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Optimization of Natural Dye Extraction from Coconut Husk

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ABSTRACT

This work aims to optimize the extraction of natural dye from coconut husks for use in the textile industry. The optimization process was carried out using the One-Factor-at-a-Time (OFAT) approach. Before optimization, different parts of the coconut husk i.e., endocarp, exocarp, mesocarp, and a mixture of mesocarp and exocarp were screened for their potential to be used as dye. It was found that the combination of mesocarp and exocarp has the highest potential in terms of physicochemical and phytochemical properties as it resulted in the best yield of extract, pH and colour intensity, and the highest Total Flavonoid Content, Total Phenolic Content, and Total Tannin Content. The optimal parameters to extract dye from coconut husk (combination of mesocarp and exocarps) are particle size of 2 mm, sample to solvent ratio of 1:100, extraction time of 60 min, and a temperature of 100 °C. The dye extract was tested for use on cotton fabric. It was found that regardless of the mordanting method, acetic acid is the best mordant to use. It resulted in a brighter and more intense colour than alum. The usage of acetic acid as the mordant combined with the pre-mordant method was found to be the best for colour fixation of the natural dye from coconut husk on cotton fabric.

Keywords: Natural dye, Coconut husk, Extraction, Colour fixation, Mordanting

1. INTRODUCTION

In recent times, the demand for natural dyes have increased due to increasing awareness of the dangers of synthetic dyes to general health and environment. Natural dyes offer protection from harmful ultraviolet radiation and are typically safe to dispose. Natural dyes are also desirable because they have a gentler and more aesthetic pleasing hue which people may find more comfortable (Wangatia *et al.,* 2018; Adeel *et al.,* 2019).

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The use of synthetic dyes over a long period of time can cause various harmful effects on human health and the environment. To humans, synthetic dyes are teratogenic and carcinogenic, and can cause skin dermatitis, and even hyperactivity in children (Toma *et al.,* 2017). To the environment, the dyes cause land and water pollution which upset various ecosystems. The impact of synthetic dyes on aquatic ecology system is especially serious. They decrease the carbon dioxide sequestration efficiency of many water bodies, leading to more unbridled greenhouse gas and therefore worsening climate change (Goodarzian & Ekrami., 2018).

One major application of natural dyes is in the batik textile industry. The batik industry especially in Kelantan and Terengganu make use of natural dyes from plants to colour their batik. Batik produced using natural dyes are exclusive and are generally regarded as more valuable than batik produced using synthetic dyes (Mohd-Shukri, 2021).

Cocos nucifera, or more commonly known as the coconut, is a member of the Arecaceae family (palm family) (Imo *et al*., 2018). The coconut is a major crop in Malaysia with a total plantation acreage of approximately 85,000 hectares with each hectare being able to produce up to 15,000 coconuts (Tan, 2008). The demand for coconut is increasing annually and this results in an accumulation of coconut waste as coconut husk.

Coconut husk is composed of 30% fiber and 70% pith. It is also high in cellulose (41%), lignin (40%), and phenolic content (Israel *et al*., 2011). The high lignin content contributes to the elasticity and durability of coconut fiber. It also makes the coconut fiber resistant to rotting. In addition, coconut husk also contains tannin and potassium.

The coconut husk consists of the endocarp, mesocarp, and exocarp which make up about 30-35% of the whole coconut (Tan *et al.,* 2007). It is estimated that 5280 kg of dry coconut husk is produced per hectare every year which is significant and (Rodiah *et al.,* 2018) there is a dire need to find a way to manage the waste. One approach is to add value to the waste by utilizing the coconut husk as a natural dye. Coconut husk waste is cheap, non-toxic, efficient, and readily available. Thus, the use of coconut waste as a natural dye can reduce the cost of waste management and help prevent environmental pollution.

One of the most challenging aspects that must be overcome before widespread use of natural dye in textile industries become possible is the fixing of the colour onto fabric. The type of mordant and mordanting method play an important role in overcoming this problem. Therefore, this study focuses on the optimization of the extraction process of natural dye from coconut husk and its application on cotton cloth using various types of mordant and mordanting method.

2. MATERIALS AND METHODS

2.1 Chemicals

Acetone, ethanol, and methanol for analysis were purchased from Sigma Aldrich (Malaysia) Sdn. Bhd. Vinegar (acetic acid) and alum were purchased from a local store in Kangar, Perlis. Cotton fabric for dyeing was purchased from a local textile market in Kota Bharu, Kelantan.

2.2 Materials

Coconut husk was obtained from a local store in Kangar, Perlis. The coconut husks were separated into four parts: i) endocarp, ii) mesocarp, iii) exocarp and iv) a combination of mesocarp and exocarp. Each of these parts was ground using a mechanical grinder (Mill Powder Tech, Taiwan) and sieved to obtain particles that are 1.5 mm in size.

2.3 Extraction of Natural Dye

Separately, 10 g of each of the coconut husk samples was boiled in 150 mL of distilled water at a temperature of 100°C for 45 min (Antima et al., 2012). Then, the extracted dye was filtered using Whatman No. 1 filter paper and stored at 7°C (Linden, Taiwan) until further analysis

2.4 Physiochemical Analysis

2.4.1 Gravimetric Analysis Materials

The gravimetric method was used to determine the yield of the natural colorant of each extracted dye using the following equation (Sivakumar *et al.,* 2017):

$$Yield\ of\ natural\ colorant\ \% = \left(\frac{Natural\ dried\ extracts\ obtained\ (g)}{Weight\ of\ sample\ used\ (g)}\right) \times\ 100$$

2.4.2 Determination of pH

The pH of the extract was determined using a handheld pH meter (Hanna Instruments pHep H198107, Malaysia). The extracted dyes were cooled to room temperature before analysis.

2.4.3 Determination of Colour Intensity

The colour intensity of the extracted dye was measured at 700 nm (Choudhury, 2014) using a UV-VIS spectrophotometer (Thermo Spectronic Genesys 20, USA).

2.5 Phytochemical Analysis

2.5.1 Total Flavonoid Content (TFC)

The total flavonoid content of the extracted dye sample was determined using the aluminium tri-chloride ($AlCl_3$) method as described by Shourove *et al.* (2020) with some modifications. Briefly, 0.5 mL of each extract was added to 2 mL of distilled water and mixed well with 0.15 mL of 5% sodium nitrate ($NaNO_2$). After reacting for 5 min, 0.15 mL of 10% aluminium tri-chloride-6-hydrate ($AlCl_3.6H_2O$) solution was added. After another 5 min, 1 mL of 1 M sodium hydroxide (NaOH) was added. Then, the reaction solution was incubated for 15 min and the absorbance was determined at 415 nm using a UV-VIS spectrophotometer (Thermo Spectronic Genesys 20, USA). Quantification was done using quercetin as the standard and the results are expressed as milligrams of quercetin equivalent per grams of dry matter (mg QE /g).

2.5.2 Total Phenolic Content (TPC)

The total phenolic content was measured using the Folin-Ciocalteu colorimetric method (Jothi & Krish, 2017). Briefly, 0.1 mL of each extract was mixed with 0.2 mL of Folin-Ciocalteu reagent and 8 mL of distilled water, and the mixture was subsequently incubated at room temperature for 3 min. A 1 mL aliquot of 20% sodium carbonate was then added to the mixture and it was further incubated in the dark. After 30 minutes of incubation, the absorbance was measured at 765 nm using a UV-VIS spectrophotometer (Thermo Spectronic Genesys 20, USA). The total phenolics content is expressed as milligrams of gallic acid equivalent per grams of dry matter (mg GAE /g), based on the standard calibration curve.

2.5.3 Total Tannin Content (TTC)

Total tannin content was determined using the Folin-Ciocalteu colorimetric method described by Haile & Kang (2019) with slight modifications. In short, 0.1 mL of each extract was mixed with 7.5 mL of distilled water, 0.5 mL of Folin-Ciocalteu reagent, and 1 mL of 35% sodium carbonate (Na_2CO_3) solution. This solution was mixed well, diluted to 10 mL with distilled water, and then kept at room temperature for 30 min. Then, the absorbance was measured at 700 nm using a UV-VIS spectrophotometer (Thermo Spectronic Genesys 20, USA). The tannin content is expressed as milligrams of tannic acid equivalents per grams of the dye sample (mg TAE /g).

2.6 Optimization of the Dye Extraction Condition Using One-Factor-At-A Time (OFAT) Approach

Based on the results obtained from the physicochemical and phytochemical analyses described in Sections 2.4 and 2.5, a combination of mesocarp and exocarp of coconut husk was investigated using the One-Factor-At-A-Time (OFAT) approach to determine the optimum dye extraction conditions. The parameters studied are: i) particle size (0.5 to 4 mm), ii) sample-to-solvent volume ratio (1:25, 1:50, 1:75, 1:100, and 1:125), iii) extraction time (20 to 60 min), and iv) extraction temperature (60 to 100 ºC). These four parameters were varied one at a time, and the optimum parameter is used in subsequent steps of the optimization process to obtain the highest TFC, TPC, TTC and colour intensity.

2.6.1 Effect of Particle Size on the Dye Quality

The effect of particle size (0.5, 0.715, 1, 2, and 4 mm) on dye quality was determined using 150 mL of water at a constant extraction temperature of 100°C and extraction time of 45 min. The extract was cooled and filtered with Whatman No.1 filter paper prior to analysis.

2.6.2 Effect of Sample-to-Solvent Volume Ratio on the Dye Quality

Based on the results obtained in Section 2.6.1, a particle size of 2 mm was chosen and the effect of sample-to-solvent volume ratio on dye quality is investigated. Four combination of sample-to-solvent volume ration was prepared (1:25, 1:50, 1:75, 1:100, and 1:125 (g/mL)) and the extraction was carried out for 60 min at a temperature of 100°C.

2.6.3 Effect of Extraction Time on the Dye Quality

Based on the results obtained in Sections 2.6.1 and 2.6.2, the particle size of 2 mm and sample-to-solvent ratio of 1:100 g/mL were chosen and the effects of extraction time on phenolic yield (TFC, TPC, TTC) and the dye colour intensity is investigated. The extraction process was carried out at 100 °C for 20, 30, 40, 50, or 60 min.

2.6.4 Effect of Extraction Temperature on the Dye Quality

Based on the results obtained in Sections 2.6.1, 2.6.2 and 2.6.3, the particle size of 2 mm, sample-to-solvent ratio of 1:100 g/mL, and 60 min extraction time were used and the effects of extraction temperature on the phenolic yield (TFC, TPC, TTC) and dye colour intensity were investigated. The extraction temperature was set at 60, 70, 80, 90, or 100 °C (Antony & Farid, 2022).

2.7 Dyeing Method

The optimum conditions for extraction of the dyes are: particle size of 2 mm, sample-to-solvent ratio of 1:100, extraction time of 60 min, and a temperature of 100 °C. The next step is to study the fixation of the extracted dye on cotton fabric and therefore we look into the effect of mordant types and mordanting methods. Three dyeing methods: i) pre-mordanting, ii) simultaneous mordanting and iii) post-mordanting; and two different types of mordants: (i) 0.5% acetic acid (v/v) and ii) 0.5% alum (w/v) were investigated in this study. Cotton fabrics of size of 15 x 30 cm were subjected to a scouring pre-treatment process beforehand by soaking the fabric in 5 L of distilled water for 30 min. The fabrics were then dried at room temperature after scouring (Verenkar & Krishnana, 2020).

2.7.1 Pre-mordanting

In the pre-mordanting method, the cotton fabric was pre-soaked in a mordant solution (150 mL) for 1 hour at a temperature of 80°C before further soaking in 250 mL of dye extract for another hour at a temperature of 85°C. The fabric was then washed with cold water and left overnight to dry (Thiyagarajan *et al.,* 2016).

2.7.2 Simultaneous Mordanting

In simultaneous mordanting, the cotton fabric was soaked in a mixture of 150 mL of mordant and 250 mL of dye extract at a temperature of 85°C for 1 hour. The dyed cotton fabric was then washed with cold water and left overnight to dry (Thiyagarajan *et al.,* 2016).

2.7.3 Post-Mordanting

In the post-mordanting method, the cotton fabric was first soaked in 250 mL of dye extract at 80 °C for 1 hour followed by soaking in mordant solution (150 mL) for another hour at 85°C. The cotton fabric was then washed with cold water and left overnight to dry (Thiyagarajan *et al.,* 2016).

2.8 Statistical Analysis

All values are reported as a mean ± standard deviation. Minitab (Version 17), LLC. (2010) statistical analysis software was used to calculate the means and standard deviations for all measurements. All experiments were carried out in triplicate. The data were subjected to one-way analysis of variance (ANOVA) followed by Tukey's Test for multiple comparisons where $P<0.05$ is the threshold for significance.

3. RESULTS AND DISCUSSION

3.1 Physicochemical and Phytochemical Properties of Different Parts of Coconut Husks as Natural Colorant

Phytochemicals such as phenolics, flavonoids, and tannins are extracted from natural materials and are polyphenolic compounds that can be used for the dyeing of textiles (Salem *et al.,* 2020; Sofyan *et al.,* 2021). Polyphenolic dyes can have various colours based on the phytochemical compounds present and its concentration. For example, dye extract from the pods of *Archidendron jiringa* contain high amounts of tannins that give a bright yellow colour (Sofyan *et al.,* 2021).

The physicochemical and phytochemical properties of different parts of coconut husk (i.e., endocarp, exocarp, mesocarp, and a combination of mesocarp and exocarp) are shown in Figures 1 and 2, respectively. As shown in Figure 1, the combination of mesocarp and exocarp of coconut husk resulted in the highest dye yield (66.67±0.79%) and colour intensity (0.50±0.07 nm) and they are statistically different at $p<0.05$ for both parameters from every other samples (Figure 1). Additionally, the pH values for the exocarp (pH 7.4±0.05) and the combination of mesocarp and exocarp (pH 7.2±0.4) were significantly higher at $p<0.05$ than that for the endocarp and the mesocarp (pH 6.5 and pH 6.7, respectively).

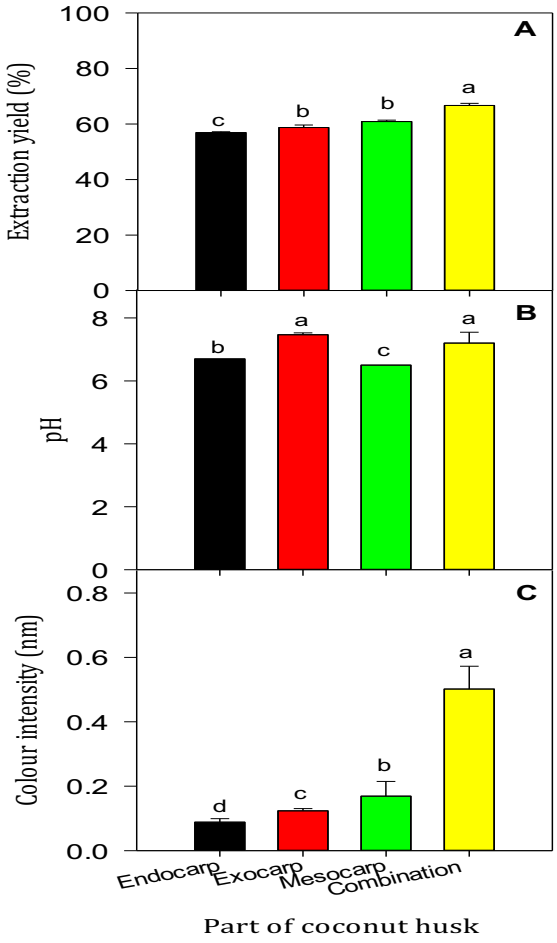

Figure 1. Physicochemical properties of different parts of coconut husk where ■ = endocarp, ■ =exocarp, ■= mesocarp and ■= combination of mesocarp and exocarp. Data are mean ± standard deviation (N=3). Values with different letters are significantly different at $p<0.05$ (One-way ANOVA followed by post-hoc Tukey's Test)

Also, among all the samples studied, the combination of mesocarp and exocarp resulted in the highest values of TPC (1.73±1.99 mg GAE/g; $p<0.05$), TFC (2.24±0.04 mg QE/g; $p<0.05$) and TTC (0.23±0.01 mg TE/g) and they were significantly different from every other sample (Figure 2). In a study conducted by Rodiah *et al.* (2018) which assessed the polyphenolic contents of mesocarp and endocarp of coconuts, the authors found that the mesocarp had a TPC value (32.24 mg GAE/g) and it was significantly higher than the TPC value of the exocarp (8.63 mg GAE/g). However, the authors did not assess the TPC values of the combination of mesocarp and exocarp.

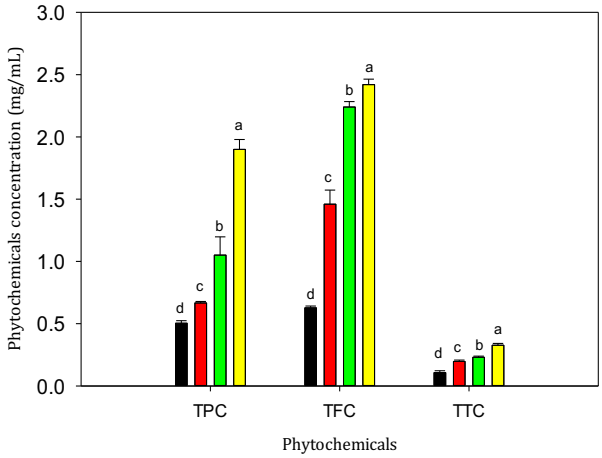

Figure 2. Phytochemical properties of different parts of coconut husk where ■ = endocarp, ■ =exocarp, ■ =mesocarp and ■ =combination of mesocarp and exocarp. Data are mean ± standard deviation (N=3). Values with different letters are significantly different at $p<0.05$ (One-way ANOVA followed by post-hoc Tukey's Test)

The exocarp is a thin, yellow-brown, watertight outer skin of the coconut husk while mesocarp is a thick, fibrous middle layer of the coconut husk. In terms of practicality, it is tedious to physically separate the exocarp and mesocarp. Among the parts of coconut assessed in this study, the combination of mesocarp and exocarp has shown a favourable result in its physicochemical and phytochemical assessments. Therefore, the combination of mesocarp and exocarp was chosen for further investigation in the subsequent optimization study.

3.2 Optimization of the Dye Extraction Condition Using One-Factor-At-A Time (OFAT) Approach

3.2.1 Effect of Particle Size on the Dye Quality Post-mordanting

The coconut husk was ground and sieved into five (5) different particle sizes in the range of 0.5 – 4 mm. In the optimization study, only phytochemical (TFC, TPC, TTC) and colour intensity analyses were carried out. The TTC of the sample was found to increase proportionately with particle size while the TFC, TPC and colour intensity of the samples were found to increase with particle size up to 2 mm and decrease thereafter (Table 1).

Table 1 Effect of particle size on the dye quality

Particle Size (mm)	Total Flavanoid Content (mg QE/g)	Total Phenolic Content (mg GAE/g)	Total Tannin Content (mg TE/g)	Color Intensity (nm)
0.5	2.51 ± 0.18^{e}	0.49 ± 0.10^{e}	0.12 ± 0.08^{e}	0.18 ± 0.06^{e}
0.715	2.63 ± 0.14^{d}	0.63 ± 0.12^{d}	0.22 ± 0.10^{d}	0.22 ± 0.08^{d}
1	3.12 ± 0.12^{c}	1.60 ± 0.14^{b}	0.31 ± 0.12^{c}	0.41 ± 0.02^{b}
2	3.89 ± 0.10^{a}	2.20 ± 0.16^{a}	0.38 ± 0.14^{b}	0.50 ± 0.06^{a}
4	3.32 ± 0.11^{b}	0.94 ± 0.11^{c}	0.40 ± 0.12^{a}	0.34 ± 0.10^{c}

Values are mean ± standard deviation (N=3). Values with different letters in each column are significantly different at $p<0.05$ (One-way ANOVA followed by post-hoc Tukey's Test)

A significant decrease in TFC, TPC and colour intensity was observed when increasing the particle size from 2 mm to 4 mm. In general, a smaller particle size has higher surface area and thus, it is expected that the extraction of the phytochemicals from the samples should increase. However, it was not the case in this study. The smaller sample particle sizes of the samples were not suitable for use in the extraction process because they tend to float to the top therefore reducing the area of contact between the solvent and sample (Yeop *et al.,* 2017). A particle size of 2 mm was chosen as the best to use in subsequent analyses.

3.2.2 Effect of Sample-to-Solvent Volume Ratio on the Dye Quality

The TPC, TFC and colour intensity of the samples were found to increase with sample-to-solvent volume ratio to a maximum when a ratio of 1:100 is used and decreased thereafter at 1:125 (g/mL). The TTC value however was found to increase proportionally with sample-to-solvent volume ratio throughout (Table 2). These results are because when a low amount of solvent is used, incomplete extraction occurs due to the solvent becoming saturated. When a higher amount of solvent is used, the concentration gradient between the phytochemicals in the sample and in the solvent is also higher and thus, the phytochemicals tend to move from the sample to the solvent more. However, further increasing the sample-to-solvent ratio to 1:125 (g/mL) decreased the extraction. This is postulated to be due to a reduction in the solvent's diffusion, transfer, and solvation capacity (Soedirga *et al.,* 2020). The optimum sample-to-solvent ratio to extract the dye from coconut husk is 1:100 (g/mL), as it resulted in the highest concentration of TPC, TFC, and colour intensity; and a considerable amount of TTC.

Table 2 Effect of sample to solvent volume ratio on dye quality

Volume ratio (mL)	Total Flavanoid Content (mg QE/g)	Total Phenolic Content (mg GAE/g)	Total Tannin (mg TE/g)	Color Intensity (nm)
1:25	2.12 ± 0.000^{e}	2.13 ± 0.08^{c}	0.11 ± 0.08^{e}	0.32 ± 0.02^{e}
1:50	2.35 ± 0.006^{d}	2.04 ± 0.11^{e}	0.20 ± 0.11^{d}	0.43 ± 0.06^{d}
1:75	2.89 ± 0.010^{b}	2.11 ± 0.14^{d}	0.27 ± 0.14^{c}	0.49 ± 0.08^{c}
1:100	3.19 ± 0.012^{a}	2.45 ± 0.16^{a}	0.35 ± 0.16^{b}	0.55 ± 0.10^{a}
1:125	2.74 ± 0.004^{c}	2.36 ± 0.12^{b}	0.39 ± 0.19^{a}	0.53 ± 0.12^{b}

Values are mean ± standard deviation (N=3). Values with different letters in each column are significantly different at $p<0.05$ (One-way ANOVA followed by post-hoc Tukey's Test)

3.2.3 Effect of Extraction Time on the Dye Quality

The effect of extraction time on the quality of the dye extracted from coconut husk was studied using samples with particle size of 2 mm, sample-to-solvent ratio of 1:100 (g/mL). The extraction time was varied between 20 to 60 min. The TFC, TPC, TTC, and colour intensity were found to increase with extraction time (Table 3). Increasing the extraction time means that there is more time for the phytochemicals to move from the sample to the solvent and therefore the yield is increased. However, it is expected that after a certain period of time, an equilibrium will be reached between the solute in the solid matrix (plant sample) and the solute in the bulk solution (extraction solvent) (Sabarudin *et al.,* 2016).

Table 3 Effect of extraction time on the dye quality

Time (mins)	Total Flavanoid Content (mg QE/g)	Total Phenolic Content (mg GAE/g)	Total Tannin (mg TE/g)	Color Intensity (nm)
20	2.02± 0.017^{e}	1.99 ± 0.006^{e}	0.12 ± 0.006^{e}	0.22 ± 0.010^{e}
30	2.09 ±0.006^{d}	2.20 ± 0.001^{d}	0.20 ± 0.010^{d}	0.29 ± 0.010^{d}
40	2.16 ± 0.012^{c}	2.22 ± 0.080^{c}	0.34 ± 0.012^{c}	0.33 ± 0.002^{c}
50	2.24 ± 0.012^{b}	2.32 ± 0.012^{b}	0.37 ± 0.010^{b}	0.39 ± 0.001^{b}
60	2.67 ± 0.000^{a}	2.43 ± 0.012^{a}	0.44 ± 0.000^{a}	0.42 ± 0.000^{a}

Values are mean ± standard deviation (N=3). Values with different letters in each column are significantly different at $p<0.05$ (One-way ANOVA followed by post-hoc Tukey's Test

3.2.4 Effect of Extraction Temperature on the Dye Quality

The effect of extraction temperature on the quality of the dye extracted was investigated using samples with particle size of 2 mm, sample-to-solvent ratio of 1:100 (g/mL), and an extraction time of 60 min. The extraction temperature was varied from 60 to 100 °C. The results show that TPC, TTC, and colour intensity of the samples all increase proportionately with temperature, while TFC decreases with increasing temperature (Table 4).

Table 4 Effect of extraction temperature on dye quality

Temp (°C)	Total Flavanoid Content (mg QE/g)	Total Phenolic Content (mg GAE/g)	Total Tannin (mg TE/g)	Color Intensity (nm)
60	2.63 ± 0.006^{a}	1.88 ± 0.006^{e}	0.15 ± 0.010^{e}	0.21 ± 0.003^{e}
70	2.57 ± 0.000^{b}	1.98 ± 0.006^{d}	0.22 ± 0.006^{d}	0.24 ± 0.003^{d}
80	2.46 ± 0.015^{c}	2.02 ± 0.012^{c}	0.27 ± 0.006^{c}	0.28 ± 0.003^{c}
90	2.30 ± 0.012^{d}	2.17 ± 0.012^{b}	0.36 ± 0.010^{b}	0.44 ± 0.001^{b}
100	2.21 ± 0.010^{e}	2.23 ± 0.001^{a}	0.45 ± 0.001^{a}	0.54 ± 0.001^{a}

Values are mean ± standard deviation (N=3). Values with different letters in each column are significantly different at $p<0.05$ (One-way ANOVA followed by post-hoc Tukey's Test

The increased TPC, TTC, and colour intensity is postulated to be due to the improvement of mass transfer of phenolic compounds with solvent temperature. As the temperature is increased, the cell walls of the sample ruptures, and more compounds can be extracted (da Rosa *et al*., 2019). This study is also in agreement with a study conducted by Sarkar & Ghosh (2017), who reported that a raised temperature could increase the TPC extraction by raising the diffusion coefficient and solubility of the TPC compound in the extraction solvent. In addition to that, the heat from the solvent also releases cell-wall phenolics and bounded total phenolics by breaking the cellular structure (Syafiq *et al.,* 2021), resulting in increased TPC extraction.

In contrast with the positive effects of higher temperatures on TPC, TTC, and colour intensity, the TFC decreases with increased heating. Elevating the temperature above a certain level causes a degradation of the flavonoid compounds (Liyana-Pathirana & Shahidi, 2005). Based on the OFAT

results, it can be concluded that the optimum conditions to extract dye from coconut husk (combination of mesocarp and exocarp) are particle size of 2 mm, sample-to-solvent ratio of 1:100. TFC, TPC, TTC, and colour intensity that are obtained at these optimal conditions are 2.21 ± 0.010 mg QE/mL, 2.23± 0.001 mg GAE/mL, 0.45 ± 0.001 mg TE/mL and 0.54 ± 0.001 nm respectively.

3.3 Application of the extracted dye on cotton fabric

The effectiveness of the extracted dye was tested on cotton fabric by using different types of mordant (alum and acetic acid) and mordanting methods (pre-mordanting, simultaneous mordanting and post-mordanting) (Figure 3).

Regardless of the mordanting method, the usage of acetic acid results in a higher colour intensity of the dyed cotton fabric compared to alum. Acetic acid enables better fixation of the dye on cotton fabric due to the better stability of the dye-mordant complex compared to alum (alkali). Alum is made up of larger molecules compared to acetic acid, and this minimizes the reaction that occurs with fiber due to the effect of space tightness, resulting in a lower dye absorption by the cotton fabric (Hubbe *et al.,* 2019).

Figure 3. Effect of different types of mordant and mordanting method on cotton fabric

When comparing the mordanting methods used, pre-mordanting was found to result in better colour intensity while post-mordanting resulted in the least. The pre-mordanting method enables better dye fixation due to the chemical bridging between the dye and fabric by the mordant (Prabhu & Bhute, 2012). In simultaneous mordanting, the mixture of mordant and dye reacts to form a complex molecule before it is adsorbed by the cotton fabric. This complex molecule minimizes absorption by the fabric (Janani *et al*., 2014; Rápó & Tonk, 2021). In the post-mordanting method, the fabric was soaked in the dye before soaking in the mordant solution. This technique causes a negative charge build-up on the cotton fabric which repulses the mordant. It also causes the formation of an insoluble dye and mordant complex, further complicating its fixation on the cotton fabric. The low coordination between the dye and mordant results in a lower colour intensity of the dyed cotton fabric (Samanta, 2020).

4. CONCLUSION

This study demonstrates the extraction of natural dyes from coconut husk and its application to dye cotton fabric. Four different parts of coconut husk (i.e. endocarp, exocarp, mesocarp and combination of mesocarp and exocarp) were screened and it was found that the combination of mesocarp and exocarp resulted the best yield of extract, TFC, TPC, TTC, pH, and color intensity. The OFAT method was used to determine the optimum extraction conditions of the samples. It is concluded that the optimum conditions to extract dye from the mesocarp and exocarps of the coconut are: particle size of 2 mm, sample-to-solvent ratio of 1:100 (g/mL), extraction time of 60 min, and a temperature of 100 °C. When carrying out the dyeing process, acetic acid and the pre-mordanting method were found to be the best to use for the dyeing of cotton fabric.

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REFERENCES

Adeel, S., Kiran, S., & Habib, N., Hassan, A., Kamal, S., Abdul-Qayyum, M., Tariq, K. (2020). Sustainable ultrasonic dyeing of wool using coconut coir extract. *Textile Research Journal*. 90(7-8):744-756.

Antima, S., Dangwal, L. R., & Mukta, D. (2012). Dye yielding plants of the Garhwal Himalaya, India: A case study. *International Research Journal of Biological Sciences*. 14, 69-72.

Antony, A., & Farid, M. (2022). Effect of Temperatures on Polyphenols during Extraction. *Applied Sciences*. 12(4):2107.

Choudhury, A. K. R. (2014). *Principles of colour and appearance measurement; Volume 1: Object appearance, colour perception and instrumental measurement. Woodhead Publishing Series in Textiles, Number 159 (1st ed.).* Woodhead Publishing Limited, UK.

da Rosa, G. S., Vanga, S. K., Gariepy, Y., & Raghavan, V. (2019). Comparison of microwave, ultrasonic and conventional techniques for extraction of bioactive compounds from olive leaves (*Olea europaea* L.). *Innovative Food Science and Emerging Technologies.* 58, 102234.

Goodarzian, H., & Ekrami E. (2018). Wool Dyeing with Extracted Dye from Pomegranate (*Punica Granatum* L). *World Applied Science Journal*. 8(11),1387–1389.

Haile, M., & Kang, W. H. (2019). Antioxidant activity, total polyphenol, flavonoid and tannin contents of fermented green coffee beans with selected yeasts. *Fermentation*. 5(1), 1-29.

Hubbe, M. A., Chandra, R. P., Dogu, D., & Van Velzen, S. T. J. (2019). Analytical Staining of Cellulosic Materials: A Review. *Bioresource.* 14(3), 7387-7464.

Imo C., Ezaonu, C. S., Imo, N. G., & Anigbo, C.J. (2018). Proximate, Mineral and Phytochemical Composition of *Cocos nucifera* Nut. *Asian Journal of Biochemistry*. 13, 9-14.

Israel., A. U., Ogali, R. E., Akaranta, O., & Obot, I. B. (2011). Extraction and characterization of coconut (*Cocos nucifera* L.) coir dust. Songklanakarin *Journal of Science and Technology*. 33(6), 717-724.

Janani, L., Hillary, L., & Phillips, K. (2014). Mordanting methods for dyeing cotton fabrics with dye from *Albizia coriaria* plant species*. International Journal of Scientific and Research Publications.* 4(10), 1-6.

Jothi D. & Krish H. (2017). Extraction of Natural Dyes from African Marigold Flower (*Tageteserecta). Textile Colouration Autex Research Journal*. 8(2), 49–53.

Liyana-Pathirana, C., & Shahidi, F. (2005). Optimization of extraction of phenolic compounds from wheat using response surface methodology. *Food Chemistry.* 93(1), 47–56.

Mohd-Shukri, N. (2021). Personal communication, 17 Nov 2021.

Prabhu, K. H. & Bhute, A. S. (2012). Plant based natural dyes and mordants: A Review. *Journal of Natural Product and Plant Resources*. 2(6),649-664.

Rápó, E., & Tonk, S. (2021). Factors Affecting Synthetic Dye Adsorption; Desorption Studies: A Review of Results from the Last Five Years (2017–2021). *Molecules*. 26(17), 5419.

Rodiah, M. H., Nur-Asma-Fhadhila, Z., Kawasaki, N., Noor-Asiah, H., & Aziah, M. Y. (2018). Antioxidant Activity of Natural Pigment from Husk of Coconut. *Pertanika Journal of Tropical Agriculture Science*. 41(1), 441-452.

Sabarudin, N. A., Abdul-Munaim, M. S., Ab-Wahid, Z., & Gambang, K. (2016). Effect of extraction condition of natural dye pigment from Bougainville flower's bract. *Australian Journal of Basic and Applied Science.* 10(17), 172-175.

Sofyan, S., Failisnur, F., & Silfia, S. (2021). The combination of natural dyes from jengkol pod waste (*Archidendron jiringa*) and liquid waste of gambier (*Uncaria gambir Roxb*) on coloring cotton fabrics. AIP Conference Proceeding. 2349, 020061.

Salem, M. Z. M., Ibrahim, I. H. M., Ali, H. M., & Helmy, H. M. (2020). Assessment of the use of natural extracted dyes and pancreatin enzyme for dyeing of four natural textiles: HPLC analysis of phytochemicals. *Processes.* 8, 59.

Samanta, P. (2020). A Review on Application of Natural Dyes on Textile Fabrics and Its Revival Strategy. In A. K. Samanta, N. S. Awwad, & H. M. Algarni (Eds.), *Chemistry and Technology of Natural and Synthetic Dyes and Pigments (pp. 1-24). Rejeka, Crotia.* Intech Open Access Publisher.

Sarkar, A., & Ghosh, U. (2017). Effect of extraction temperature and technique on phenolic compounds and antioxidant activity of *Tamanrindus indica* seeds. *Research Journal of Recent Sciences*. 6(2), 10-15.

Shourove, J. H., Zzaman, W., Chowdhury, R. S., & Hoque, M. M. (2020). Effect of Thermal Treatment on Physicochemical Stability and Antioxidant Properties of Locally Available Underutilized Star Fruit Juice. *Asian Food Science Journal.* 14(3), 41–53.

Sivakumar, V., Vijaeeswarri, J., & Anna, J. L. (2016). Effective Natural Extraction from Different Plant Materials Using Ultrasound. *Industrial Crops and Products.* 33(1),116–122.

Soedirga, L.C., Hardoko & Prameswari, G. (2020). Determination of solvent and ratio sample-solvent towards the production of oligo-glucosamine obtained from fermented tiger shrimp (*Penaeus monodon*) shell's chitin by using precipitation method. *Food Research*. 4(6), 2163–2168.

Shafiq, F., Siddique, A., Pervez, M. N., Hassan, M. M., Naddeo, V., Cai, Y., Hou, A., Xie, K., Khan, M. Q., & Kim, I. S. (2021). Extraction of natural dye from aerial parts of Argy wormwood based on optimized Taguchi approach and functional finishing of cotton fabric. *Materials* (Basel). 4(19), 5850.

Tan, I. A. W., Ahmad, A. L., & Hameed, D. B. (2008). Preparation of activated carbon from coconut husk: Optimization study on removal of 2, 4, 6-trichlorophenol using response surface methodology. *Journal of Hazardous Materials*. 153(1-2), 709-717T.

Tan, Z. Y. (2019). Agriculture: a coconut revival. The Edge Markets (Online Magazine). Retrieved from https: https://www.theedgemarkets.com/article/agriculture-coconut-revival.

Thiyagarajan, S., Balakrishnan, K., & Tamilarasi, S. (2016). A study on extraction and application of eco-friendly natural dye extracted from flowers of *Dichrostachys cinerea* on silk fabric using of chemical mordants. *International Journal of Advanced Research*. 4(1), 1053–1059.

Toma, M., Vinatoru, M., Paniwnyk, L., & Manson, T. J. (2017). Investigation the effects of ultrasound on vegetal tissues during solvent extraction. *Ultrasonics Sonochemistry*. 8(2), 137–142.

Verenkar, N., & Krishnana, S. (2020). Extraction of natural dye from *Mammea suriga* and its application on silk and cotton fabrics. *Indian Journal of Fibre and Textile Research*. 45(4), 402-410.

Wangatia, L. M., Tedesse, K. & Moyo, S. (2018). Mango bark as a mordant dyeing cotton with natural dye. *International Journal of Textile*. 12(4),11–15.

Yeop, A., Sandanasamy, J., Pang, S.F., Abdulla, S., Yusoff, M. M. & Gimbun, J. (2017). The effect of particle size and solvent type on the gallic acid yield obtained from *Labisia pumila* by ultrasonic extraction. MATEC Web of Conferences. 111, 02008. EDP Sciences.