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REGULAR ARTICLE

Stability Improvement of Betalains Recovered from Red Dragon Fruit Peels *(Hylocereus polyrhizus)* by Cellulose-Based Encapsulation

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Abstract
This study promoted a valorization pathway of Red Dragon Fruit Peel, including extraction of betalains and pectin, stabilization of batalains and expanding the application of betalains and pectin in jam and jellies. Betalains were extracted by the same weight of ethanol 96% at 45 ℃ for 1 h and obtained with the content of 2.09 ± 0.03 mg/g of dry peels. To minimize the solid wastes, the alcohol-insoluble residues of betalains extraction were utilized to extract pectin by citric acid 0.1 M at 85 °C for 120 min with 19.8% yield and 56.8% DE (degree of esterification). Betalains stabilization focused on the encapsulation in support of the freeze-drying technique and microcrystalline cellulose (MCC) as a wall material. Freeze-dried MCC/betalains complexes were prepared by different weight ratios of betalains and MCC (1:3, 1:5, and 1:10). Encapsulation of betalains promoted significantly higher stability at different storage conditions: cold (4 °C) and room temperature (27 °C) with daylight and without daylight. The stability of encapsulated betalains was improved at high temperatures (80 °C and 100 °C), various pH levels (1.2, 3.6, 5.6, and 7.4) and water activities (0.089 and 0.898) when compared with the nonencapsulated betalains. The incorporation of encapsulated betalains into pineapple jam and gummy candy demonstrated storage stability after a two-week storage period.

Keywords Red dragon fruit peel · Betalains encapsulation · Freeze drying · Microcrystalline cellulose · Biomass waste valorization

1 Introduction

Fruit waste or fruit processing by-products is a growing problem in our modern society. The long-term disposal of these fruit wastes into the environment leads to global issues. Developing novel processes to convert these byproducts into value-added products could provide a viable way to manage this waste problem while simultaneously creating sustainable economic growth from a bioeconomy perspective. In some tropical countries, red dragon fruits (*Hylocereus polyrhizus*) are of high potential as a profitable fruit crop, whose peels contribute 22–44% of the fruit weight and are commonly disposed of as wastes during processing [1]. However, red dragon fruit peel wastes are rich sources of betalains, a natural pigment, and pectin, a commonly used food additive, promising to create value-added products for the future food industry.

Natural pigments in food have been produced as an alternative to replace synthetic dyes, as these artificial dyes were reported to affect health negatively [2]. The research groups working on betalains highlighted their unique chemical properties, such as water solubility and color intensity, that offer them used as a vivid red food colorant. In addition, many studies demonstrated the antioxidant, antibacterial, and anticancer properties of betalains [3]. However, despite exhibiting interesting properties under food and biological aspects, one of the main constraints that prevent their potential use has been their instability in the presence of high temperature, pH < 3 or > 7, light, oxygen, and high water activity [4]. Therefore, the objective of studies in recent years has been based on finding ways to stabilize them to

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increase their commercial applications and take advantage of their benefits.

Encapsulation represents a promising method for the stabilization of betalains [4]. Several groups have reported that three main factors, including the type of matrix, the encapsulation technology, and the matrix's porosity, contribute to improving betalains stability in an encapsulated form [4]. The matrix selection is critical since it depends on the final application of the encapsulated compound and an efficient result. Several matrices have been employed to encapsulate betalains as polysaccharides, proteins, or in combination with polysaccharides [4, 5]. Maltodextrin is most used as a matrix to encapsulate betalains [4, 5]. Several other encapsulating agents, such as guar gum, gum Arabic, xanthan gum, and pectin have been used alone and in combination to upgrade betalains encapsulation, generating more solid knowledge about the influence of coating materials and involved processes on encapsulation and the desired characteristics of the final products [4]. Microcrystalline cellulose (MCC) has more natural attributes relevant to food applications, including the degree of crystallinity, degree of polymerization and molecular weight, particle size and shape, mechanical properties, thermal stability, porous structure, and surface area and moisture content characteristics, which can be a novel candidate replaceable for maltodextrin in betalains encapsulation [6]. De Barros et al. reported the improved survival of probiotic cells during extrusion/spheronization and excellent protection from gastric acid when MCC calcium crosslinked alginate was fabricated as wall material [7]. The researchers discussed that the combination of MCC and sodium alginate produced efficient sphere disintegration and rapid release of probiotics in intestinal conditions. Owing to their barrier effects, MCC could produce coats to protect the sensitive bioactive food components against moisture, oxygen, light, and temperature [8]. Koupantsis et al. reported good shell formation with good protective ability of essential oils when they investigated the complex coacervation of milk proteins using carboxymethylcellulose (CMC) and MCC as the wall materials [9]. No studies have used MCC as wall material in betalains coating to the best of our knowledge.

Several encapsulation techniques have been used to produce microparticles, such as spray drying, freeze drying, ionic gelation, emulsions, etc. [4, 5]. The most used technology for the encapsulation of betalains is spray drying due to its low cost, simple processing, and its rapid obtaining of encapsulated betalains. However, the main disadvantage of spray drying is the use of high temperatures to evaporate the soluble solution of matrix and solvents quickly, and betalains may be degraded [4]. Some previous works demonstrated that the freeze-drying technique might be a good way to tackle the limitations of spray drying in coating betalains [10]. Few studies have addressed the role of cellulose-based carrier agents and freeze-drying technique in the encapsulation of bioactive compounds. Vukoja et al. observed that the amount of cellulose used as a carrier polymer for volatiles and complexation time influenced the adsorption efficiency [11]. Then, these authors found that cellulose could be a good encapsulation polymer for delivering raspberry bioactive compounds, especially when cellulose was used in lower percentages to formulate encapsulates [12]. Siccama et al. found that partial replacement of maltodextrin by cellulose-based carriers resulted in freeze-dried powders with similar physical properties as the control sample and did not detrimentally influence the aroma compounds [13]. Feng et al. demonstrated that CMC-Na and MCC could encapsulate astaxanthin and increase the astaxanthin solubility, stability, and antioxidation activity [14]. Their expanding application in yogurt showed that CMC-Na and MCC microencapsulated astaxanthin could enhance yogurt's stability and antioxidation activity as a food thickener, stabilizer, and coloring additive.

Our group has worked on building a valorization strategy for biomass waste to produce add-valued products [15, 16]. We developed a pathway to entirely convert the red dragon fruit peels, a waste product of fruit processing, into useful additives in the food industry [16]. As inherited from the results of this study, betalains and pectin were extracted in turn by a simple procedure with competitive efficiency. To increase betalains' commercialized value and applicability, MCC is investigated as a wall material of freeze-dried encapsulation to improve betalains stability for commercial purposes. The freeze-dried betalains and pectin obtained from red dragon fruit peel were applied in producing jam and jellies, which has not been reported previously to our best knowledge.

2 Materials and Methods

2.1 Betalains and Pectin Extraction from Red Dragon Fruit Peels

2.1.1 Extraction Methods

Fresh Red Dragon Fruit peels were collected from HG Food Joint Stock Company, Long An Province, Viet Nam. The green parts of the fresh peel were removed to obtain the red one and then blanched in hot water (90 °C) for 1 min to deactivate pectic enzymes. The peels were cut into 2–3 cm pieces and were stored at − 4 °C for further experiments.

Betalains and Pectin Extraction from Red Dragon Fruit Peel were modified from our previous work [16]. First, red dragon fruit peels were blended in a homogenizer with the same weight of 96% ethanol to obtain a pulp. The pulp extraction process was then carried out in a thermostat water bath,

maintaining the temperature at 45 °C for 60 min. The mixture was filtered and pressed to obtain a colored solution and solid waste. The colored solution was then vacuum filtered through a 45 µm filter paper. The filtrate was concentrated by a rotary evaporator (Yamato RE 301A-W) at 35 °C for 30 min to remove solvents. The concentrated extract was stored at 4 °C for further use.

The solid waste of the filter press process was dried at 60 °C in an oven (UM500, Memmert GmbH, Schwabach, Germany) to obtain a dry matter with 17.2% moisture content and utilized for pectin extraction. First, the extraction process of Pectin was carried out using acidified water (0.1 M citric acid) at 85 °C for 120 min in a thermostat water bath. The weight ratio of solid and acidified water was 1:40 (w/v). The hot acid extract was then filtered using a cloth to remove the pulp. The filtrate was then cooled to 4 °C and precipitated using a double volume of 95% ethanol. Finally, the precipitate was filtered and washed with the solution of 70% ethanol via vacuum filtration. The resulted pectin was dried in the oven at 65 °C until a constant weight was reached. The residual pulp was kept at 4 °C for other experiments.

2.1.2 Quantification of Betalains and Color parameters

UV–Vis spectroscopy is the most widely used analytical technique for quantitatively identifying two structural groups of betalains extracts (betacyanins and betaxanthins) [17]. The maximum absorbance of betacyanins and betaxanthins was measured at 537 ± 3 nm and 480 ± 3 nm, respectively, using a UV spectrophotometer (Biochrom Libra S22), according to the method of Stintzing et al. [18]. The total pigment content was calculated by Eq. (1):

$$\begin{aligned}&\text{Total pigment content (mg/L)}\\&= \text{betacyanin content (mg/L)} + \text{betaxanthin content (mg/L)}\\&= \frac{A_{537} \bullet M_{w(betacyanin)} \bullet D_F \bullet 1000}{E_{betacyanin} \bullet l} + \frac{A_{480} \bullet M_{w(betaxanthin)} \bullet D_F \bullet 1000}{E_{betaxanthin} \bullet l}\end{aligned} \quad (1)$$

where A: absorbance (betacyanin at 537 nm and betaxanthin at 480 nm), M_w: the molecular weight of betalains (308 g/mol for betaxanthin, 550 g/mol for betanin), D_F: dilution factor, E: molar extinction coefficient of betacyanin (48,000 L/mol·cm for betaxanthin, 60,000 L/mol·cm for betanin in H_2O), l: path length (1 cm).

2.1.3 Yield of Pectin Extraction and Structural Analysis of Pectin

The dried pectin powder was weighed for yield calculation. Then, the mass yield of pectin was calculated according to Eq. (2):

$$Y_p(\%) = \frac{M_p}{M_r} \times 100 \quad (2)$$

where Y_p is the pectin extraction efficiency based on the weight of solid residue, M_r is the weight of dried solid residue, and M_p is the weight of dry pectin.

The structure characteristic of the pectin samples was evaluated using Fourier-transform infrared spectroscopy (Frontier FT-IR/NIR) with wavelengths in the range 4000–400 cm^{-1}, measuring the resolution of 4 cm^{-1} [19] with a total of 20 scans. The spectra were then compared with that of the commercial Pectin (Pectin Classic CS 502, Corporate Group Herbstreith & Fox).

The degree of esterification (DE) of pectin of dragon fruit peel was determined by the titrimetric method of Food Chemical Codex [19]. The data were then compared with commercial pectin.

2.2 Encapsulation of Betalains Using a Freeze-Drying Technique

2.2.1 Preparation of Encapsulated Betalains

Betalains were encapsulated based on previous works with modifications [11, 20, 21]. Microcrystalline cellulose (Ambicel) was supplied by Maple Biotech Pvt. Ltd., India. First, MCC was dissolved in distilled water to form a 10% solution of wall material (w/v). Next, Betalains extract was mixed with wall material solution in core-to-shell ratios of 1:3, 1:5, and 1:10 (w/w) and then sonicated by an ultrasound sonicator (200 W) in 5 min. For the Betalain-to-wall ratio of 1:10, acetic acid was added into the complex (1% w/w) to increase the solubility of MCC. Finally, the sonicated solution was pre-frozen at − 60 °C and then dried with a vacuum freeze dryer (Toption TPV-50F Freeze Dryer) under the following conditions: the temperature of sublimation from − 50 °C to 0 °C; the temperature of isothermal desorption varied from 0 °C to 40 °C. The complete process lasted for 48 h. MCC-encapsulated BE samples were set names based on the weight ratio of Betalains and MCC. The ratio of 1:3, 1:5, and 1:10 was called "BE:MCC = 1:3", "BE:MCC = 1:5", and "BE:MCC = 1:10", respectively.

2.2.2 Encapsulation Efficiency

The method to determine the encapsulation efficiency was modified from the work of Feng et al. [14]. Encapsulated efficiency (EE) was calculated on the basis of the total quantity of betalains in the microencapsulated betalains and nonencapsulated betalains present on the surface. First, 50 mg of powder were washed twice in a 5 mL acetone solution and then centrifuged at 4000 rpm for 10 min. The Betalain content of the supernatant was calculated by Eq. (1). Total betalains in 50 mg of powder were extracted entirely by crushing and mixing the microparticles in 0.5 mL acidic water at pH 5 (using acetic acid to adjust pH) and 1 mL acetone, then vortexed thoroughly, followed by centrifugation to release the betalains completely into the ethanol solution. This was followed by a decantation and the bottom layer was re-extracted in 0.5 mL acidic water and 1 mL acetone. All supernatants were collected to measure total betalain content, and then calculated by Eq. (1). Encapsulation yields (EE) were estimated as Eq. (3).

$$EE(\%) = \frac{C_o - C_1}{C_o} \times 100 \tag{3}$$

where C_o: total betalains content of 50 mg powder (mg/L); C_1: nonencapsulated betalains present on the surface (mg/L).

2.2.3 Morphology of Betalains Encapsulates

Scanning electron microscopy (SEM) evaluated the particle morphology of the freeze-dried betalains powders. Particles were loaded onto a specimen stub, coated with gold layer by a sputter coater. Examinations were made at a high voltage of 5 kV with magnification = 3000 × [15].

2.2.4 Total Phenolic Content

Phenolic compounds in encapsulated samples were extracted by the procedure of Feng et al. with some modifications as described in Sect. 2.2.2 [14]. The total phenolic content (TPC) of encapsulated and noncapsulated samples was determined by Folin – Ciocalteu's method [22]. An aliquot of betalains was added to Folin – Ciocalteu's reagent (10%), which was diluted in distilled water and Na_2CO_3 7.5% w/v. The mixture solution was vortexed, covered with parafilm and left for 30 min. TPC was calculated by the absorbance at a wavelength of 765 nm using a spectrophotometer. The standard curve was prepared using solutions of GA. The total phenolic content was expressed in GA equivalents (mg GA/100 g fresh weight).

2.3 Evaluation of the Stability of Betalains

The effects of temperature, pH, water activity, and storage conditions on the stability of encapsulated betalains and betalains extract were observed. The procedure was similar to the work of Rodriguez et al. [23]. All experiments were performed in triplicate, and the data were reported as mean ± standard deviation. The stability of betalains was calculated according to Eq. (4):

$$Betalains\ retention(\%) = \frac{C_i}{C_o} \times 10 \tag{4}$$

where C_o represents the concentration of betalains at initial (mg/L); C_i represents the concentration of betalains at specific time (mg/L).

2.3.1 pH Stability

The nonencapsulated and encapsulated betalains were added to different buffers at different pH: 0.1 M HCl and 0.2% NaCl buffer solution (pH 1.2) and citrate–phosphate buffer solutions (pH 3–7). Betalains retention was monitored by determining the absorbance of the solution by UV spectrophotometer at 537 nm for 3 h.

2.3.2 Heat Stability

Aqueous solutions of encapsulated and noncapsulated betalains (0.1% w/w) were heated at 80 and 100 °C. The solution was immediately cooled to stop thermal degradation, and the betalains content was calculated at time intervals.

2.3.3 Effect of Water Activity

Nonencapsulated and encapsulated betalains were stored in desiccators at two different water activities (A_w). One was attained using saturated NaOH solution with $A_w = 0.089$, and another was a saturated $BaCl_2$ solution with $A_w = 0.898$. Betalains retention was monitored at 537 nm over 7 days.

2.3.4 Storage Stability

Noncapsulated and encapsulated betalains were kept at four storage conditions: cool and dark (black ziplock bags, amber bottles for BE and kept at 4 °C refrigerator); room temperature, dry and dark (black ziplock bags for powder, amber bottles for BE extract and kept in a desiccator), room temperature, dry and daylight (transparent ziplock bags, transparent bottle for BE extract and kept in a desiccator); atmosphere and daylight (transparent ziplock bags of powders and

a transparent bottle of BE extract were put at free space in the laboratory with 80% relative humidity). The total betalains content was monitored for 30 days.

2.4 Application of Pectin and Encapsulated Betalains in Food Systems

Pineapple jam and gummy candies were prepared to mimic real food models but with very simple compositions. Pineapple was purchased from local market.

One fresh pineapple has removed the top and sliced off the peel. The fruit was quartered vertically, and the core was removed. Half of the pineapple was placed in a food processor and pulsed until it was fine but not completely pureed. Next, the crushed pineapple was cooked in a jam pot to boil (100 °C), and 2.5 g of Red dragon fruit pectin was mixed with puree. Citric acid was added to adjust pH 3.5. The mixture was stirred to incorporate in 5 min. Then, 50 g of sugar was added, mixed well, and kept heating at 80–100 °C for 2 min. Next, 0.05 g of MCC coated BE powder was mixed with jam to make it red. Finally, the colored jam was cooled down and poured into sterilized jars.

To prepare gummy candy, 8 g of red dragon fruit pectin and 12 g of commercial gelatin (Ewald-Gelatine GmbH, Germany) were mixed with 30 g of water at 80 °C until complete dissolution. Citric acid was added to adjust pH 5.0. The mixture was stirred to incorporate and kept gentle heating (80–100 °C) for 5 min. 50 g of sugar was added, mixed well, and kept gentle heating lower to 80 °C for 2 min. 0.1 g of MCC coated BE powder was mixed with the mixture before being poured into multishape silicon templates and cooled at room temperature to form the gels.

The model jam and gummies were stored at room temperature in vacuum-sealed plastic bags and at 4 °C in fridge. Subsequently, the shelf-life stability of food samples was monitored over 2 weeks. Samples were pureed, weighed, and dissolved in distilled water (1:2 w/v). The solution was filtered, centrifuged and the clear liquid was analyzed for betalains content. In addition, microbiological assays of two food systems were conducted after 2-week storage to identify and restrict harmful microorganisms, which can spoil foods and ensure safety (as seen in Table 1).

3 Results and Discussion

3.1 Betalains and Pectin Extraction from Red Dragon Fruit Peel

The extraction of betalains is the first step for valorizing red dragon fruit peel; therefore, it is crucial in the following process. Betalains in red dragon fruit peel are a group of water-soluble pigments consisting of betacyanin and betaxanthin. The maximum absorbance of betacyanin was detected at 537 ± 3 nm, but no betaxanthin was detected at 480 ± 3 nm. When compared with other conventional procedures, the content of betacyanin in this study was 208.8 mg/100 g of dry matter, which is higher than that of 13.8 mg/100 g extracted by Wu et al. and 18.67 mg/100 g extracted by Ramli et al. [29, 30]. The total phenolic content of BE extract was 307.9 mg of GAE/100 g dry matter. These values are different from some previous works due to the discrepancy in the growing environment, varieties, and extraction method of the red dragon fruit [1]. As the consumption of fresh dragon fruit is preferred, most of the processed dragon fruits are of low-quality varieties. In addition, fresh residual peels from processing were not purposely stored, resulting in the loss of beneficial biology compounds. Some authors mentioned that conventional extraction has some limitations in preserving bioactive compounds [4]. However, it cannot deny the advantages of the method used in this study as it demonstrated a competitive yield and short extraction time, in addition to simple equipment, eco-friendly solvent, and most notably, it made the full use of solid residues to produce pectin.

In this study, the solid residue after the filtration step of the extraction was utilized to obtain pectin. Pectin was recovered in 19.8% yield, and DE was 56.8%. The red dragon fruit peel pectin is classified as high-methoxyl pectin (DE > 50%) at such DE value. The pectin extracted by a similar condition reported by Muhammad et al. (2014) also showed a high DE value with 63.74% [31]. The structure of Pectin obtained was determined using Fourier-transform infrared spectroscopy (FT-IR), which is consistent with commercial pectin (Fig. 1). A broader absorption band around 3400 cm^{-1} was attributed to the stretching of O – H group, and an absorption band at 2900 cm^{-1} was due to the C – H stretching of CH2 group.

Table 1 Methods for Microbiological assays (was tested by Chan Nam TSS—Chan Nam Science Technology Service Joint Stock Company)

Criteria	Unit	Methods
Total aerobic microorganisms	CFU/g	TCVN 4884-1:2015 (ISO 4833-1:2013) [24]
Escherichia coli	CFU/g	TCVN 7924-3:2017 (ISO 16649-2:2001) [25]
Staphylococcus aureus	CFU/g	TCVN 4830-1:2005 (ISO 6888-1:1999 and 1:2003) [26]
Salmonella app	/25 g	TCVN 10780-1:2017 (ISO 6479-1:2017) [27]
Yeast and mold	CFU/g	AOAC 2014.05 [28]

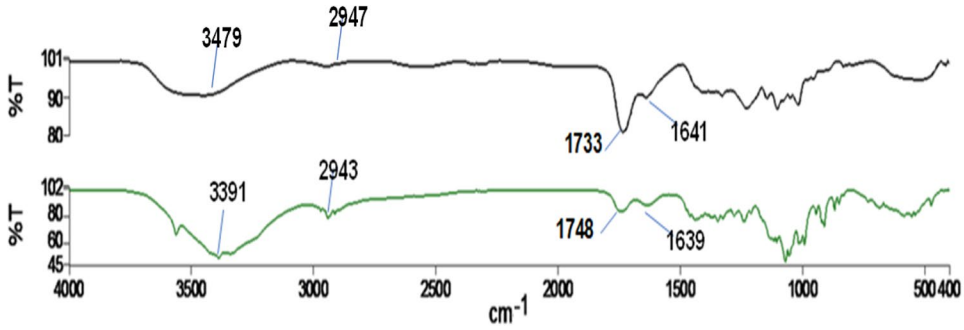


Fig. 1 FT-IR spectra of Red dragon fruit peel Pectin (black) and commercial Pectin (green) (colour figure online)

Table 2 FTIR spectra of dragon fruit peel's pectin and commercial pectin

Functional Groups	FTIR wavenumber (cm^{-1})	
	Red dragon fruit pectin	Commercial pectin
OH group	3479	3391
CH group	2947	2943
C–O from esterified carboxyl group	1733	1749
Free carboxyl group	1641	1639

The absorption bands in the wavelengths 800–1200 cm^{-1} are the fingerprint regions of the typical pectin polymers. The absorption bands at 1630–1650 cm^{-1} and 1730–1760 cm^{-1} indicated the free and esterified carboxyl groups, respectively. This structural characteristic was also consistent with previous works [31, 32]. The comparison between the typical peaks of the extracted and commercial pectin is shown in Table 2. Pectin was used to make jams and jellies.

3.2 Encapsulation of Betalains Using Freeze-Drying Technique

Drying of betalains extract is to obtain a pure and easy-to-use pigment in powder form, and freeze drying was chosen for betalains encapsulation due to its favorable mild process parameters (low temperature) to avoid degradation of bioactive compounds [4]. MCC was used as coat in encapsulation to provide protection for the sensitive bioactive food components against moisture, oxygen, light, and temperature [9]. Using MCC as a wall material with different ratios of core and wall showed high encapsulation yields of 96–98%, which indicates that MCC can hold betalains inside the particles through hydrogen bonding between the carboxyl group of betalains and polyhydroxy of MCC (Fig. 2). Although MCC has a Higher Degree of Polymerization (DP > 500), making it slightly insoluble in water as compared to Maltodextrin [14], employing a mild ultrasound treatment (200 W, 5 min) helped improve the dispersion of MCC in water and promote the interaction between MCC and Betacyanin, resulting in excellent EEs. Similarly, Li et al. demonstrated that ultrasound treatment at 200 W for 5 min resulted in the best EE due to interactions between Maltodextrin and Betalains [20].

Table 3 indicates the total phenolic content and betacyanin content of freeze-dried powders. Betalains extract was freeze-dried and analyzed as a reference sample to compare with the encapsulated samples. As seen in Table 3, the total phenolics and color contents did not change much after freeze-dried, and the content values are similar among the powders. The powders' total phenolic and betacyanin contents were slightly lower than the BE extract. These declines corresponded to adding a solid amount of MCC.

SEM micrographs revealed the morphology of betalains microcapsules (Fig. 3). From Fig. 3, it can be seen that the betalains microcapsules were irregular rigid, similar to freeze-drying encapsulation in previous studies. The morphology of the particles was uneven and not spherical. Most microencapsulated particles are less than

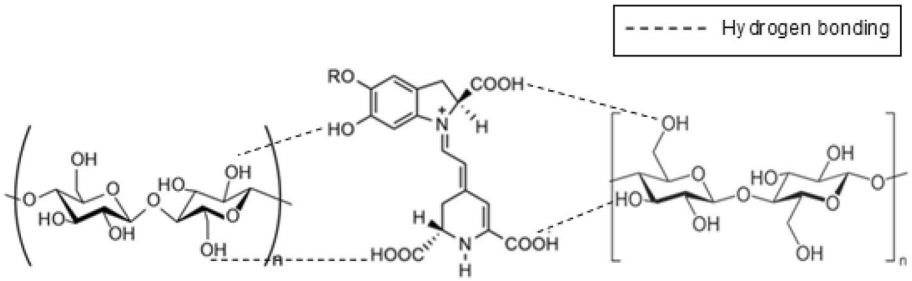


Fig. 2 Hydrogen bonding between Betacyanin and MCC (similar to the hydrogen bonding between betacyanin and maltodextrin reported by Rodriguez et al. [22])

Table 3 Characteristics of MCC encapsulated betalains

	Freeze-dried BE extract	MCC encapsulated BE*		
		BE:MCC = 1:3	BE:MCC = 1:5	BE:MCC = 1:10
Encapsulation yield (%)	–	96.37 ± 1,92	97.80 ± 1.10	97.02 ± 0.28
Belatains content (mg/100 g dm)	150 ± 1.32	124 ± 0.76	129 ± 0.20	145 ± 0.55
Total phenolics content (mg GAE/100 g dm)	284 ± 1.99	235 ± 1.05	224 ± 0.33	269 ± 0.07

*MCC encapsulated BE samples were set name based on the weight ratio of Betalains and MCC, the ratio of 1:3, 1:5 and:10 was called "BE:MCC = 1:3", "BE:MCC = 1:5", and "BE:MCC = 1:10", respectively

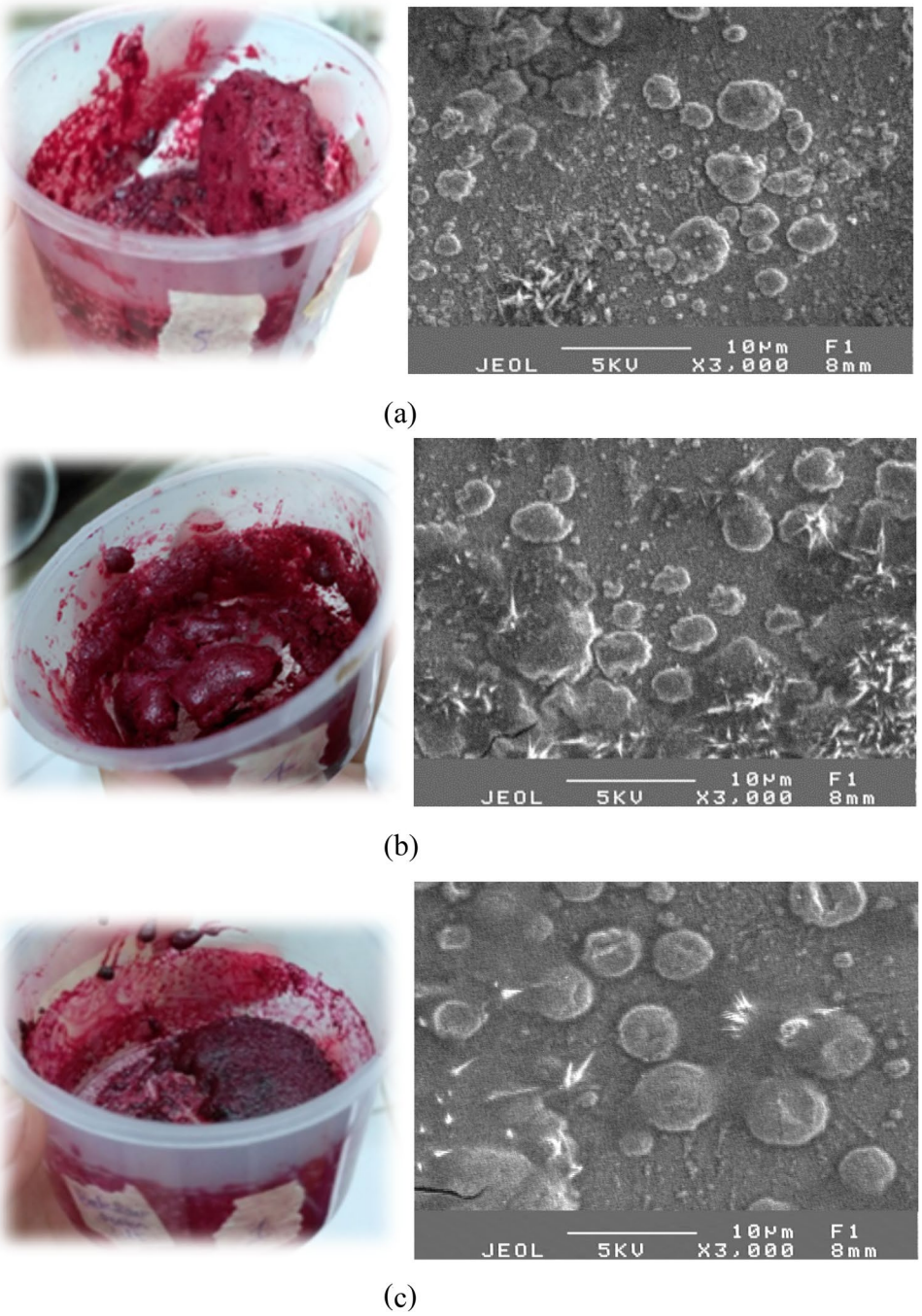


Fig. 3 SEM images of MCC encapsulated BE with a core-to-wall ratio of (**a**) 1:3; **b** 1:5; and **c** 1:10

10 μm in size. The difference in particle size may be associated with the concentration of wall material and the crushing of freeze-dried powders after the freeze-drying process. Microencapsulated particles in samples with high MCC concentration (ratio of BE:MCC = 1:10) tended to stick together. Romero-González et al. [33] reported that the morphology of the freeze-dried microcapsules was not spherical like spray-dried ones but irregularly disrupted porous structure. The gum Arabic-microencapsulated pigment has a continuous compacted structure with a very fine surface, while the inulin-coated pigment has a rough surface but dense inside.

3.3 Thermal Stability of Encapsulated Betalains

Heat treatment such as cooking, canning, or pasteurization remains the most significant problem that needs to be addressed in using betalains in food. In this study, the thermal stability of freeze-dried powders was investigated at 80 and 100 °C, which was a typical range of food processing. As in Fig. 4a, at 80 °C, BE extract was degraded entirely only after 60 min, while the three freeze-dried powders remained 30–40% of betalains content. The BE protection by the MCC coating from physiochemical factors in the aqueous solutions slowed the degradation rate of BE. The thicker the coat, the more BE remains. Apparently, after 90 min, the BE encapsulated by the thickest MCC wall (with a 1:10 core-to-wall ratio) still has 20% remaining, while the thinner walls are entirely decomposed. A much loss of color begins to occur even below the pasteurization conditions of 70–80 °C [4].

An increase in the temperature always accelerates betalains degradation (Fig. 4b). At 100 °C, BE encapsulated by MCC with the 1:3 core-to-wall ratio disappeared after 30-min heating. BE protected by the thickest MCC wall indicated the best thermal stability, retaining over 20% of BE after 45 min, but the one with a 1:5 core-to-wall ratio was degraded completely. The loss of BE extracts at 100 °C in this study was consistent with de Mello et al. [34]. This thermal instability of BE in aqueous solutions was explained that prolonged heating in the presence of oxygen and light could dramatically lead to the degradation and structural modification of betalains via a series of reactions, such as hydrolysis, isomerization, dehydrogenation, deglycosylation, and decarboxylation [35]. Although the degradation profiles of the cellulose-based betalains delivery system are slightly different from the maltodextrin-based system in previous works, the MCC-based one with the core-to-wall weight ratio of 1:10 indicated an improvement in heat properties of BE as compared to nonencapsulated BE.

3.4 Effect of Water Activity on the Stability of Encapsulated Betalains

Water activity is a factor that must be controlled in products containing betalains. This factor affects the rate of water-dependent hydrolytic reactions for aldimine bond cleavage of betalains [35]. As seen in Fig. 5, BE contents of the extract and powders were gradually decreased at two levels of water activity after 7 days. High water activity has also been reported to potentiate betalains degradation, while low water activity can improve stability [36]. Similarly, the degradation rates of encapsulated and nonencapsulated at $a_w = 0.089$ were slower than those at $a_w = 0.898$. Notably, the decline in BE contents of freeze-dried powders was just under 6% after a week at $a_w = 0.089$, but the BE contents of these powders were decreased by over 20% at a higher water activity of 0.898. The previous work also demonstrated that the water activity below 0.63 is the most effective in betalains stabilization [37].

3.5 Effect of pH on the Stability of Encapsulated Betalains

Similar to previous works, Fig. 6 showed that the encapsulated and nonencapsulated Betalains were stable in pH between 3 and 7. Most of them remained over 88% of the pigment content at that pH range, and the best results were shown at pH 5.3. At pH 1.2, the powder with the highest MCC content (the core-to-wall ratio of 1:10) did not protect BE well from degradation. Previously, Yousefi et al. also discussed that a high MCC concentration detrimentally affected the encapsulation of bioactive compounds in spray-dried black raspberry juice due to the structural changes of the powder particles [38]. This detrimental effect might be more intense at low pH. In contrast, the other two with thinner wall layers remained over 80% of Betalains content after 3 h at pH 1.2, which can be a stable colorant in food applications to pH 3.6–7.4 as they might protect betalains and other bioactive compounds from the stomach's acidic environment (pH 1.2) from being released and then, appropriately absorbed in the intestines.

3.6 Storage Stability of Encapsulated Betalains

The effects of four different storage conditions on betalains and their freeze-dried powders are shown in Fig. 7. In four storage conditions, three MCC-encapsulated powders indicated an improvement in the stability of betalains. In general, betalains retentions of all powders were excellent when they were stored in the fridge and desiccator. All samples exposed to the atmosphere indicated a considerable decrease in betalains content after one month. These results were consistent with previous works mentioned in a review

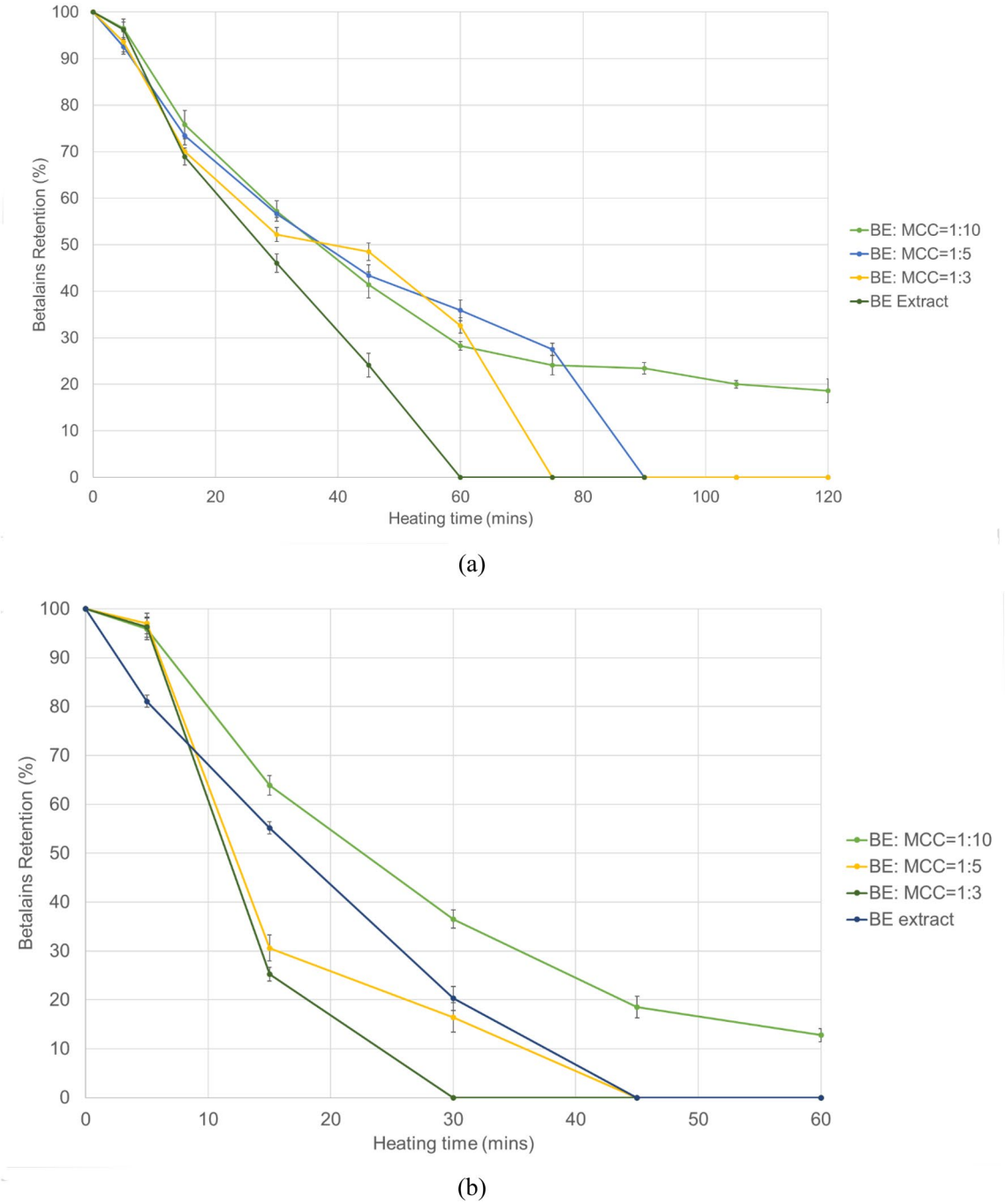
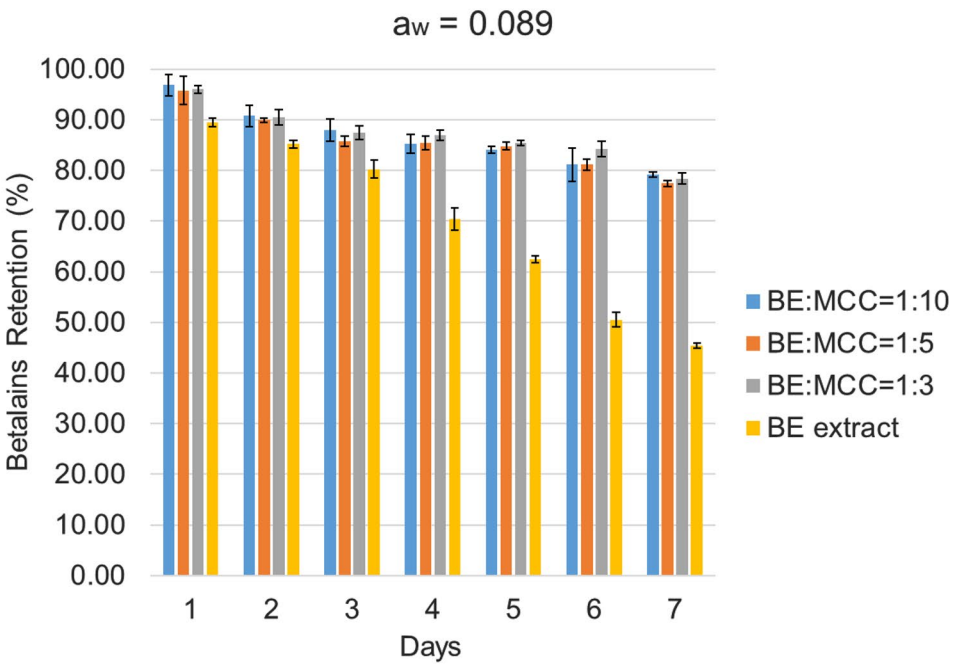


Fig. 4 Degradation of betalain extract and freeze-dried powders at (**a**) 80 °C and (**b**) 100 °C

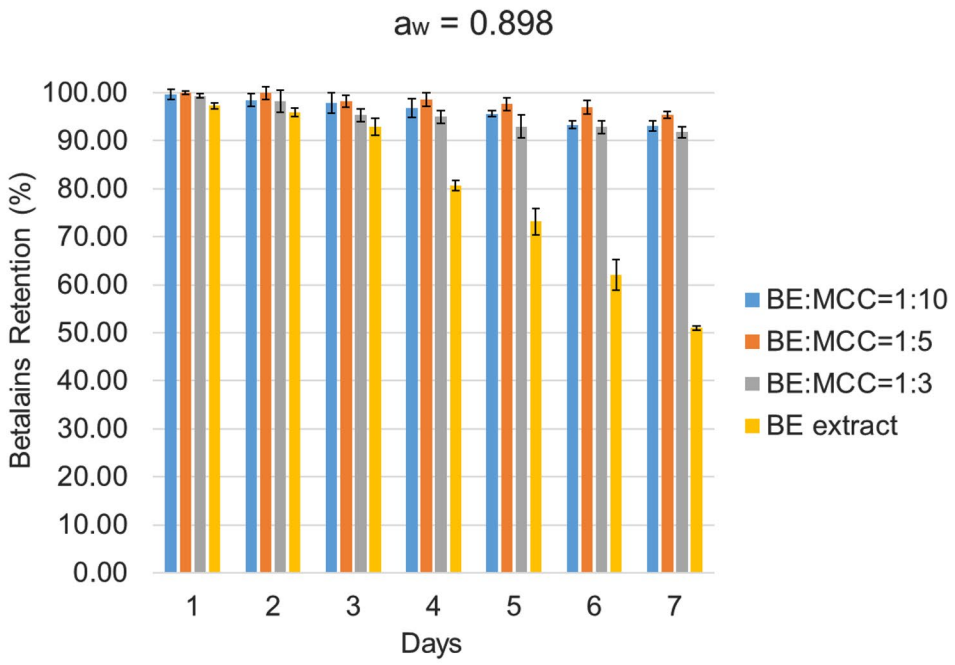
of Castro-Enríquez et al. [4]. However, the three powders remained about 60% of betalains content when exposed to air, compared to BE extract being almost colorless after 30 days. There is no difference between the remaining percentage of BE powders with light exposure and without light exposure, which means that light was not a negative factor for betalains when they avoided moisture. Thus, simply by packaging in vacuum bags to prevent moisture, betalains encapsulated with MCC could potentially be commercialized.

3.7 Application of Pectin and Encapsulated Betalains in Food Systems

Converting discharged red dragon fruit peel waste into natural pectin and stable betalains for producing value-added



(a)



(b)

Fig. 5 Degradation of betalain extract and freeze-dried powders at (**a**) a_w= 0.089 and (**b**) a_w= 0.898

products is a novel step in sustainable production toward circular bioeconomy. This study chose jam and jell as model products to show the initial applicability of encapsulated betalains and pectin. Owing to being stable over a wide pH range between 3 and 7, betalains may be used in low-acid foods such as dairy products (yogurt and ice cream) [39] and in syrups, sausages, and confectionaries. In addition, betalains may offer red colors at neutral pH conditions, and this property may be exploited to create more expensive and regulated color alternatives [35].

Pectin is one of the important fiber parts because it contributes to maintaining the colonic microflora, which is beneficial to human health [40]. The pectin extracted from dragon fruit peels was reported to display similar textural

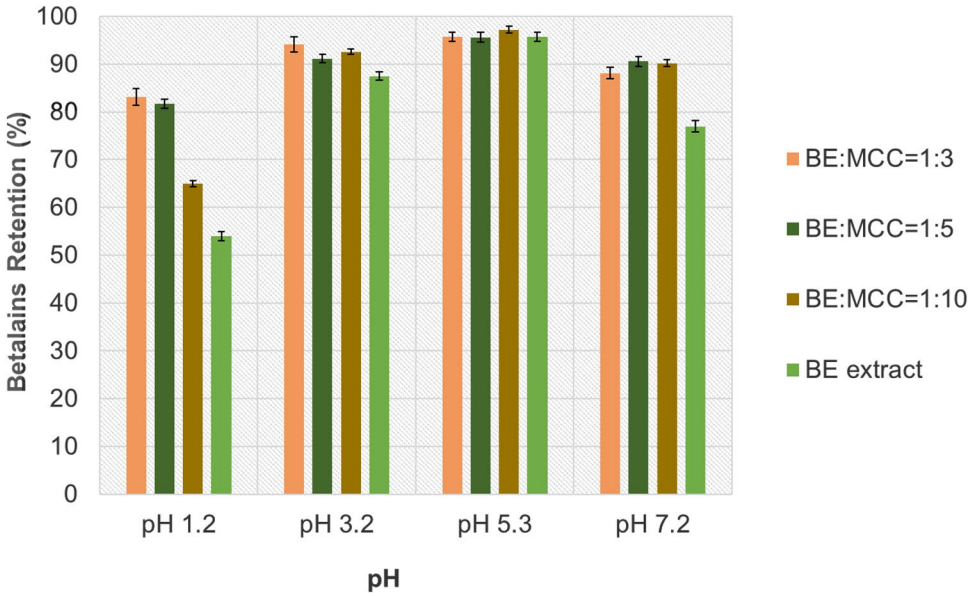


Fig. 6 Degradation of betalain extract and freeze-dried powders at pH 1.2, 3.2, 5.3, and 7.2

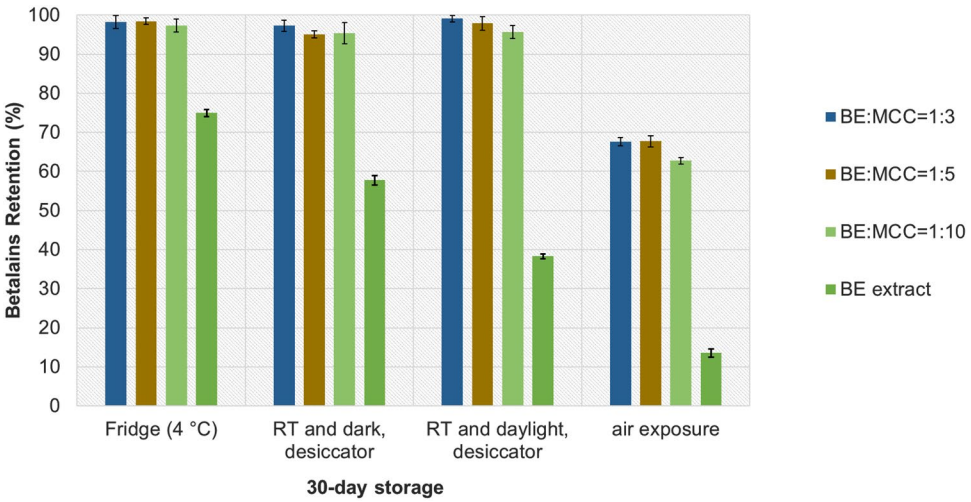


Fig. 7 Retention of betalain extract and freeze-dried powders at different storage conditions after 30 days

properties to those from apple and citrus peels, which are its common sources [1]. Previously, pectin from dragon fruit (*Hylocereus polyrhizus*) peel was applied successfully in pineapple jam, which was reported by Izalin et al. [41]. This group showed that dragon fruit peel pectin possessed several textural characteristics which were similar to apple and citrus pectin, for example, water and oil holding capacity and swelling capacity. Also, sensory evaluation and consumer acceptance of pineapple jam in the study demonstrated this pectin as an effective thickener in low-viscous food and beverages. Soedirga and Marchellin published a work that applied red dragon fruit peel pectin in combination with carrageenan to make jelly candy [42]. Their product improved some properties such as hardness, cohesiveness, gumminess and chewiness. Therefore, in this study, the food matrices structured by pectin are expected to be a suitable medium that assisted in protecting betalains during processing and storage. Pineapple jam and gummy candy were chosen as model food systems to apply pectin as a food thickener and freeze-dried betalains powder with the betalains-to-MCC ratio of 1:5 as a colorant. Pectin obtained from red dragon fruit peels has 58.6% of DE, which is proper for model food systems like jam and jellies. The microbial profile of the applied pectin powder is presented in Table 3. As discussed above, the loss of MCC-coated betalains was only 5–7% during the first 5-min heating at 80–100 °C. Thus, the colorant was added to the jam and jelly matrice at the end of the heating process. Betalains powder with the core-to-wall ratio of 1:5 (called BE:MCC = 1:5) after 30-day storage with light in the desiccator was also conducted microbiological tests before employing in the food system (as shown in Table 3). The overall microbial load data indicates that pectin and

freeze-dried betalains is safe for consumption and has a good shelf life. Table 4 also indicated the data on the viable microbial profile of jam and jellies after 2-week storage in the sealed vacuum bag. Total aerobic microorganisms of pineapple jam and gummy candy were 7800 and 590 CFU/g, respectively. These results were within the acceptable limit (10^4 CFU/g food) of the National technical regulation of Microbiological contaminants in food (QCVN 8-3:2012/BYT) [43]. There was no detectable growth of *E. coli, Staphylococcus, Salmonella*, yeast, and mold. Generally, although jam and jellies were preservative-free and were not pasteurized, they are safe for consumption after 2-week storage.

After two weeks of being stored in a vacuum-sealed bag, no considerable loss of betalains content was observed for both food systems colored with freeze-drying powder. Some previous works discussed that the light-induced degradation of betalains is boosted by oxygen [37]. Similarly, the effects of light exposure on the two food systems in this study are negligible under anaerobic conditions of vacuum-sealed bags. As in Table 5, the stability of betalains is greater than 90% after two-week vacuum storage. For both model food products, storage at low temperatures (4 °C in the fridge) ensured the best color stability, up to 97%. The investigation confirms that the MCC-encapsulated betalains can be used as a red colorant in jam and jellies stored under vacuum-sealed packaging or refrigeration and extends our knowledge about the potential application of betalains in pectin-based foods. Since no preservatives were used and the maintenance of the vacuum condition was limited, the observation was only conducted for two weeks. Further research is necessary to justify the actual applications.

3.8 Valorization Pathway of Red Dragon Fruit Peels

The design of sustainable production strategies to efficiently reduce fruit and vegetable waste is highly required and particularly valuable because they are underexploited sources of bioactive compounds. Red dragon fruit peels are the main solid waste obtained in high quantities after the extraction of juices, drying fruits, wine fermentation, etc. The extraction of phytochemicals, the recovery of dietary fiber, or the use of the whole red dragon fruit peel to functionalize and add value to food products are promising ways to use these fruit losses and wastes efficiently. A treatment procedure for red dragon fruit peels to yield different value-added products, including betalains, pectin, and ethanol, is proposed in this paper. A simplified flow diagram of the valorization process is proposed in Fig. 8. In the next phase of valorization pathway, the solid waste disposed of by the pectin extraction step will be reused to produce bioethanol by fermentation. In such a biorefinery approach, red dragon fruit peel waste can be used to produce a more varied range of biomaterials (pectin and betalains) and bioenergy (bioethanol or methane) via sustainable production processes and solid waste minimization. This idea will offer not only high-added value inputs for the formulation of food products, but also address the clean energy issue. It will also stimulate the sustainable development of local food industries, contributing to reducing carbon footprint [44]. Valorization of such wastes to produce innovative biomaterials, biochemicals, and biofuels can open potential market opportunities if efficiently exploited and developed.

Table 4 Microbial profile of pineapple and gummy candy after 2-week storage and colorant powder

Criteria	Unit	Pectin Powder	Dried BE Powder	Pineapple Jam	Gummy candy
Total aerobic microorganisms	CFU/g	nil*	470	7800	590
Escherichia coli	CFU/g	nil*	nil*	nil*	nil*
Staphylococcus aureus	CFU/g	-	nil*	nil*	nil*
Salmonella app	/25 g	–	nil*	nil*	nil*
Yeast and mold	CFU/g	nil*	nil*	nil*	nil*

**No colonies were observed in 10^{-1} dilution*

Table 5 Betalain Retention in food products: Pineapple jam and Gummy candies after 2-week storage in the fridge and vacuum-sealed bags

	Pineapple jam			Gummy candies		
	4 °C in fridge	Vacuum-sealed bag, daylight, RT	Vacuum-sealed bag, dark, RT	4 °C in fridge	Vacuum-sealed bag, daylight, RT	Vacuum-sealed bag, dark, RT
Betalains retention after 2-week storage (%)	98.23 ± 0.13	88.66 ± 0.87	92.30 ± 0.55	97.94 ± 0.30	91.02 ± 0.91	93.16 ± 0.78

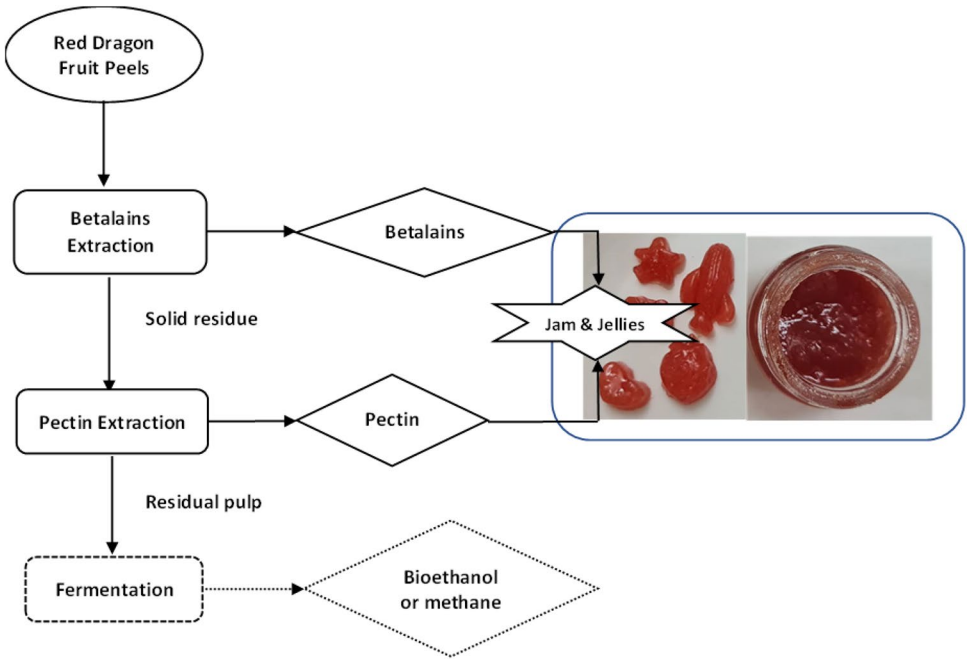


Fig. 8 Flow diagram for the production of betalains, pectin, and bioethanol from red dragon fruit peels

4 Conclusions

In this study, we demonstrated that betalains from red dragon fruit peels could be protected and stabilized by the different content of microcrystalline cellulose under various physicochemical factors (temperature, pH, water activity) and storage conditions (fridge and desiccator). MCC was proven an effective wall material for betalains encapsulation by freeze-drying technique that promises to replace maltodextrin. The MCC encapsulated betalains were incorporated as colorants for jam and jellies, and they had good stability of 90% for two weeks under vacuum storage at room temperature. Worthy of note in this work is the simultaneous extraction of betalains and pectin from red dragon fruit peels disposed of by local food processing companies and utilizing pectin in jam and jellies promoted a more protective food matrix for the encapsulated betalains. The processing technique used can contribute to the effective exploitation of the extensive red dragon fruit peel waste stream to produce high-quality natural food ingredients. Within the scope of the circular bioeconomy, a biorefinery approach proposed via the entire valorization of red dragon fruit peel waste to produce biomaterials and clean energy will be published in due course.

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Data Availability The generated data used to support the findings of this study are included within the article.

Declarations

Conflict of interest The authors declare that there are no conflicts of interest requiring disclosure in this article.

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