

Optimization of Ultrasonic Extraction Method of Natural Dyes from *Xylocarpus Moluccensis*

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Abstract—Problem statement: Traditionally the extraction of natural dyes from plant materials was done by soaking method or boiling method which requires longer extraction time, higher temperature and higher consumption of water. Extractions of colorant using boiling method normally produce a small amount of yield. Low yield of natural dyes is one of the factors that limit the use of natural dyes in comparison with synthetic dyes. Hence, there is a growing demand for developing suitable extraction technique for more efficient and effective extraction of natural textile dyes. Approach: This study demonstrated ultrasonic cleaner as a technique to extract colorant from a selected plant, i.e. *Xylocarpus moluccensis* species which can be found abundantly in peninsular Malaysia. Colorant from combination of heartwood and bark of *Xylocarpus moluccensis* was extracted at different ultrasonic volume, time and temperature. The color strength of the dye was analyzed by using Lab Scan XE UV-visible spectrophotometer. Result: Results show the highest percentage of total yield dye extract using ultrasonic cleaner was produced at optimum condition (extraction time: 80 minutes, extraction temperature: 49°C and sonic volume: medium) with 80ml of 0.1M sodium hydroxide as the solvent. The yield produced was higher than that of boiling method and thus gave higher intensity of red crude extract. Conclusion: The utilization of ultrasonic cleaner was found to have significant improvement in the extraction of natural dyes compared to traditional boiling method.

Index Terms—UV-visible spectrophotometer, ultrasonic cleaner, natural dye, colorant, anthraquinone

I. INTRODUCTION

Xylocarpus moluccensis, commonly known as *Nyireh batu* in Malay belongs to family of Meliaceae, which can be found abundantly in littoral forest of the Malaysia Peninsula, Bengal, Burma, the Andaman's, Australia, Fiji and Africa [1]. The features of the *Xylocarpus moluccensis* are 5 to 20m tall, compound leaf comprising of 2 to 3 pair of leaflets that are oblong with more pointed tip, thin and leathery (green color), the flowers are tiny white to yellowy, the fruits are green in color and round shape and bark are longitudinal fissures and hath dark grey color.

According to Giesen *et al.*, [2], *Xylocarpus moluccensis* is used widely in timber production for construction of houses and boats. Aborigine people (Mah Meri community) of Pulau Carey, Malaysia are famous for its unique wooden statues and masks carved from *Xylocarpus* heartwood. The

waste from this handcraft production is useful as a source of natural dyes for this study. Previously, Abd Razak *et al.*, [3] reported *Xylocarpus moluccensis* heartwood as one of the potential natural dyes sources. They discovered that the colorant extracted corresponds to anthraquinone compound.

Recently, there is a growing demand for developing suitable efficient extraction technique for natural dyes from plants materials. Ultrasonic approach has been widely used mainly in food industry [4]-[6] and also in textile industry [7]-[9].

Cuoco *et al.*, [8] used ultrasonic-assisted extraction based on central composite design to extract dyes originally biosynthesized by the plant. Recently, Sivakumar *et al.*, [7] found that the application of ultrasonic probe can increase the extraction of dyes from different parts of various plants resources. The reason for the improvement could be due to better leaching of natural dyes materials from plant cell wall membranes and mass transfer to solvent assisted by acoustic cavitations provided by ultrasonic. Kamel *et al.*, [9] used ultrasonic bench top cleaner bath for dyeing cloth fabric using colorant from Cochineal dye. They reported that the ultrasound approach improved dyeability as well as enhancement in the fastness properties of the dyed fabric.

In this study ultrasonic cleaner approach was used in both extraction and dyeing process. Ultrasonic cleaner has a more uniform and wider waves compared to ultrasonic probe and ultrasonic- assisted extraction based on central composite design. As a result, various pattern can be examined at the same time hence reduce time, cost and energy. Whereas in the other two latter methods, waves occur only at a limited area. Therefore, more time and exertion are required to examine each pattern which consumes more energy and cost.

II. MATERIALS AND METHOD

A. Raw Materials.

The waste of heartwood and bark of *Xylocarpus moluccensis* was collected from the craft small industry located in the village at Pulau Carey, Klang Selangor, Malaysia. The sample was then ground and stored at room temperature before use.

B. Chemical

Sodium hydroxide

C. Ultrasonic Extraction Method

Approximately 7g of bark and 1g of heartwood was weighed and placed in the bottle. 80mL of 0.1M sodium hydroxide was added. The bottle was then placed into the ultrasonic cleaning for extraction process. Parameter optimized were ultrasonic volume (low, medium and high),

Manuscript received September 19, 2012, revised November 30, 2012.

This work was supported by research funding from Universiti Teknologi MARA (UiTM).

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extraction time (20, 40, 60, 80 and 100 minutes) and temperature was read after the extraction.

All extracts were filtered by using vacuum pump and dried using freeze dried. After sample was dried, the weight was measured and recorded.

$$\% \text{Yield of total extract} = \frac{\text{Total extract obtained (g)}}{\text{Amount of sample used (g)}} \times 100$$

D. Boiling Method

Approximately 7g of bark and 1g of heartwood was weighted and placed in the bottle. 80mL of 0.1M sodium hydroxide was added. The sample was boiled using for 80 min at atmospheric pressure. All extracts were filtered by using vacuum pump and dried using freeze dried. After sample was dried, the weight was measured and recorded.

E. Chemical Analysis

The reflectance/ transmittance and color strength of the extract sample was measured using Lab Scan XE UV-visible spectrophotometer at the wavelength range of 400-700nm.

F. Color Measurement

Color of extract was measured using Lab Scan XE UV-visible spectrophotometer based on CIE system (International Commission on Illumination). The reference illuminant was D65 (standard daylight), expressed as L^* , a^* and b^* which representing lightness (L^*), redness ($+a^*$), greenness ($-a^*$), yellowness ($+b^*$), and blueness ($-b^*$). From the L^* , a^* and b^* coordinates, chroma (C^*) and hue angle (h°) values were calculated by using the following equation:

$$G. \quad C^* = \sqrt{(a^{*2} + b^{*2})} \text{ and } h^\circ = \tan^{-1}(b^*/a^*)$$

Chroma measures the intensity or saturation of the colorant while hue angle (h°) is expressed on 360° grid to show the tonality of the color. Hue angle (h°) represents red at 0° (or 360°), blue at 270° (or -90°), yellow at 90° and green color at 180° (or -80°).

III. RESULTS

Extraction of natural dyes from combination of bark and heartwood of *Xylocarpus moluccensis* was done using 0.1M sodium hydroxide as a solvent. A preliminary study consisting of various experiments were carried out in order to determine the role of extraction time and ultrasonic volume involved in the ultrasonic-assistance extraction of *Xylocarpus moluccensis*. The percentage yield of dye extract of *Xylocarpus moluccensis* obtained is tabulated in Table I and the plot is shown in Fig. 1.

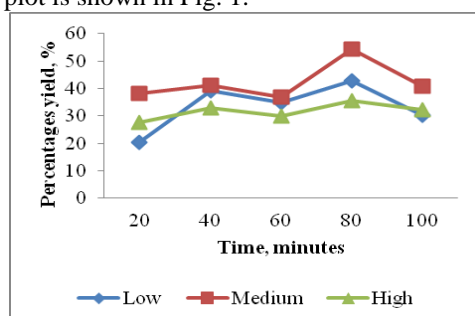


Fig. 1. Effect of extraction time and sonic volume on % yield

As shown in Fig. 1, the percentage yield of dye extract of *Xylocarpus moluccensis* increased with the increasing of the extraction time and reached maximum at 80 minutes. A further increase in time resulted in a declined plot.

Ultrasonic volume is an important parameter for the ultrasonic-assistance extraction as the effectiveness of the dye extract depends on it. Results in Table I show that the highest percentage yield of dye extract was produced at medium sonic volume.

The extraction using ultrasonic cleaner at optimum condition was then compared to the classical boiling method. The results in Fig. 2 show that the extraction yield obtained by ultrasonic extraction method increased by 54.22% as compared to boiling method.

The analytical measurement of aqueous plant extract is important in order to control the quality and standardization of the natural colorant prior to further application such as textile dyeing. In this study, the measurement of L^* , a^* , b^* as well as h° and chroma C^* values was used to evaluate the color of crude dyes from combination of bark and heartwood of *Xylocarpus moluccensis* obtained by both ultrasonic and boiling methods.

TABLE I: PERCENTAGE YIELD OF TOTAL EXTRACT USING ULTRASONIC

Time, minutes	Percentage yield of total extract, %		
	Low frequency	Medium frequency	High frequency
20 minutes	20.27	38.1	27.72
40 minutes	39.31	41.22	32.86
60 minutes	34.81	36.83	30.04
80 minutes	42.96	54.22	35.69
100 minutes	30.38	40.71	32.38

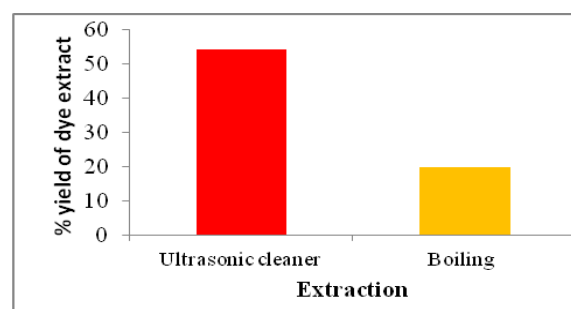


Fig. 2. Comparison of % yield of dye extract obtained using ultrasonic and boiling methods in 80 minutes time.

Table II shows the values of L^* , a^* , b^* obtained from measurement using Lab Scan XE UV-spectrophotometer based on CIE system. Hue angle, h° and chroma, C^* values were calculated using the formula mentioned earlier.

The value of a^* which indicates the redness effect for crude dye extract was significantly higher by using ultrasonic extraction compared to boiling method. The values of a^* and b^* contribute to h° value of 45° which show that the color of crude dyes extract obtained by ultrasonic extraction method was red-orange. While, the color of crude dye extract obtained by boiling method was more to yellowish orange as represent by hue angle of 55.4°

TABLE II: COLOR MEASUREMENT OF CRUDE DYE EXTRACTS OBTAINED BY USING ULTRASONIC AND BOILING METHODS.

Method	L^*	a^*	b^*	C^*	h°
Ultrasonic	0.18	0.31	0.31	0.44	45.0
Boiling	0.17	0.2	0.29	0.35	55.4

L^* =lightness, a^* =redness, $-a^*$ =greenness, b^* =yellowness, $-b^*$ =blueness, h° =hue angle, C^* =chroma

The intensity or saturation of colorant is represented by chroma, C^* value. The C^* value of 0.44 indicates that dye extract obtained by ultrasonic cleaner extraction was more saturated compared to extract obtained by boiling method (0.35).

IV. CONCLUSION

The use of ultrasonic extraction method was found to have significant improvement in the percentage yield, color intensity and redness color of crude dye extract compared to traditional boiling method. In this study, the operating condition for the ultrasonic extraction of natural dye from *Xylocarpus moluccensis* was optimized at 80 minutes with medium sonic volume and temperature of 49°C.

ACKNOWLEDGEMENT

We wish to express our gratitude to the Faculty of Applied Sciences, Universiti Teknologi MARA Shah Alam for the support.

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