains anthocyanins, which are reported to be the most abundant flavonoid constituents of fruits and vegetables (Patras *et al*., 2010). Exploring the potential of extracting these pigments, and investigating the possibility of replacing synthetic food colorants with the extract would be beneficial to consumers, particularly due to increased consumer preference for foods containing natural colorants (Özkan & Bilek, 2014). Moreover, the inner whitish thick parts of the peel turn brown upon cutting, which indicates the presence of phenolic compounds other than anthocyanins (Rohman, 2017). The ethanolic extract of rambutan peel is reported to be non-toxic as revealed by acute toxicity studies (Rohman, 2017). Moreover, rambutan peel is reported to be a good source of natural antioxidants, thus can be utilized without discarding as agricultural waste (Sun *et al*., 2011). Therefore, use of the discarded rambutan peels for extraction of these bioactive compounds and other phytochemicals, and exploring the potential of incorporating the peel extract in product formulations would be beneficial to the food industry and environment.

However, most of these constituents in the peel extracts may degrade upon exposure to some food processing and/or environmental conditions such as light, high temperature, pH, and oxygen, possibly due to instability of the unsaturated bonds in the molecular structure of these constituents. Thermal processing is reported to degrade anthocyanins due to thermal instability, mainly attributable to presences of

conjugated double bonds in their structures (Patras *et al*., 2010). Moreover, stability of anthocyanins is affected by the chemical structure as reported by showing the highest stability by cyanidin-3-rutinoside at 95 °C. Also cyanidin and delphinidin rutinosides showed the highest stability at 8 °C (Rubinskiene *et al*., 2005).

Therefore, it is necessary to minimize the rate of degradation of these bioactive compounds during food processing and/ or storage. Microencapsulation of the extract could possibly reduce the rate of such degradations, as bioactive compounds or the core materials are surrounded by a tiny coating or a wall material known as encapsulant, resulting in small capsules (Khoo *et al*., 2017). Moreover, microencapsulation ensures controlled and targeted release of the core materials at a specific rate and also improves their dispersibility (Ghosh *et al*., 2022). Therefore, this study aimed to explore the possibility of extracting the discarded rambutan peels of '*Malwana*' cultivar and using the microencapsulated peel extract in food product development.

Materials and Methods

Pretreatment screening for frozen peels

Rambutan (*N. lappaceum* L. cv Malwane) is the mostly grown and most popular cultivar in Sri Lanka. Discarded rambutan peels of *Malwana* were washed with tap water and divided into three portions of 500g each. One fruit peel portion was stored at -18 °C (S-1), another portion was first microwave blanched for 3 min and stored at -18 °C (S-2), the third portion was steam blanched for 3 min and stored at -18 °C (S-3).

Color extraction of frozen peels

The S-1, S-2, and S-3 samples (10 g) were thawed by keeping them at 25±2 °C for 1 hr. and blended for 2 min with distilled water (25 mL) in a laboratory blender. Then it was filtered through muslin cloth followed by Whatman no 1 filter paper (GE Healthcare, Life sciences, City Name, Germany). The filtrates or the crude extracts were visually compared for their color intensity.

Solvent screening for extraction of color

The S-1 peels were thawed as mentioned above, and the pigments were extracted by blending the thawed peel (10 g) with 25 mL each of distilled water, acetone, and ethanol (1:1:1) for 2 min in a laboratory blender. The blended samples were filtered, and the filtrates or the crude extracts were visually compared for color intensity. Both S-2 and S-3 samples were subjected for the same treatment. Anthocyanin contents of the crude extracts were determined as given below.

Screening for temperature to extract color

The S-1 peels were thawed as mentioned above. The pigments were extracted by mixing the thawed peel (10 g) with the distilled water (25 mL) and exposing the mixtures to three temperatures (25, 50, and 80 °C) for 1 hr in a shaking water bath (Gemyco, YCW-01C, CITY NAME, Korea), followed by blending for 2 min in a laboratory blender (RTX-023, COMPANY NAME< Phoenix, AZ, USA) and filtering through muslin cloth and Whatman no 1 filter papers (GE Healthcare, Life sciences, CITY NAME, Germany). The filtrates or the

crude extracts were visually compared for color intensity.

Final extraction of colorant through optimized conditions (above)

All the three S-1, S-2, and S-3 samples were tested for the color intensity, and finally got the S-1 was succeeded. Only S-1 sample was subjected for the further analysis due to resultant higher color intensity. The S-1 peels thawed as given above were extracted by mixing them (40 g) with distilled water (100 mL) and exposing the mixtures to 50 °C for 1 hr in a shaking waterbath, followed by blending for 2 min in a laboratory blender. Then it was filtered through muslin cloths and Whatman no 1 filter papers. Anthocyanin and total phenolic contents and antioxidant capacity of the filtrate (crude extract) were determined as given below (AOAC, 2000). The crude extract was spray-dried after mixing with maltodextrin as given below.

Spray drying of the crude extract of colorant

Maltodextrin (Daejung, Kosaq Company, City name, Korea) was used as the carrier agent in the preparation of an emulsion for spray drying (Idham *et al*., 2012). The crude extract (200 mL) was mixed with maltodextrin (20 g), and the mixture was thoroughly mixed using stirring hotplate (MSH 20 D, Dathan Scientific Company Ltd., City name, Korea)) at 25±2 °C.

Afterwards, the mixture was kept at 25±2 °C for 6 hr. and filtered through muslin cloth. The filtrate (200 mL) was spray-dried in a spray dryer (L-8, Ohkawara Kakohki Co. Ltd., CITY NAME, Japan) at the inlet temperature of 120 °C and the outlet temperature of 60 °C to obtain the microencapsulated spray dried powder (MESP), which was collected in the cyclone. Anthocyanin, total phenolic contents, antioxidant capacity, solubility, color, moisture content, and water activity of MESP were determined using the methodologies given below.

Parameters of physicochemical analysis

Determination of anthocyanin content

Anthocyanin content was determined according to a spectrometric method with slight modifications (Boyano-Orozco *et al*., 2020). The crude extract sample (4 ml) was diluted by a factor of 10 with distilled water, the absorbance of the solution was measured at 525 nm using a spectrophotometer (Jenway, 6300, Barloworld Scientific, CITY NAME, England). Total anthocyanin content was calculated as follows. Blank samples were prepared using distilled water, ethanol, and acetone. MESP (0.4 g) was dissolved in distilled water.

$$\text{Total anthocyanin content (g/kg)} = \left[\frac{(A525 \times 10 \times V \times DF)}{1000 \times V1} \right]$$

where;

$A525$ = absorbance

V = Total Volume (mL)

$V1$ = Collected sample volume

DF = Dilution Factor

Determination of total phenolic compounds content

The contents of total phenolic compounds of the crude extract and MESP were determined according to the Folin-Ciocalteu method (Asimi *et al*., 2013).

Determination of total antioxidant activity

Total antioxidant activity was determined according to the method of Baliyan *et al*. (2022) with some modifications. MESP (1 g) was dissolved in 12 ml of methanol. 2,2-Dihenyl-1-Pyicryl Hydrazyl (DPPH) working solution (3 mL) was mixed with 0.1 ml of the sample and kept in a dark place for 1 hr. Same procedure was followed for rambutan peel extract. Absorbance was measured at 517 nm using a spectrophotometer (Jenway 6300, Barloworld Scientific, CITY NAME, England).

$$\text{\% of Total antioxidant activity} = \left[\frac{(Ac\text{-}As)}{Ac}\right] \times 100\%$$

Where;

Ac = Control reaction absorbance

As = Testing specimen absorbance

Determination of solubility

Solubility of MESP was determined according to the method of Osorio-revilla (2020). MESP (1 g) was dissolved in 25 mL of distilled water at 25 °C and shaken for 5 min in a vortex mixer (JEIO tech, J-02, CITY NAME, Germany). The mixture was centrifuged at 3400 rpm for 5 min (CT4D) and filtered through Whatman no. 1 filter papers. The filtrate (25 mL) was dried for 5 hr at 105 °C and weighed.

$$\text{Solubility (\%)} = \frac{\text{weight of the dried solid}}{\text{Initial weight of powder}} \times 100\%$$

Where;

w= Initial weight of powder

w= weight of the dried solid

Determination of color

Color of the extracts were read according to the RHS color chart (6th edition Royal Horticultural Society, RHS Media, London, England). The color of MESP was determined using a colorimeter (Hanna, CS-10, CITY NAME, Thailand).

Determination of moisture content

Moisture content was calculated by drying MESP (1 g) in an oven (OF-22, JEIO Tech. Co., CITY NAME, Germany) at 110 °C until a constant (app. 0.01 difference) weight was obtained.

$$\text{Moisture content (\%)} = \frac{\text{Initial weight} - \text{final weight}}{\text{Initial weight}} \times 100\%$$

Determination of water activity

Water activity was determined by using a smart water activity meter (MRC, WAM -1, MRC Ltd., CITY NAME, Israeal). Following the placement of 10 g of MESP in the dish, the machine was started, and the resulting data were recorded.

Product formulations with MESP

Pasta: Wheat flour (120 g), MESP (3 g), and semolina (20 g) were mixed well with water (15 mL) and ½ of an egg to produce pasta. The mixture was extruded in a pasta extruder (Dolly-Mini P3, Dolly (Pvt)

Ltd, CITY NAME, Denmark), followed by steam-blanching for 15 min and drying in a dehydrator (SL-1N, Olympia Pvt., Ltd., CITY NAME, Japan) for 1 hr. The dehydrated and the boiled pasta samples were visually observed.

Gelly: 20 g of Gelatin (Delmage, Motha Confectionary Works Pvt. Ltd., CITY NAME, Sri Lanka) was mixed with water (100 mL), followed by mixing further with hot water (300 mL), sugar (100 g), a flavoring, and MESP (1 g) dissolved in water (10 mL) and then cooled. The resultant product was compared with the market sample to identify any remarkable difference between them.

Statistical analysis

The data were statistically analyzed using SAS software (SAS Enterprise Guide versions, SAS Institute INC., Cary, NC, USA) followed by one-way analysis of variance (ANOVA). The treatments were compared by LSD mean comparison tests at p=0.05. The results were presented as mean values of triplicate with standard deviations (SD).

Results and discussion

As rambutan is a seasonal fruit, it is important to identify an easy method for the preservation of the peels in bulk. Therefore, the effect of microwave blanching and steam blanching as treatments before freezing of the peel on the color intensity of the water extracts was studied. The highest color intensity in the samples frozen without blanching, the lowest color intensity in the microwave blanched samples and moderate color intensity in the steam blanched samples were evident (Figure 1). Difference in color intensity revealed that blanching before freezing was not suitable as a pretreatment probably due to thermal instability of anthocyanins in the extract. Similarly, thermal instability of anthocyanins (Sun *et al*., 2011) due to presence of conjugated double bonds in their structures was reported (Patras *et al*., 2010). Moreover, type of anthocyanins was reported to affect the thermal stability of anthocyanins as revealed by cyanidin-3-rutinoside showing the highest stability at 95 °C (Rubinskiene *et al*., 2005). A number of extraction methods of anthocyanins has evolved from traditional solvent extraction methods to many other emerging technologies such as deep eutectic solvent extraction, ultrasonic assisted extraction, microwave-assisted extraction, and supercritical fluid extraction (Tena & Asuero, 2022; Tan *et al*., 2022).

Figure. 1. Effect of blanching on color intensity of the water extracts of frozen (-18 °C) rambutan peels (1- No blanching; 2- Steam blanching for 3 min; 3 microwave blanching for 3 min)

Where according to the RHS color chart reading for 1, 2 and 3 tubes;

1. Red Group 42 vivid reddish orange A
2. Orange – red 34 Group strong reddish orange C
3. Orange – red 33 Group vivid reddish orange B

However, the objective of this research was to find out a simple method of extraction of rambutan peel with minimum resources and cost. As anthocyanin is a polar compound, ethanol, acetone, and methanol are commonly used in traditional solvent extraction methods (Khoo *et al*., 2017). Therefore, effect of acetone, ethanol, and water on the color intensity of the peel extracts was studied. Visual observation of the color intensity of the extracts revealed the highest anthocyanin extraction with acetone, the second higher with ethanol and the lowest with distilled water (Figure 2). Moreover, anthocyanin contents (mg Kg^{-1}) in the peel extracted with acetone, ethanol, and distilled water are presented in Table 1. Although acetone resulted in the highest extraction of anthocyanin (Table 1), its use is not desirable, as it is not a food grade chemical. Moreover, removal of acetone is costly, and its complete removal needs additional resources. Although the extraction of anthocyanins with 55% ethanol at 60 °C for 1 hr is reported to be a suitable method, using ethanol as a solvent is also costly (Khoo *et al*., 2017). Therefore, distilled water was selected as the most economical solvent, regardless of the low extractability of anthocyanin.

Figure. 2. Effect of solvent on color intensity of the extracts of frozen (-18 °C) rambutan peels (1- Acetone; 2- Ethanol; 3 – Distilled water)

Where according to the RHS color chart reading for 1,2 and 3 tubes of;

1. Red Group 46 strong red A
2. Red Group 45 vivid Red A
3. Orange red 34 Group vivid reddish orange B

Table 1. Effect of solvent on anthocyanin contents of the extracts of frozen (-18 °C) rambutan peels

Solvent	Anthocyanin content (mg Kg^{-1} FW)
Acetone	13.04 ± 0.42a
Ethanol	8.30 ± 0.46b
Distilled water	2.94 ± 0.04c

Values are averages of triplicate. Results are expressed as the mean ± standard deviation. Mean values within a column with different superscripts are significantly different ($p<0.05$).

As the extractability is affected by extraction temperature, effect of extraction temperature (25, 50, and 80 °C) on the color intensity of the peel extracts was studied. The highest color intensity was evident in the samples extracted at 50 °C (Fig. 3). Therefore, extraction of the frozen peel with distilled water at 50 °C, while shaking in a shaking water bath was found to be suitable as reflected by the color intensity of the extract (Figure 3 and 4).

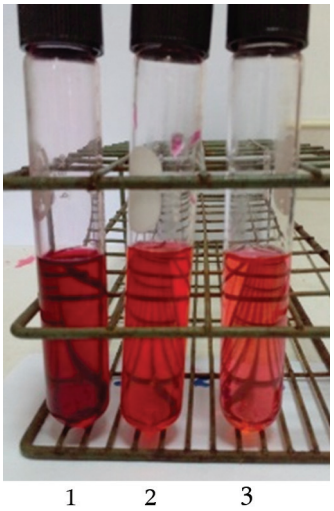

Figure. 3. Effect of extraction temperature (1: 50 °C; 2: 25 °C; 3: 80 °C) on color intensity of the extracts of frozen (-18 °C) rambutan peels

Where according to the RHS color chart reading for 1, 2 and 3 tubes;

1. Red Group N 45 Moderate Red C
2. Orange – red N 34 B strong reddish orange B
3. Orange – red N 34 Moderate Red C

Figure. 4. Color of the extract of frozen (at -18 °C) rambutan peels extracted with distilled water at 50 °C

According to the Table 2, the contents of total phenolic compounds from the water extract (50 °C) of the frozen peel was 24.52 ± 0.47 mg mL^{-1}, amounting to 2.5 % (g mL^{-1}) of the crude extract and 12.3 % (g g^{-1} FW) of the frozen peel. The antioxidant capacity of the water extract (50 °C) of the frozen peel was 80.06 ± 0.01%, when determined using the DPPH assay. Therefore, incorporation of water extract of frozen rambutan peel in product formulations can add value to finished products. Moreover, based on acute toxicity studies, as the ethanolic extract of rambutan peel is reported to be non-toxic (Rohman, 2017), discarded rambutan peel can be utilized as safe and health-beneficial ingredients without throwing them away. However, it is necessary to make the water extract of the frozen peel available in a convenient form for possible applications in food processing. Moreover, the degradation of anthocyanin and other bioactive needs to be minimized, when the extract is incorporated into food, as the stability of these compounds is reduced during processing and storage (Patras *et al*., 2010; Sun *et al.*, 2011).

Table 2. Phytochemicals of water extract at 50 °C from the frozen rambutan peels

Parameter	Water extract at 50 °C	% from crude extract	% from frozen peel
Total phenolic content	24.52 ± 0.47 mg mL-1	2.5	12.3
Antioxidant activity	80.06 ± 0.01 %		

Microencapsulation is reported to protect the sensitive constituents, thereby ensuring their safe delivery (Choudhury *et al*., 2021). Encapsulation efficiency of 99.7% after spray drying of emulsions of anthocyanin and maltodextrin is reported (Idham *et al*., 2012). Therefore, the crude extract encapsulated with maltodextrin was spray-dried to obtain MESP, which can easily be incorporated as an ingredient in product formulations. According to the Table 3, the yield of MESP was 16.5%. Anthocyanin content of 24.76 ± 0.04 mg Kg^{-1}, total phenolic compounds content of 373 ±7 g Kg^{-1} and antioxidant activity of 81% (DPPH assay) were evident in MESP.

Physical properties such as solubility and color of MESP are important parameters for a food processor to find out its suitability as an ingredient. Water solubility of a powder is a measurement of the degree to which it can be readily solubilized in water before use (Hardy & Jideani, 2018). Solubility of MESP, which is an important physical property that facilitates its incorporation as an ingredient in product formulations, was found to be 74.29 ± 0.04% at 25±2 °C (Table 3). Lower solubility, than that of spray dried milk powder (Sulieman *et al*., 2014), evident in this study is possibly adequate for using MESP as an ingredient. As spray-drying may increase the rates of browning reactions, and color of spray dried powders affects the color of finished products when incorporated them as ingredients, color of MESP was measured. The L^* (lightness) value of MESP, which indicates lightness and blackness, was 71.3 ± 2.25 (Table 3). If necessary, temperature of spray-drying and direction of feeding can be adjusted as reported by Sulieman *et al*. (2014) to make MESP lighter in color.

Moreover, moisture content and water activity of MESP are important parameters to decide a suitable packaging material and to predict the shelf life. Based on the moisture content of 7.80 ± 0.02% and water activity of 0.41 ± 0.04 evident in this study (Table 3), revealed that packaging in air, moisture and light barrier pouches made of tin, laminate (Al mixed triple layer), and so on are suitable to pack the MESP, soon after producing to keep successful storage life.

MESP was incorporated as an ingredient in formulations of pasta and jelly to find out the effectiveness of MESP, particularly when exposed to high temperatures of processing under dry and moist conditions, and also to different types of ingredient such as wheat flour and gelatin. Incorporation of MESP in the formulation of pasta imparted a reddish color to the finished products. No significant change in color of pasta during boiling was also evident, showing the ability of MESP to impart color in starch-based products processed at high temperature and re-constituted in boiling water. However,

coagulation and darkening of color were evident when MESP was incorporated in the formulation of jelly. Effect on such physical properties of jelly may be attributable to formation of hydrogen bonds between hydrophilic groups of gelatin with the $^{-}$OH groups of maltodextrins, anthocyanin molecules and other phenolic compounds that are considered relatively polar hydrophilic compounds, which were extracted when frozen rambutan peel was subjected to water extraction at 50 °C. A previous study revealed perfect miscibility of biopolymers such as gelatin and maltodextrin at low concentrations and precipitation with increase in their concentrations, resulting in a liquid coexisting with a solid phase (Beldengrün *et al*., 2018). Therefore, further studies are necessary to find out the effectiveness of MESP in gelatin based products. Moreover, further research is necessary to find out the stability of bioactive compounds during storage of MESP. However, the results of this study indicate the potential of extracting bioactive compounds from discarded rambutan peels and producing spray dried powder for adding value to different types of food.

Conclusion

The extraction methodologies employed for rambutan peels revealed that the highest anthocyanin extraction was achieved using acetone, followed by ethanol, and distilled water yielded the lowest results. Despites this, it is crucial to develop a water extract that can be conveniently utilized in food processing applications. To address this, the crude extract was encapsulated with maltodextrin and subsequently spray-dried to obtain a microencapsulated powder of rambutan peel extract. Apart from its phytochemical and physiochemical properties, the powder demonstrates positive characteristics such as solubility and stability at high temperatures, making it an ideal ingredient for various product formulations. The moisture content and water activity of the resulting powder are within the favorable ranges, indicating that it can be stored successfully under ambient conditions for an extended period when packaged appropriately. Consequently, there is significant potential in extracting bio-active compounds from discarded rambutan peels and producing spray dried powder, which can enhance the value of a wide range of food products.

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