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Research

Optimization of Natural Dyes Extraction from *Mangifera indica* (Gadung) Leaves

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ABSTRACT

In recent years, due to the massive pollution spread, the switchover to the eco-friendly process is being considered. Particularly, in textile industries, the wastewater containing dangerous substances damages the condition of the rivers directing to the use of natural dyes. The existing traditional industries often use natural sources with lack of study about the method and process condition to gain the best quality and the highest quantity of colorants. The common natural dye source, *Mangifera indica* leaves, particularly from Gadung species, a common type of *Mangifera indica* in Indonesia, is used in this study using reflux method combined with methanol and water as its solvent separately. The study is a modification from the preceding process, which includes tannin and fatty acids in the final product. Spectral analysis indicated that the best time for extraction using water, which produces pale yellow color is 60 minutes, and the best time for extraction using methanol, which produces green color is 360 minutes.

Keywords: Extraction; Natural dyes; Gadung; *Mangifera indica* ; Pollution; Reflux; Methanol; Water

INTRODUCTION

Commonly, textile industries are using synthetic dyes instead of natural dyes due to its easier application on fabrics [1]. This, however, leads to destructible effects on the body of water, because the usage of synthetic dyes mostly needs a particular type of mordant, which contains chemical substances like heavy metals that affects the quality of its wastewater [2]. Despite being treated before disposal, there is a value limit for those metals that are allowed to be disposed of that can accumulate on the body of water [3]. Therefore, the textile industries are interested to use natural sources for dyeing their fabrics [4]. The traditional textile industries have been using those natural sources, yet with a lack of research on the optimization [5]. Owing to this, research is conducted to burgeon the textile industries' productivity [1].

One of the sources of natural dyes is *Mangifera indica* leaves. In Indonesia, there are many types of *Mangifera indica* ; this research particularly studies Gadung species, which is common in Indonesia. It produces pale yellow color for water as its solvent and green color for methanol as its solvent. Many studies had been conducted with a different type of *Mangifera indica* . Shinde SS et al. studied Kesar and Hapoos species in the effects of solvent, particle size, and temperature upon the extract percentage. Methanol, ethanol, and acetone were used as its solvent, also, a varied range of temperature and size of particles. The results showed that methanol is the best solvent for extraction. Also, the extract percentage increased by increasing temperature and reducing the size of particles. The extract percentage using Hapoos species leaves gave 48.66% w/w and yield of 2.43% w/w, while Kesar species leaves gave extract percentage of 34.95% w/w and yield of 1.75% w/w [6]. On the other hand, the extraction upon Rajapuri leaves conducted by Shinde SS et al. gave extract percentage of 60.71% w/w and yield of 3.035% w/w [7]. These studies indirectly claim that species take part in determining the amount of mangiferin extracted.

Despite those previous studies, no studies are observing in detail about the fluctuation of the dyes extracted; also, all of those studies were using Soxhlet method instead of reflux method. This research tried to optimize the extraction time with consideration of the closeness to the real industries' circumstances which does not include tannins and fatty acids removal.

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Reflux method was used instead of Soxhlet because of its resemblance between laboratory equipment and the traditional textiles industries equipment-just boiling the materials with its solvent on a fixed ratio. This method is combined with two common solvents, methanol and water, which are the best solvent [6] and practical solvent, respectively.

MATERIALS AND METHODS

Materials

Mangifera indica leaves were used specifically Gadung species, which were gathered directly from the trees planted in Surabaya, Indonesia. The leaves were washed and sun-dried for about 4-5 days and ground using kitchen blender until the powder was attained. The powder was then screened to get 40/70 mesh particle size and extracted using distilled water and methanol 95% (technical grade, purchased from Brataco Chemika, Jakarta, Indonesia) as solvents.

Methods

The ratio of materials to solvent 1:10 was used in case of each variable. Powder from the leaves was extracted using reflux set equipment and maintained by TP-101 digital temperature indicator at 100°C on the three-neck rounded flask for distilled water and 64°C for methanol. Samples were collected at 30 minutes or 1 hour interval depending on the extraction duration, filtered using filter paper (Whatman Grade 40), and the absorbances were measured using HP-8453 Double Beam UV-Vis Spectrophotometer at a wavelength of 361 nm. Time variation was employed in three ways i.e. 2, 4, and 6 hours. Moreover, LC-HRMS analysis was done using Thermo Scientific™-Q Exactive™ as the high-resolution mass spectrometer. Two solvents of 0.1% formic acid in water and 0.1% formic acid in acetonitrile were used along with Hypersil GOLD aQ 50 × 1 mm × 1.9 u particle size analytical column. Throughout the analysis process, column oven is maintained at 30°C.

RESULTS AND DISCUSSION

In this study, there is a modification process instead of using old process for extracting natural dyes from *Mangifera indica* s leaves by isolating mangiferin in dried condition that is common in the past research to support the big industries with their powder colorants production. In contrast this process directly gets the natural dye in aqueous condition and with appropriate mordant that can be applied on fabrics. In the preceding process (Figure 1), powder from leaves has to be extracted with petroleum ether to remove its fatty acids and acetone to remove its tannins, then can be extracted using ethanol or methanol as the recent research suggests. This process is applicable to big dyes industry in which the goal is to export the natural dyes powder as a product. However, this research was conducted to help traditional textile industries which lack the optimization process; therefore, with these research results, they can directly produce their colorants and reduce the usage of colorants powder which will be more friendly economically for a long term.

In contrast to the traditional industries which mostly use boiling process with water, this experiment tried to resemble that condition by using reflux method. This method allows the materials to be mixed with the solvent directly. Moreover, distilled water was used because of the practicality as the traditional industries do and methanol for the comparison. So much for the aim, this study measures the highest point for the time of extraction for optimizing the process. This aim can be achieved by using spectral analysis that shows the highest absorbance due to the variation in time of extraction. The results below show the exact value of the absorbance point.

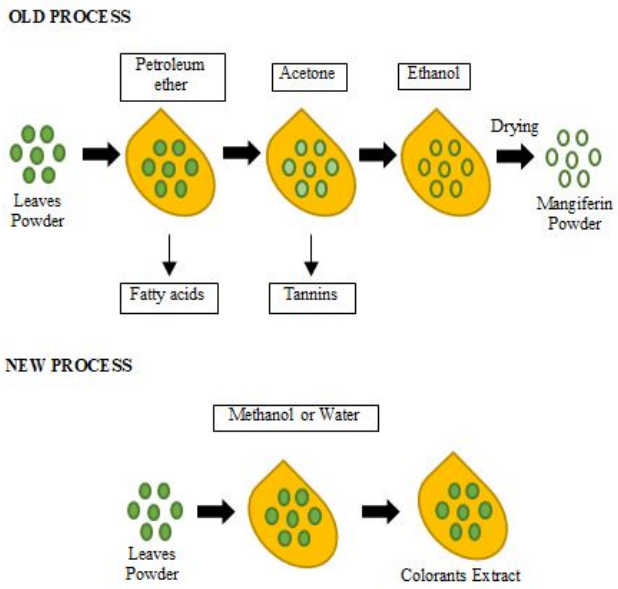

Figure 1: Old and New Process Comparison

Spectral Analysis

The absorbance data of *Mangifera indica* leaves extraction for each solvent were presented in (Figures 2 and 3) respectively. Same wavelength) was used in case of both solvents for spectral processing due to the highest absorbance from wavelength seeking process.

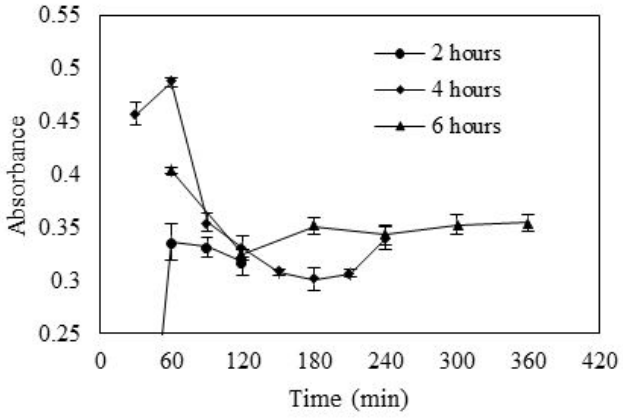

Figure 2: Water Solvent Absorbance vs Time (min) Relation

Reflux method produces high absorbance due as its materials mixed with the solvent face high temperature. The reasons for high temperature excellence are increased kinetic energy in the

solvent and the lower solvent's viscosity that eases the diffusional process through the leaves' matrix. Also, the higher intermolecular forces within the solvent that causes the higher local temperature in the leaves' matrix increases the mangiferin's solubility [6]. Based on this evidence, it was observed that the maximum time for *Mangifera indica* extraction using the reflux method and water as its solvent is 60 minutes.

Despite the optimum time for the extraction process, each variable indicates degradation through its absorbance. These results match with the previous studies about degradation of mangiferin [8]. Degradation happens since the process running for 60 minutes. In this study, distilled water was used for the extraction process. The preceding experiments indicated that using pH 7 will affect the thermal stability of mangiferin to be less stable. Also, it indicates that the degradation happens after mangiferin being observed at 100°C using pH 7, therefore, these results match with the preceding experiment.

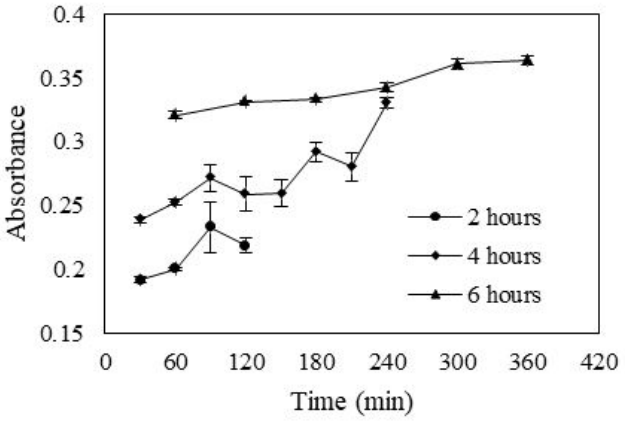

Figure 3: Methanol Solvent Absorbance vs Time (min) Relation

The absorbance value using methanol as a solvent is not as high as using water (Figure 3). The maximum absorbance is 0.3639 in 360 minutes. However, this result cannot directly be compared to the water solvent's result, because of the different colors produced by each solvent as described in the following explanation.

Color differences

In addition to the efficiencies of water solvent that has been explained before, the usage of water in extracting natural dyes also produces pale yellow color (Figure 4a). However, methanol produces the green color (Figure 4b). This phenomenon happened because of the absence of several steps for extracting mangiferin-the chemical substance that produces pale yellow color on *Mangifera indica* as described by Bhatia VK et al. [9]. This research is intended for users to directly color their fabrics instead of isolation of mangiferin powder, which later can be used for the coloring process. Therefore, the steps for removing tannin and fats are not considered. Because the solubility of tannin and fats is extremely low in water, only mangiferin was extracted because of its polarity towards water.

However, in methanol solvent, because of its solubility towards chlorophyll, it leads to the extract having green color instead of pale yellow. Still, the methanolic extract contains mangiferin. This claim is supported by the LC-HRMS analysis (Figure 5) that produced m/z value of 423.09 which match with the mangiferin mass spectrometry data [10].

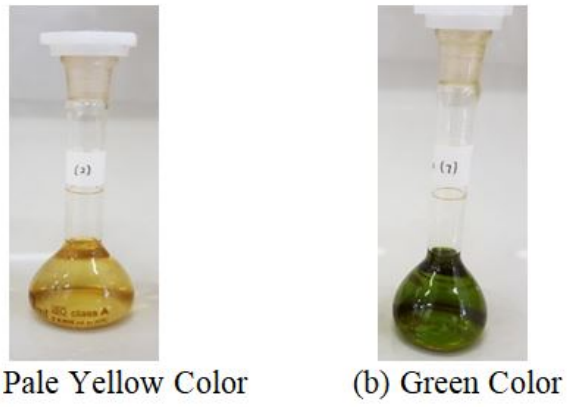

Figure 4: Color differences

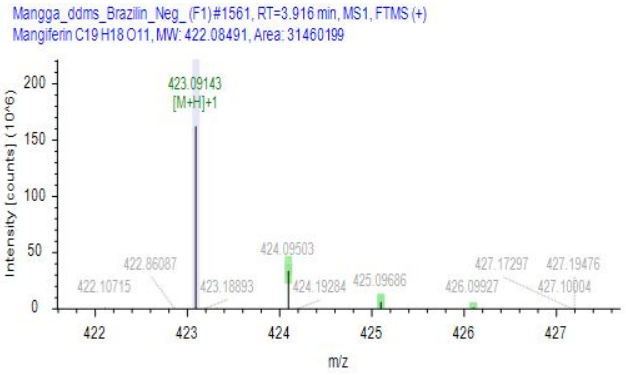

Figure 5: LC-HRMS Chromatogram on Methanolic Extract of *Mangifera indica* (Gadung) Leaves

CONCLUSION

The best time for extracting natural dye from *Mangifera indica* (Gadung) leaves using water as a solvent is 60 minutes and using methanol as the solvent is 360 minutes. This is due to the degradation after 60 minutes of extraction using water and constant absorbance after 360 minutes of extraction using methanol. After 360 minutes of extraction, any results of degradation will have no impact since textile industries work mostly 8 hours a day with the initial preparation that shortens their work time to approximately 6 hours. Direct extraction of powder from *Mangifera indica* leaves using methanol will produce green color because it dominantly contains chlorophyll instead of mangiferin substance.

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AUTHOR CONTRIBUTIONS

Hadi RP and Valentina JJ contributed equally under the supervision of Sutrisna PD and Priyantini HR.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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