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# Ultrasound-assisted Extraction of Natural Colourant from Husk of *Cocos nucifera*: A Comparison with Agitated-bed Extraction

**Rodiah, M. H.*, Nur Asma Fhadhila, Z., Noor Asiah, H., Aziah, M. Y. and Kawasaki, N.**

*UNISEL Bestari Jaya Campus, Faculty of Engineering and Life Sciences, Department of Science and Biotechnology, Jalan Timur Tambahan, 45600 Batang Berjuntai, Selangor, Malaysia*

## ABSTRACT

This study investigated the use of ultrasound-assisted extraction (UAE) and agitated-bed extraction (ABE) to improve extraction efficiency of dyes from exocarp and mesocarp of *Cocos nucifera*. The UAE method was carried out at a frequency of 37 kHz and power of 150 W at 30°C for 1 hour and ABE technique was performed at 30°C for 24 hours at the agitation of 150 rpm. The effects of different solvent types (methanol, ethanol and acetone) and the different percentage of solvent (20%-70%) were determined. The results showed although the colourant yield increased with the increase of solvent concentration up to 60%, statistic value indicated no significant difference ($p > 0.05$) for the value of yield using solvent range from 20% to 70% for both UAE and ABE methods. Therefore, the solvent concentration of 20% was selected as the optimum concentration for each type of solvent used for both ABE and UAE methods. In comparison, higher extraction yields were achieved by UAE method where the optimal yield of the colourant of the mesocarp was 7.6% using acetone as solvent and the exocarp yielded about 6.4% using acetone or methanol. The recovery of natural colourant using ultrasound was found to be highly dependent on the type of solvents (acetone > methanol > ethanol) for both mesocarp and exocarp. The present study suggests that UAE method should be employed for hauling out colouring materials from exocarp and mesocarp due to its effectiveness in terms of time and economical usage of solvent.

*Keywords:* Agitated-bed extraction (ABE), *Cocos nucifera*, exocarp, mesocarp, natural dye, ultrasound-assisted extraction (UAE)

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*E-mail addresses:*
rodiah@unisel.edu.my (Rodiah, M. H.)
asmafhadhila90@gmail.com (Nur Asma Fhadhila, Z.)
noorasiah@unisel.edu.my (Noor Asiah, H.)
aziahmy@unisel.edu.my (Aziah, M. Y.)
kawasaki@unisel.edu.my (Kawasaki, N.)
*Corresponding Author

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## INTRODUCTION

There is a growing demand for eco-friendly and non-toxic colourants or natural dyes, particularly for food colouration and textile as well as leather garments (Palanivel et

al., 2010). The demand for natural dyes all over the world is estimated to be 10,000 tonnes, which account for only 1% of the world synthetic dyes consumption but expected to rapidly grow in the near future (Sivakumar et al., 2011). *Cocos nucifera* (coconut) is a member of the family Arecacea (palm family), an ornamental tree growing in villages and towns in Malaysia (Abdulelah et al., 2011). The fruit contains three layers, namely exocarp (outer layer), mesocarp (fibrous husk) and endocarp (flesh). The exocarp and mesocarp make up the husk of coconut (Victor, 2013).

In Malaysia, there is an abundance of coconut husk which should be recycled. The researchers' previous study has successfully extracted natural dye using microwave-assisted extraction from these waste in order to make better use of this cheap and abundant agricultural waste. Five compounds were detected in dye extract of mesocarp, namely catechin conjugate, petunidin-3-glucoside, proanthocyanidin trimer, cyanidin-3-sambubioside and delphinidin-3-glucoside whereas four compounds were identified in dye extract exocarp which were catechin, epicatechin, petunidin-3-glucoside and cyanidin-3-(6'-feruloylsophoroside)-5-glucoside (Zulrushdi, 2018).

Previous studies have shown that agitated-bed extraction (ABE) is one of the methods to extract dyes due to its effortless and inexpensive protocol. The ABE is a type of mechanical agitation which is a simple and low-cost method that uses agitation or mixing action to extract certain phytochemical content from plant samples in a shake-flask which is placed onto a rotary shaker, or with a magnetic stirrer submerged into the flask directly (Lau et al., 2010). The ABE was also used in a study on the extraction of anthocyanins from skins of jabuticaba. In this study, ABE was compared with pressurised liquid extraction (PLE). However, the results showed PLE resulted in higher extraction efficiency compared with ABE method (Veggi, Santos, & Meireles, 2011).

The ABE method was also used in the super-critical fluid extraction (SFE) process for the recovery of phenolics using grape bagasse from Pisco residues. An agitated bed extraction was performed by placing the grape bagasse into a 250 ml of Erlenmeyer flask containing ethanol (96%). The extractions were performed in a shaker with agitation (168 rpm) for 6 hours. The findings indicated higher extraction yields were achieved by agitated bed extraction than those obtained with the supercritical fluid methods (Farías-campomanes, Rostagno, & Meireles, 2013). In another study, ABE method was compared with microwave assisted extraction (MAE) of flavonoids from cultivated *E. sagittatum* and the result showed microwave technique resulted a significant increase in extraction yield of flavonoids (Zhang et al., 2013).

Sivakumar et al. (2011) reported another improved method called ultrasound-assisted extraction (UAE) which could extract higher yield of dyes. The UAE has the capacity to improve extraction efficiency by promoting mass transfer and possible rupture of cell wall due to its effect in acoustic cavitation (Shirsath et al., 2012). Apart from that, UAE can produce cavitations in the investigated sample, and the collapse of these cavitations bubbles induces a mechanical stress on the cells. These resulted in cell disruption and penetration of the solvent into the cells (Leonelli & Mason, 2010). The UAE technology can possibly improve extraction of components, such as anthocyanins, polyphenolics, polysaccharides, aromatic compounds, and oils (Shirsath et al., 2012).

The utilisation of ultrasonic has been investigated recently in the extraction of the bioactive compound from plant materials. Examples are the extraction of total phenolics from coconut shell powder (Rodrigues & Pinto, 2007), rice bran (Tabaraki & Nateghi, 2011), the extraction of anthocyanins from Garcinia indica Choisy (Nayak & Rastogi, 2011), *Delonix regia* tree flowers (Adjé et al., 2010), carotenoids (Sun et al., 2011) and betalains (Sivakumar et al., 2009). The efficacy of ultrasound-assisted extraction is also proven to be more predominant in getting higher yield with shorter extraction time when contrasted with other extraction methods, such as the conventional reflux in the extraction of phenolic acid from *Citrus unshiu Marc* peels (Ma et al., 2009) and the extraction of salvianolic acid B from *Salvia miltiorrhiza* root (Dong et al., 2010). The use of ultrasound was found to have a significant enhancement in the extraction efficiency of colourant obtained from beet root. Around 80% improvement in the yield of the colourant was accomplished with ultrasound compared with the magnetic stirring process using 1:1 ethanol–water (Sivakumar et al., 2009). Zou et al. (2010) demonstrated that natural melanin from dried fruit bodies of *Auricularia auricula* was successfully extracted using ultrasound with significant yields of the melanin.

The nature of the bioactive compound present in a plant is varied and depends on the plant materials. Hence, in general, it is extremely difficult to suggest an appropriate extracting solvent for individual plant materials. The main objective of the present study is to evaluate the effect of various solvents (methanol, ethanol and acetone) and different types of extraction methods (agitated bed extraction and ultrasound assisted extraction) on the extractability of natural dye.

## MATERIALS AND METHODS

### Preparation of Sample

The exocarp (outer layer) and mesocarp (fibrous husk) of *Cocus nucifera* (coconut) were obtained from Tanjung Karang, Selangor (Malaysia). In this study, only mature (brown colored) husks were used. Exocarp was separated from the mesocarp before both parts were cut and dried in an oven at 50°C for 2 days. The samples were finely ground using a grinder, sieved with a 0.5 mm sieve and kept in a clean plastic container, away from heat and moisture prior to conducting the experiments.

### Methods of Extraction

**Ultrasound-assisted extraction (UAE) and Agitated-bed extraction (ABE).** Approximately 3g of ground coconut exocarp and mesocarp were weighed separately before each of the samples was mixed with 60 mL of ethanol. In this study, the concentration of solvents varied ranging from 20% to 70%. The samples were sonicated at a frequency of 37 kHz and power of 150 W in a sonicator bath (Elmasonic, Germany) at 30°C for an hour based on the previous method adopted by Asma et al. (2015). The extraction process could not be prolonged beyond one hour as the temperature of water in the sonicator bath was increased to 65 to 70°C. The increased temperature caused dryness to the sample due to solvent evaporation. After ultrasound

extraction, the samples were filtered using filter paper (CHM, Germany) in order to remove plant materials before the solvent was evaporated using a rotary evaporator at room temperature for 5 minutes. The samples were then centrifuged at 10,000 rpm for 10 minutes. The supernatant was stored at 4°C in the dark prior to analysis. The same procedures were employed using different solvents (methanol and acetone).

The ABE was performed using an incubator shaker (Stuart, United Kingdom) at 30°C for 24 hours at an agitated speed of 150 rpm. The parameters for ABE extraction were examined in previous studies which indicated that maximum yield was obtained at 24 hours. A shorter period of extraction resulted in non-significant yield (unpublish data). Similar procedures of sample preparation, filtration, and removal of solvent and collection of the sample were the same as those used in the UAE technique.

## Analytical Methods

**Gravimetric Analysis.** The yield of the natural colourant of each sample was determined by a gravimetric method which was performed according to the method described by Sivakumar et al. (2011). Sample extracts were dried in a hot air oven at 80°C, overnight until all the water evaporated. The samples were then cooled in a desiccator and weighed until a constant weight was achieved. The yield for total extract was calculated using the following formula:

$$\% \text{ yield of natural colourant} = \frac{\text{Natural dye extract obtained (g)}}{\text{Weight of sample used (g)}} \times 100$$

$$\% \text{ improvement of UAE compared to ABE} = \frac{\% \text{ yield of (UAE process–ABE process)}}{\% \text{ yield of ABE process}} \times 100$$

**Colour Measurement Analysis.** The colour was evaluated using a colorimeter (Konika, Minolta, Japan). The colour of the sample was quantified in terms of L*, a*, b*, c* and h* values which are the variables in the CIELAB color space and described as follows: A negative value of L* denotes darker shade while a e positive value of L* indicates a a lighter shade. Negative value of a* is indicated by greener color and positive value of a* is indicated by red. Negative value of b* is indicated by blue color whie positive value of b* is indicated by yellow color. c* signifies chroma or purity of colour while h* represents hue (shade) of color.

**Statistical Analysis.** All the experiments were carried out in triplicate, and the results were expressed as means ± SD (standard deviation). Statistical analysis was done using SPSS software version 16 using the Tukey test. A value of $p<0.05$ was considered statistically significant. Meanwhile, the statistical significance for comparison between UAE and ABE was evaluated using Student's t-test and set at <0.05.

## RESULTS AND DISCUSSION

### Colourant yield between UAE and ABE

**Mesocarp Extract.** The percentage yield of natural dye of mesocarp extracted using agitated bed and ultrasound are shown in Table 1. In order to identify the most effective solvent which contributes the highest yield of colourant, varying the types and compositions of solvent were done. This is crucial as Shirsath et al. (2012) also mentioned that the amount of solvent utilised in the operation is essential and in some cases, it is beneficial to add some amount of water to the solvents. In this study, the concentration of solvent was chosen from 20% to 70% because the preliminary study showed that solvent at the concentration less than 20% did not give much effect on dye extraction within 24 hours. The concentration of the solvent that is more than 70% will evaporate very quickly within 30 to 60 minutes when employed, causing dryness to the sample. Based on the results, the highest yields were obtained when mesocarp was extracted with acetone at the concentration range between 50% and 60% using UAE method. This indicated that extending the concentration of solvent beyond 60% was unnecessary and also could result in reducing the amount of colourant extracted, which is supported previous findings. Shirsath et al. (2012) reported that to enhance the extraction of isoflavones from soybeans it was important to include a specific amount of water (40–60%) to the extracting solvent. This was probably due to the relative polarity of the compounds, and the increased propagation of ultrasonic waves in aqueous solvents. When large volumes of water (higher than 60%) were added, the effectiveness of extraction was reduced possibly attributed to increased production of radicals from the ultrasound induced dissociation of water (Shirsath et al., 2012).

Table 1
*Percentage yield of natural dye from coconut husk (Mesocarp) extracts in different concentration of solvent*

| Extraction methods | Concentration (%) | Type of Solvent | | |
|---|---|---|---|---|
| | | Ethanol (%) | Methanol (%) | Acetone (%) |
| Ultrasound-assisted extraction | 20 | $7.2^{a} \pm 1.13$ | $5.2^{a} \pm 1.69$ | $7.6^{a} \pm 1.69$ |
| | 30 | $7.2^{a} \pm 1.13$ | $6.8^{a} \pm 0.56$ | $8.4^{a} \pm 0.56$ |
| | 40 | $7.6^{a} \pm 0.56$ | $7.2^{a} \pm 2.26$ | $8.0^{a} \pm 2.26$ |
| | 50 | $6.8^{a} \pm 1.69$ | $6.8^{a} \pm 0.56$ | $9.2^{a} \pm 0.56$ |
| | 60 | $6.0^{a} \pm 0.56$ | $6.8^{a} \pm 0.56$ | $9.2^{a} \pm 0.56$ |
| | 70 | $5.2^{a} \pm 0.56$ | $6.0^{a} \pm 0.56$ | $6.8^{a} \pm 0.56$ |
| Agitated-bed extraction Agitated-bed | 20 | $4.0^{a} \pm 1.13$ | $5.2^{a} \pm 0.56$ | $6.0^{a} \pm 2.82$ |
| | 30 | $5.6^{a} \pm 1.13$ | $4.4^{a} \pm 1.69$ | $5.6^{a} \pm 0.00$ |
| | 40 | $6.0^{a} \pm 1.69$ | $8.0^{a} \pm 1.13$ | $5.2^{a} \pm 2.82$ |
| | 50 | $6.0^{a} \pm 0.56$ | $8.0^{a} \pm 2.26$ | $7.2^{a} \pm 0.00$ |
| | 60 | $6.4^{a} \pm 1.13$ | $6.8^{a} \pm 0.56$ | $6.8^{a} \pm 2.82$ |
| | 70 | $3.6^{a} \pm 1.34$ | $5.6^{a} \pm 1.13$ | $3.1^{a} \pm 0.35$ |

Results as means from triplicates. $^{a–b}$ Different superscript letters between the concentration of solvent for each extraction method denote significant differences ($p < 0.05$)

On the other hand, in ABE method, methanol extract at the concentration of 40%-50% revealed the highest percentage of colourant yield (8.0%) compared with other solvents. Apparently, for both methods, the yield of colourant increased markedly with the increase of the concentration of each type of solvent and reached a peak value with 50% of concentration. However, there was no significant difference ($p > 0.05$) among the concentration of each solvent used (20%-70%). Therefore, the solvent concentration of 20% was selected as the optimum concentration for each type of solvent used for both ABE and UAE methods.

**Exocarp Extract.** The percentage yield of the natural dye of exocarp for extraction using agitated bed and ultrasound is shown in Table 2. Similarly, an effective solvent for extracting natural dye from exocarp of *Cocos nucifera* was identified by varying the types and composition of the solvent. Based on the results, the highest yield (11.6%) was obtained when exocarp was extracted with acetone at the concentration of 50% using UAE method. Whereas, in ABE method, methanol extract at the concentration of 50%-60% gave the highest percentage of colourant (7.6%). However, there was no significant difference ($p > 0.05$) among the concentration of each solvent used (20%-70%). Therefore, the solvent concentration of 20% was selected as the optimum concentration for each type of solvent used in the exocarp sample.

Table 2
*Percentage yield of natural dye from outer layer (Exocarp) extracts in different concentration of solvent*

| Extraction methods | Concentration (%) | Type of Solvent | | |
|---|---|---|---|---|
| | | Ethanol (%) | Methanol (%) | Acetone (%) |
| Ultrasound-assisted extraction | 20 | $6.0^{a} \pm 0.56$ | $6.4^{a} \pm 1.13$ | $6.4^{a} \pm 1.13$ |
| | 30 | $5.2^{a} \pm 0.56$ | $5.6^{a} \pm 1.13$ | $7.2^{a} \pm 2.26$ |
| | 40 | $9.2^{a} \pm 1.69$ | $6.0^{a} \pm 1.69$ | $10.4^{a} \pm 1.13$ |
| | 50 | $6.8^{a} \pm 0.56$ | $7.6^{a} \pm 0.56$ | $11.6^{a} \pm 2.28$ |
| | 60 | $8.4^{a} \pm 2.82$ | $6.8^{a} \pm 1.69$ | $8.8^{a} \pm 1.13$ |
| | 70 | $5.2^{a} \pm 0.56$ | $6.8^{a} \pm 1.69$ | $7.2^{a} \pm 1.13$ |
| Agitated-bed extraction | 20 | $5.2^{a} \pm 0.56$ | $2.8^{a} \pm 0.56$ | $2.1^{a} \pm 0.14$ |
| | 30 | $6.8^{a} \pm 0.56$ | $5.6^{ab} \pm 1.13$ | $5.6^{a} \pm 0.00$ |
| | 40 | $6.4^{a} \pm 1.13$ | $3.6^{a} \pm 0.56$ | $6.0^{a} \pm 1.69$ |
| | 50 | $7.2^{a} \pm 2.26$ | $7.^{6b} \pm 1.69$ | $6.0^{a} \pm 1.69$ |
| | 60 | $6.8^{a} \pm 0.56$ | $7.6^{b} \pm 0.56$ | $4.4^{a} \pm 0.56$ |
| | 70 | $5.2^{a} \pm .69$ | $5.2^{ab} \pm 1.69$ | $4.0^{a} \pm 1.13$ |

Results as means from triplicates. Different superscript letters between the concentration of solvent for each extraction method denote significant differences ($p < 0.05$).

Based on the observation, the increase of solvent concentration (ethanol, methanol and acetone) up to 70% resulted in decreasing pattern of the extraction yields for both UAE and ABE. The alcohol vaporised easily during the extraction process and resulted in slight dryness to the medium of extraction. The finding is in agreement with Cacace and Mazza (2003) who

reported that the increase in ethanol concentration reduces the dielectric constant of the solution and consequently reduces the interaction energy between the solute and solvent (Cacace & Mazza, 2003).

**Comparison between UAE and ABE based on Selected Optimum Condition.** In order to validate the efficiency of extraction method, a comparison was made between UAE and ABE methods based on the optimum concentration (20%) for each type of solvent used (Table 3). Under the optimised conditions, the greatest colourant yield was extracted by ethanol for the mesocarp and there was 80% of improvement in the percentage yield of coloruing matter extract obtained due to the use of ultrasound compared with ABE method. This may be due to the intensity of ultrasonic cavitation in the ethanol mix with water increase as a result of the increase in surface tension and the decrease in viscosity as stated by Boonkird et al. (2008). Meanwhile, a three-fold increase in the percentage yield of the colourant was seen using the UAE method when the exocarp was extracted using acetone. This enhancement could be attributed to cavitation and thermal effects of the ultrasound technique, which cause disruption of the cell wall and intensification of mass transfer (Esclapez et al., 2011; Vajic et al., 2015).

Table 3
*The effect of ultrasound on the yields of colourant extracted from mesocarp and exocarp using ethanol, methanol and acetone at 20% concentration*

| Plant sample | Type of solvent | % yield of UAE (A) | % yield of ABE (B) | % improvement due to ultrasound = ((A-B)/B) × 100 |
|---|---|---|---|---|
| Mesocarp | Ethanol | $7.2^{a} \pm 1.13$ | $4.0^{b} \pm 1.13$ | 80.0 |
| | Methanol | $5.2^{a} \pm 1.69$ | $5.2^{a} \pm 0.56$ | 0.0 |
| | Acetone | $7.6^{a} \pm 1.69$ | $6.0^{a} \pm 2.82$ | 26.6 |
| Exocarp | Ethanol | $6.0^{a} \pm 0.56$ | $5.2^{a} \pm 0.56$ | 15.4 |
| | Methanol | $6.4^{a} \pm 1.13$ | $2.8^{b} \pm 0.56$ | 128.6 |
| | Acetone | $6.4^{a} \pm 1.13$ | $2.1^{b} \pm 0.14$ | 204.8 |

Results as means from triplicates. $^{a–b}$ Different superscript letters between the percentage of yield between UAE and ABE for each solvent denote significant differences (independent t-test, $p<0.05$)

In this study, the mesocarp and exocarp samples were sonicated at a frequency of 37 kHz and power of 150 W in a sonicator bath at 30°C for an hour. In a similar manner, Khan et al (2010) discovered that the UAE method was faster and more efficient than industrial hot maceration process. The results revealed that UAE at ultrasonic bath operating at 35 kHz for 3 h gave 87.4% yield compared with 79.4% of the maceration process for 15 h. Rouhani et al. (2009) studied the effect of UAE on the extraction of curcuminoids from turmeric plant rhizomes using ultrasonic bath at 35 kHz for 15 min and discovered that the yield of UAE was three times higher than the traditional method (solvent extraction, soxhlet extraction and maceration).

The efficacy of ultrasonic-assisted extraction not only improved natural colourant yield recovery but also reduced extraction time. Despite the long period of extraction up to 24 h, the low yield of colourant values was observed for both mesocarp and exocarp produced using the

agitated-bed. In this study, the extraction time is shortened to 1 h from 24 h if the ultrasound is chosen as the preferable method. Thus, the UAE process with 20% ethanol and 20% acetone as extracting solvent seems to be the best combination for the extraction method of natural dye recovery for both mesocarp and exocarp.

However, it is crucial that the sonication time is carefully optimised, since exposure to ultrasonic irradiations may damage the quality of the solute in certain heat sensitive materials. (Romdhane & Gourdon, 2002). Indeed the findings are in agreement with Rouhani et al. (2009) who reported that extraction of curcuminoids from turmeric plant rhizomes using ultrasonic bath increased but if the extraction time was longer than 15 min, the extraction yield declined. The finding showed that the induced cavitation can degrade compounds when exposed for a longer time.

Generally, agitated bed extraction mainly depended upon continuous permeation and solubilisation processes to leach target constituents. Extraction of colouring matter is a solid-liquid leaching process involving mass transfer problem (Sivakumar et al., 2011). Since the colouring matter is tightly bound to plant cell membranes, extraction could be improved by methods such as ultrasound which use shorter time (less than an hour). This finding is supported by Sivakumar et al. (2011) who concluded that the improvement of yield of extraction was due to better leaching of natural dye material from plant cell membranes and mass transfer to solvent assisted by acoustic cavitation from the ultrasound.

The percentage of recovery of natural colourant using ultrasound was found to be highly dependent on the type of solvents (acetone > methanol > ethanol) for both mesocarp and exocarp. The determination of the best solvent for ultrasound assisted extraction typically relies on the physical properties (surface tension, vapor pressure and viscosity) of the solvent because these properties influence the cavitational intensity in a liquid phase. Solvents vary in their extraction abilities depending on their own and the solute's chemical structure. Once the functional group of the solute is identified, possible solvents can be chosen. Common solvents used are acetone, alcohols, and ether to extract bio-active substances from natural products due to their broad solubility propensity (Shirsath et al., 2012).

In the present study, acetone exhibited the highest yield which may be attributed to its semi-polar characteristic which might attract both polar and nonpolar compounds. In fact, it was verified that most natural antioxidants from plants, similar to phenolic compounds, flavonoids and anthocyanins, are easily dissolved by low-polar organic solvents, such as ethanol, methanol, and acetone, with different levels of water (Xu et al., 2017). Indeed, these findings indicated that organic solvents diffuse into solid material during extraction procedure and then solubilise the compound with similar polarity. The nature of the solvent used decides the sort of chemicals likely extracted from plant materials (Tiwari et al., 2011). Polarity is the capacity of a molecule to engage in strong interactions with other polar molecules (Barwick, 1997). Most of the bioactive components of plant matrices are medium-sized molecules. The presence of aromatic delocalised μ-electrons may cause the molecules to be highly polarisable (Kolar et al., 2002).

Jadhav et al., (2009) reported that various solvents for extraction of vanillin demonstrated that the utilisation of polar solvents resulted in a maximum degree of extraction. The use of

polar and non-polar solvents combined with ultrasonic energy has been reported to result in quick and effective extraction of total lipids from solid matrices (Metherel et al., 2009). Furthermore, Shirsath et al. (2012) described that solvents, such as methanol, ethanol and hexane are frequently used for UAE instead of water.

## Colour Strength

The results of different types and solvent proportions on the color of the natural dye of mesocarp and exocarp using ultrasound-assisted extraction (UAE) and agitated bed extraction (ABE) are shown in Table 4 and Table 5. The color values of the samples were measured in terms of L*, a*, b*, c* and h. This indicated that the method employed for the extraction slightly affected the colour values. All mesocarp and exocarp extracted from UAE and ABE had almost similar color, with lightness (L*) values ranging from 23.2 to 27.7 (mesocarp) and 23.3 to 31.8 (exocarp). However, the colour was lighter using UAE method (compared with ABE method). L* represents lightness value, the lower the lightness value, the higher the colour yield. In addition, the results obtained were correlated with the result from the percentage yield of colourant extracted using both methods of UAE and ABE (Table 1).

Table 4
*Color coordinate of mesocarp extracts using ultrasound-assisted extraction (UAE) and agitated-bed extraction (ABE) technique*

| Method of extraction | Solvent | Concentration | Color Coordinates | | | | |
|---|---|---|---|---|---|---|---|
| | | (%) | L* | a* | b* | c* | h* |
| UAE | Ethanol | 20 | 24.4±0.8 | 1.4±0.4 | 3.9±0.7 | 4.1±0.8 | 70.6±2.9 |
| | | 30 | 25.8±0.2 | 0.3±0.1 | 1.9±1.3 | 2.0±1.4 | 73.2±7.5 |
| | | 40 | 24.0±0.1 | 1.3±0.2 | 1.2±0.2 | 1.8±0.1 | 47.4±0.5 |
| | | 50 | 24.5±0.5 | 0.7±0.4 | 2.2±0.8 | 2.3±0.5 | 68.0±18.1 |
| | | 60 | 23.5±0.5 | 0.4±0.1 | 0.9±0.3 | 0.9±0.2 | 68.2±8.1 |
| | | 70 | 24.1±1.8 | 4.4±2.4 | 3.5±1.2 | 5.7±2.3 | 40.1±12.9 |
| | Methanol | 20 | 25.0±0.1 | 1.4±0.0 | 3.5±0.6 | 3.8±0.5 | 67.9±3.1 |
| | | 30 | 24.9±0.3 | 0.3±0.0 | 5.2±0.5 | 5.2±0.5 | 87.1±0.4 |
| | | 40 | 25.1±0.0 | 1.6±0.3 | 3.6±0.4 | 4.0±0.6 | 66.2±2.9 |
| | | 50 | 25.2±0.7 | 2.0±0.9 | 3.6±0.1 | 4.2±0.4 | 61.5±11.2 |
| | | 60 | 24.9±0.2 | 0.5±0.2 | 3.7±0.8 | 3.8±0.7 | 81.5±4.4 |
| | | 70 | 24.9±1.5 | 2.2±1.4 | 5.3±1.3 | 5.8±1.6 | 69.3±10.2 |
| | Acetone | 20 | 25.1±0.1 | 0.4±0.2 | 4.1±0.1 | 4.1±0.1 | 84.8±2.2 |
| | | 30 | 25.1±0.4 | 0.7±0.1 | 3.0±0.2 | 3.1±0.2 | 74.1±2.7 |
| | | 40 | 24.7±0.4 | 1.2±0.4 | 3.7±0.2 | 3.8±0.2 | 72.6±4.7 |
| | | 50 | 24.0±0.4 | 1.5±1.3 | 3.7±1.0 | 4.2±0.4 | 66.9±21.7 |
| | | 60 | 23.3±0.6 | 2.9±0.8 | 2.8±0.1 | 4.0±0.7 | 45.0±6.9 |
| | | 70 | 24.5±0.3 | 3.1±0.4 | 3.6±0.4 | 4.8±0.5 | 49.3±0.3 |

Table 4 *(continue)*

| Method of extraction | Solvent | Concentration | Color Coordinates | | | | |
|---|---|---|---|---|---|---|---|
| | | (%) | L* | a* | b* | c* | h* |
| ABE | Ethanol | 20 | 24.5±1.1 | 1.7±0.2 | 10.6±1.3 | 10.7±1.3 | 80.9±0.1 |
| | | 30 | 25.3±1.3 | 0.5±0.3 | 5.1±2.8 | 5.2±2.8 | 84.5±1.5 |
| | | 40 | 25.8±0.6 | 1.7±0.2 | 5.8±2.5 | 5.9±2.3 | 79.8±15.6 |
| | | 50 | 27.4±1.0 | 0.6±0.3 | 4.4±1.3 | 4.5±1.2 | 80.2±8.7 |
| | | 60 | 26.3±1.2 | 1.7±0.9 | 6.2±3.1 | 6.5±2.7 | 71.2±18.4 |
| | | 70 | 27.5±0.6 | 0.1±0.2 | 4.9±0.2 | 4.9±0.2 | 88.3±2.7 |
| | Methanol | 20 | 31.8±0.1 | 0.6±0.2 | 1.3±0.1 | 1.5±0.0 | 67.2±7.8 |
| | | 30 | 31.3±1.0 | 0.7±0.4 | 1.5±0.9 | 1.7±1.0 | 64.2±4.2 |
| | | 40 | 29.5±0.0 | 0.5±0.4 | 2.2±0.4 | 2.3±0.3 | 75.7±11.8 |
| | | 50 | 30.2±0.4 | 0.1±0.3 | 0.4±0.5 | 0.5±0.5 | 74.3±2.2 |
| | | 60 | 31.4±0.6 | 0.9±0.5 | 3.4±0.4 | 3.5±0.1 | 75.2±9.5 |
| | | 70 | 30.3±0.0 | 0.6±0.5 | 3.0±0.2 | 3.1±0.1 | 78.6±9.8 |
| | Acetone | 20 | 30.9±0.1 | 0.3±0.2 | 2.4±0.2 | 2.5±0.2 | 81.4±5.0 |
| | | 30 | 31.5±0.1 | 0.4±0.3 | 1.6±0.1 | 1.7±0.0 | 77.8±9.0 |
| | | 40 | 31.5±0.1 | 0.5±0.1 | 1.6±0.1 | 1.7±0.1 | 73.5±3.0 |
| | | 50 | 31.8±0.1 | 0.6±0.2 | 1.3±0.1 | 1.5±0.0 | 67.2±7.8 |
| | | 60 | 31.2±0.1 | 5.5±0.2 | 11.4±0.1 | 12.6±0.0 | 64.4±7.8 |
| | | 70 | 30.4±0.4 | 1.5±0.1 | 3.4±0.5 | 3.8±0.5 | 67.6±2.5 |

Table 5
*Color coordinate of exocarp extracts using ultrasound-assisted extraction (UAE) and agitated-bed extraction (ABE) technique*

| Method of extraction | Solvent | Concentration | Color Coordinates | | | | |
|---|---|---|---|---|---|---|---|
| | | (%) | L* | a* | b* | c* | h* |
| UAE | Ethanol | 20 | 24.1±0.2 | 1.8±0.2 | 1.7±0.2 | 2.4±0.1 | 43.8±7.9 |
| | | 30 | 23.3±0.6 | 2.9±0.8 | 2.8±0.1 | 4.0±0.7 | 45.0±6.9 |
| | | 40 | 23.9±0.1 | 1.9±0.3 | 2.9±0.3 | 3.5±0.4 | 56.7±1.8 |
| | | 50 | 23.7±0.6 | 1.9±1.1 | 2.5±0.6 | 3.2±0.8 | 53.5±17.1 |
| | | 60 | 23.2±1.5 | 3.2±2.9 | 2.0±0.5 | 4.0±2.3 | 40.8±28.5 |
| | | 70 | 23.4±1.6 | 3.3±2.8 | 2.5±0.9 | 4.6±1.8 | 45.1±29.2 |
| | Methanol | 20 | 23.3±0.6 | 2.9±0.8 | 2.8±0.1 | 4.0±0.7 | 45.0±6.9 |
| | | 30 | 24.2±0.5 | 3.7±0.9 | 3.2±1.0 | 5.2±0.7 | 48.1±1.9 |
| | | 40 | 25.8±0.5 | 2.7±0.6 | 3.7±0.8 | 4.6±0.8 | 53.5±5.8 |
| | | 50 | 23.4±1.6 | 3.3±2.8 | 2.5±0.9 | 4.6±1.8 | 45.1±29.2 |
| | | 60 | 24.2±1.1 | 1.9±0.6 | 2.6±0.4 | 3.2±0.5 | 54.4±9.5 |
| | | 70 | 24.1±0.3 | 2.4±0.9 | 2.4±0.3 | 3.4±0.8 | 45.9±11.1 |

Table 5 *(continue)*

| Method of extraction | Solvent | Concentration | Color Coordinates | | | | |
|---|---|---|---|---|---|---|---|
| | | (%) | L* | a* | b* | c* | h* |
| | Acetone | 20 | 24.4±0.3 | 2.4±0.4 | 4.3±0.3 | 4.9±0.4 | 61.2±2.2 |
| | | 30 | 24.5±1.2 | 2.8±0.2 | 5.3±0.2 | 6.0±0.2 | 62.3±1.1 |
| | | 40 | 23.9±0.1 | 1.9±0.3 | 2.9±0.3 | 3.5±0.4 | 56.7±1.8 |
| | | 50 | 24.5±1.0 | 3.0±0.6 | 3.6±0.9 | 4.6±0.7 | 49.7±9.9 |
| | | 60 | 23.6±0.3 | 2.0±0.3 | 2.0±0.3 | 2.8±0.2 | 45.8±7.9 |
| | | 70 | 23.4±0.7 | 2.7±0.7 | 1.8±0.9 | 3.4±0.2 | 33.8±19.1 |
| ABE | Ethanol | 20 | 26.2±1.0 | 0.7±1.2 | 3.1±0.5 | 3.3±0.8 | 80.4±15.5 |
| | | 30 | 27.4±1.0 | 0.6±0.3 | 4.4±1.3 | 4.5±1.2 | 80.2±8.70 |
| | | 40 | 27.2±0.3 | 0.5±0.5 | 5.8±1.3 | 5.8±1.2 | 84.7±6.40 |
| | | 50 | 27.2±1.5 | 0.6±0.3 | 4.8±3.3 | 5.5±4.4 | 72.8±15.3 |
| | | 60 | 26.1±1.2 | 0.8±0.0 | 5.8±1.4 | 12.1±4.0 | 85.8±1.80 |
| | | 70 | 26.8±0.6 | 0.6±0.2 | 5.5±2.5 | 7.8±2.3 | 81.1±15.6 |
| | Methanol | 20 | 27.0±0.0 | 0.2±0.0 | 3.1±0.1 | 3.1±0.1 | 85.7±0.2 |
| | | 30 | 27.0±0.1 | 0.3±0.1 | 3.4±0.5 | 3.4±0.6 | 85.1±0.8 |
| | | 40 | 27.4±1.0 | 0.6±0.3 | 4.4±1.3 | 4.5±1.2 | 80.2±8.7 |
| | | 50 | 27.2±1.5 | 0.6±0.3 | 4.8±3.3 | 5.5±4.4 | 72.8±15.3 |
| | | 60 | 27.4±1.0 | 0.6±0.3 | 4.4±1.3 | 4.5±1.2 | 80.2±8.7 |
| | | 70 | 27.7±0.4 | 1.5±0.2 | 4.2±1.3 | 4.4±1.2 | 69.3±7.3 |
| | | 20 | 26.0±2.1 | 1.2±0.6 | 3.0±1.3 | 3.3±1.4 | 69.5±3.6 |
| | Acetone | 30 | 26.8±1.1 | 1.0±0.6 | 5.0±4.0 | 5.1±4.0 | 77.4±6.0 |
| | | 40 | 26.4±1.7 | 0.7±1.0 | 3.9±1.9 | 4.0±2.0 | 82.8±9.2 |
| | | 50 | 27.1±0.8 | 0.3±0.2 | 6.3±1.7 | 6.3±1.7 | 86.7±2.0 |
| | | 60 | 27.6±1.2 | 1.8±0.5 | 15.3±3.6 | 15.4±3.6 | 83.4±0.3 |
| | | 70 | 27.1±0.4 | 1.0±1.3 | 3.9±0.6 | 4.1±1.0 | 77.1±13.2 |

Based on Table 4 and Table 5, all samples (mesocarp and exocarp) indicated that natural colourant extracted had lighter hue (yellow and red) due to low a* and b* values. In comparison, natural colourant extracted from UAE and ABE did not have a remarkable increase in a* and b* value throughout the experiment. The emergence of plant pigments and phenolic compounds (colourants) was closely related to colorful substances consisting of biochromes, which absorb or reflect light of varying wavelengths (Boonsong et al., 2012).

For all mesocarp samples extracted from ABE, hue angle (h*) was slightly lower (33.8 till 62.8) than UAE (69.3 till 86.7) which indicated that the longer the extraction time, the darker the color of extract. As previously reported, ABE method took about 24 hours to complete the extraction process compared with UAE which required only one hour. Overall, the color difference (mesocarp and exocarp) using UAE method was smaller than ABE method. The color of the mesocarp and exocarp from both UAE and ABE methods was dark (low L* value) but with low intensity (less vivid).

## CONCLUSION

Two methods were performed and compared known as UAE and ABE method in maximising the extraction of natural dyes from the husk of *Cocos nucifera*. The result indicated that UAE performed better in 20% of ethanol and 20% of acetone (80% and 204.8%, respectively; in the improvement of percentage yield of colorant due to ultrasound) in mesocarp and exocarp respectively. In addition, UAE proved to be better technique due to acoustic cavitations provided by ultrasound. Moreover, UAE is a more promising method as the time taken to complete the process was only one hour compared with the ABE method which required 24 hours. Finally, the positive results on color analysis highlighted the potential of utilising *Cocos nucifera* husk as a source of natural colourant.

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