Comparisons between Conventional and Microwave-Assisted Extraction of Natural Colorant from Mesocarp and Exocarp of *Cocos Nucifera*

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Abstract: This study aimed at exploring the feasibility of using the mesocarp and exocarp of *Cocos nucifera* as a natural colorant using microwave-assisted extraction in which often regarded as a waste materials. The effects of microwave power and heating time on the yield of colorant, pH and colour determination were investigated. The highest colorant yield in mesocarp and exocarp was found to be 6.0% (8 min of extraction at 100 W) and 6.8% (12 min of extraction at 300W), respectively. While the control, extraction of colorant by conventional method (90 min at 90 °C), showed a percentage of 5.2% (mesocarp) and 3.6% (exocarp) yield of colorant. Meanwhile, pH of all extracts fluctuated throughout the experiment. The results proved that heating time and microwave power slightly affected the pH of all samples due to pH being inconsistent throughout the experiment. In colour determination, lightness (L^*) was slightly decreased for all samples which extracted at all microwave power which indicated that the longer the heating time the darker colour was extracted. Overall, the colour of exocarp was darker than mesocarp extract due to L^* , c^* and h^* values were lower than the mesocarp extract. The utilization of microwave-assisted extraction has significant improvement in the extraction of natural colorant compared to traditional boiling method. In fact, application of microwave irradiation method proved to be a rapid and improved technique for natural colorant extraction and extensively reduced the extraction time.

Key words: MAE (Microwave-assisted Extraction), exocarp, mesocarp, *Cocos nucifera*, natural dye.

1. Introduction

Nowadays, consumers are posed concerned on the safety of synthetic colorants or synthetic dyes. The use of synthetic dyes causes environmental pollution as majority of these dyes are toxic and are non-biodegradable [1]. Therefore, there is a growing demand for ecofriendly and non-toxic colorants or natural dye, particularly for food colouration and dyeing of child textile and leather garments [2]. Hence, natural dyes and pigments have become as an imperative option to potentially harmful synthetics dyes [3]. Natural colorants can be derived from plants, minerals and even some insects [4]. Plants are typical sources of natural colorant which have the greatest

biological diversity of all living organisms and produce a large variety of pigment compositions [5] and generally have a higher compatibility with the environment [1].

Cocos nucifera (coconut) is a member of the family Arecaceae (palm family), an ornamental tree grown in villages and towns in Malaysia [6]. The coconut is a functional food because it offers health benefits beyond its nutritional content [7]. The fruit contains three layers namely exocarp, mesocarp and endocarp. The exocarp (outer layer) and mesocarp (fibrous husk) make up the husk of coconut [8]. Globally, about 40 to 50 million tonnes of coconut are grown annually [9], producing 15 to 20 million tonnes of husks. Moreover, there are significant amounts of coconut waste available in Malaysia which should be recycled rather than discarded. Reusing and recycling industrial and

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agricultural residues can minimize the environmental impacts and consequently this has contributed to reconsideration of using this waste which comprises exocarp and mesocarp for extraction of natural colorant.

Recently, MAE (Microwave-assisted extraction) is widely employed for plant extraction method as findings from many researchers claimed that MAE can lead to a considerable increase in the yield and quality of plant's materials. MAE is stated to be as a simple, environment friendly and economical method for the extraction of biologically active compounds from diverse plant materials [10]. MAE is also recognized as a green technology because it reduces the application of organic solvent [11]. In summary, this work reveals the effects of microwave on the yield of natural colorant extracted, pH and colour measurement in comparison to the conventional method.

2. Experimental Setup

2.1 Preparation of Sample

Cocos nucifera was obtained from Tanjung Karang, Selangor. The matured (brown in color) coconut was chosen as the raw material in this experiment. The exocarp from the mesocarp was peeled off from cleaned coconut using a knife. Both parts were cut and dried in an oven at 60 °C for 24 h. The samples were finely ground using a grinder and was kept in dry place until further analysis.

2.2 Methods of Extraction

2.2.1 Extraction of Pigment Using MAE

Microwave-assisted extraction was performed using a microwave (Samsung, Korea). Approximately 2 g of ground coconut exocarp and mesocarp were weighed separately in conical flask before each sample was mixed with 40 mL of distilled water (ratio of 1:20). The mixtures were placed in the center of microwave containing a circular, 360° rotating carousel with different powers: 100, 300, and 500 W and with different durations of exposure: 2, 4, 6, 8, 10, 12

minutes. At the end of extraction, the conical flasks were cooled down to room temperature before the mixtures were strained using a tea strainer and followed by filtration using filter paper (CHM, Germany) (150 mm) in order to remove plant materials. The filtrates were kept at 4 °C in the dark prior to analysis. All experiments were performed in triplicates.

2.2.2 Extraction of Pigment by Conventional Method

To extract the pigment with conventional method, all of the procedures in section 2.2.1 were carried out but instead of microwave heating in extracting stage, a water bath heating was employed at constant temperature of 90 °C. The extraction was conducted in 90 min.

2.3 Analytical Methods

2.3.1 Yield of Natural Colorant

Approximately 1 g of sample was mixed with 20 mL of distilled water. To determine the yield of natural colorant, all the procedures in section 2.2.1 were performed. The yield of natural colorant of each sample was conducted by gravimetric method [12]. The filtrate was dried in a hot-air oven at 80 °C for overnight to make sure all the water was evaporated. The extract was then cooled down in a desiccator before sample was weighed. The yield of total colorant extracted was quantified using formula as follows:

$$\text{Yield of natural colorant\%} = \frac{\text{natural dye obtained}}{\text{weight of sample used (g)}} \times 100$$

2.3.2 PH Determination

The pH of filtrate was determined using a pH meter (Hanna, USA). The sample was measured in triplicate.

2.3.3 Colour Determination

Sample for colour determination was measured using colour reader (Konika Minolta, Japan). The colour of the sample was quantified in terms of L*, a*, b*, c* and h* values. The values L*, a*, b*, c* and h* are the variables in the CIELAB colour space where L*, a* and b* are representing lightness (L*), redness (+a*), greenness (-a*), yellowness (+b*) and blueness (-b*).

While c^* represents chroma or purity of colour and h^* represents hue (shade) of colour [13].

2.3.4 Statistical Analysis

All the experiments were carried out in triplicate, and the results were expressed as means $\pm$ SD (Standard deviation). Statistical analysis was done using SPSS software version 16 using Turkey test. A value of $p < 0.05$ was considered statistically significant.

3. Results and Discussion

Application of MAE method in extraction dramatically improved the extraction time and yield of natural colorant (Table 1 and Table 2). Table 1 shows no significant difference was observed ($p > 0.05$) for all mesocarp extracts. However, the experimental results indicated that the extraction of natural colorant from mesocarp using microwave irradiation (100 W) was highest at 8 min of extraction with 6.0% of natural colorant extracted. Though, a further increase in time resulted in a declined plot. In addition, natural

colorant extracted at 300 W and 500 W showed similar trend where the highest natural colorant extracted was at 12 min of extraction time which were 3.6% and 4.8%, respectively.

Employing conventional method, 90 min was required for yielding 5.2% of same natural colorant as microwave extracted colorant. This comparison reveals that the reaction time reduces from 90 min to 8 min but the yield of extract material was better in the microwave method rather than that from the traditional boiling method. The reason for the increment of yield could be due to better leaching of natural dye material from plant cell membranes and mass transfer to solvent assisted by microwave (electromagnetic) energy provided by microwave. A study has reported that microwave enhances the mass transfer of colouring matter from natural plant material and transport to the solvent medium [14].

As shown in Table 2, that no significant difference was observed ($p > 0.05$) for all exocarp extracts.

Table 1 The effect of heating time and microwave power on total yield of natural colorant of mesocarp extracts (yield obtained from conventional method: 5.2 (%w/w)).

Time (min)	Microwave power (W)		
	100	300	500
2	2.0 ^a $\pm$ 0.566	1.6 ^a $\pm$ 1.131	2.4 ^a $\pm$ 1.131
4	3.2 ^a $\pm$ 1.131	2.8 ^a $\pm$ 0.566	3.6 ^a $\pm$ 0.566
6	5.2 ^a $\pm$ 1.697	3.6 ^a $\pm$ 0.566	4.4 ^a $\pm$ 0.566
8	6.0 ^a $\pm$ 0.566	2.8 ^a $\pm$ 0.566	3.6 ^a $\pm$ 1.697
10	2.0 ^a $\pm$ 0.566	3.2 ^a $\pm$ 1.132	3.6 ^a $\pm$ 0.566
12	2.8 ^a $\pm$ 1.697	3.6 ^a $\pm$ 0.566	4.8 ^a $\pm$ 0.000

Values are represented as means $\pm$ standard deviation. Means that do not share the same letter in the same column were significantly different at $p < 0.05$.

Table 2 The effect of heating time and microwave power on total yield of natural colorant of exocarp extracts (yield obtained from conventional method: 3.6 (%w/w)).

Time (min)	Microwave power (W)		
	100	300	500
2	2.8 ^a $\pm$ 1.697	3.2 ^a $\pm$ 1.131	3.6 ^a $\pm$ 0.566
4	3.6 ^a $\pm$ 0.566	2.4 ^a $\pm$ 1.131	5.2 ^a $\pm$ 1.697
6	2.8 ^a $\pm$ 0.566	2.8 ^a $\pm$ 0.566	4.8 ^a $\pm$ 1.131
8	3.6 ^a $\pm$ 0.566	3.6 ^a $\pm$ 0.566	3.6 ^a $\pm$ 0.566
10	3.2 ^a $\pm$ 1.131	5.6 ^a $\pm$ 2.263	4.4 ^a $\pm$ 0.566
12	4.4 ^a $\pm$ 0.566	6.8 ^a $\pm$ 0.566	4.0 ^a $\pm$ 1.131

Values are represented as means $\pm$ standard deviation. Means that do not share the same letter in the same column were significantly different at $p < 0.05$.

However, the highest percentage yield of natural colorant extract of exocarp was at 300 W. The amount of colorant increased with the increasing extraction time and reached maximum (6.8%) at 12 min. Meanwhile, the yield extracted at 100 W was the highest at 12 min (4.4%). However, the microwave power at 500 W indicated the highest yield (5.2%) attained at 4 min before the value gradually declined. Due to the features of microwave, power level of 100 W, 300 W, and 500 W could not be prolonged above 12 min because the action resulted in burning of the sample. Meanwhile, the control for exocarp, extraction of colorant by conventional method showed a percentage of 3.6% yields of colorant.

Theoretically, microwave radiation loosens the cell wall matrix and leads to severing of the parenchymal cells [15], thereby the skin tissues are rapidly and extensively opened up by the microwave. This will enhance interaction between extracting agent and source material in extraction procedure [16]. As a result, permeation of the extracting agent will rise and leads to effective increase in the yield of colorant extracted.

Based on Fig. 1, 2 and 3, pH of all mesocarp extracts indicated a fluctuating result throughout the experiment where the pH ranged from 4.83 till 5.16. Similar pattern was also observed for exocarp extract which pH was between 6.11 and 6.41. The results proved that heating time and microwave power were slightly affected by the pH of all samples due to pH being inconsistent throughout the experiment. In addition, most of the sample showed marginal reduction in pH from initial time to the end of experiment and this might be caused by the leaching of natural dye material from plant cell membranes which affected the pH value. From the results obtained, mesocarp extracts were more acidic than the exocarp extracts. In comparison, pH obtained from conventional method was 4.77 (mesocarp) and 5.94 (exocarp) which also agreed with the previous statement.

The colour of sample extracted was evaluated by measurement of the CIEL*-a*-b* coordinates. As shown in Table 3, the results indicated that temperature and microwave power slightly affect on colour values. All mesocarp extracted with water had similar colour, with hue angle values was ranging from 70 to 85. For

Fig. 1 The effect of heating time and microwave power at 100 W on pH of natural colorant from mesocarp and exocarp extracts.

Fig. 2 The effect of heating time and microwave power at 300 W on pH of natural colorant from mesocarp and exocarp extracts.

Fig. 3 The effect of heating time and microwave power at 500 W on pH of natural colorant from mesocarp and exocarp extracts.

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Table 3 Colour coordinate of mesocarp and exocarp extracts.

Sample	Microwave power (W)	Time (min)	Color coordinates				
			L	a	b	c	h
Mesocarp	100	2	25.7	0.7	3.5	2.8	84.6
		4	25.8	0.5	3.2	3.3	82.0
		6	25.6	0.5	3.8	4.1	78.7
		8	24.5	0.9	2.8	3.0	79.0
		10	24.4	1.7	2.8	3.0	72.5
		12	23.8	1.0	2.3	3.3	68.3
	300	2	26.0	0.3	3.6	3.6	85.9
		4	25.6	0.7	3.9	4.0	80.6
		6	25.4	1.0	3.8	4.1	70.0
		8	25.9	0.9	4.1	4.2	83.5
		10	25.2	1.0	3.1	3.2	78.4
		12	24.7	1.9	3.8	4.2	80.5
	500	2	25.5	1.1	4.0	3.5	78.8
		4	25.4	1.2	3.9	4.0	72.9
		6	25.6	0.3	3.5	3.5	84.5
		8	26.4	1.2	3.0	3.7	75.5
		10	27.1	0.4	2.8	2.8	84.8
		12	27.2	0.3	2.2	2.2	80.5
Exocarp	100	2	22.7	1.9	2.9	3.2	57.5
		4	24.2	1.3	3.3	3.5	68.0
		6	22.8	1.0	3.5	3.7	79.5
		8	25.1	0.6	3.6	3.7	80.6
		10	24.4	0.9	2.7	2.9	71.1
		12	23.9	0.7	2.9	3.0	76.3
	300	2	25.1	0.9	3.4	3.7	75.6
		4	24.5	1.3	2.5	2.9	64.1
		6	24.7	1.2	3.1	3.3	66.8
		8	24.4	1.1	2.9	3.2	68.0
		10	26.8	1.1	4.5	1.3	72.9
		12	25.7	0.5	4.2	4.2	83.1
	500	2	24.3	1.5	2.8	3.2	61.0
		4	25.0	1.2	3.4	3.7	70.4
		6	24.9	1.1	2.5	2.9	67.9
		8	24.0	0.9	3.1	3.3	71.9
		10	23.8	0.5	2.2	2.3	77.6
		12	25.9	0.5	4.4	4.4	83.7
Mesocarp (control)	-	90	24.5	1.0	3.4	3.5	73.2
Exocarp (control)	-	90	24.3	2.7	3.4	4.7	57.2

all samples extracted at all microwave power, lightness (L^*) was slightly decreased which indicated that the longer the heating time, the darker the colour of extract. The L^* values for sample extracted at 100 W, 300 W and 500 W dropped from 25 to 24.6, 24.6 to 23.2 and 26.5 to 23.9, respectively. L^* represents lightness value,

the lower the lightness value represent higher the colour yield. In comparison, the L^* values for mesocarp and exocarp extracts conducted by boiling method were 24.5 and 24.3, respectively. Moreover, water extraction at high temperature could increase the solubility of plant pigments and phenolic compounds

(colorants) [17].

Overall, the colour of exocarp was darker than mesocarp extract due to L^* , c^* and h values were lower than the mesocarp extract. a^* and b^* represent the tone of the colour, positive values of a^* and b^* represent redder and yellower tones while negative shows greener and bluer tones. Based on Table 3, all samples indicated that natural colorant extracted was less yellow and red due to low a^* and b^* values. The emergence of plant pigments and phenolic compounds (colorants) was closely related to colourful substances consisting of biochromes, which absorb or reflect light of varying wavelengths [5].

4. Conclusions

In this study, experimentation conducted showed that extraction time and microwave power markedly influenced the natural colorant extraction from mesocarp and exocarp of *Cocus nucifera*. Application of microwave irradiation method in the extraction of natural colorant dramatically reduced extraction time as well as improved the yield of natural colorant compared to conventional method which take longer time but lower yield of colorant extracted. Moreover, the use of water as a solvent extraction can contribute to low operating cost, eco-friendliness, low energy consumption, non-flammability and non-toxicity.

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