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

Ronaldo Pangestu Hadi (ronaldopangestu21@gmail.com)
Universitas Surabaya https://orcid.org/0000-0001-7085-2426

Jessicalina Joscelind Valentina
Universitas Surabaya

Research

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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 dangers the condition of the rivers directing to the use of natural dyes. The existing traditional industries often use natural sources with lacks of study about the method and process condition to gain the best quality and the highest quantity of colorants. This study aim is to find the time for the extraction using common materials and simple method resembling the real situation. The common natural dye source, *Mangifera indica* leaves, particularly from Gadung species – a common type of *Mangifera indica* in Indonesia – is on this study using reflux method combined with methanol and water as its solvent separately. The steps used 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 min, and the best time for extraction using methanol, which produces green color is 360 min. Therefore, the next application of boiling the materials with water can be reduced to the written times above.

1. Introduction

Commonly, textile industries are using synthetic dyes instead of natural dyes due to its easier application on fabrics. 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. Despite it will be treated before disposal, there is a value limit for those metals that allowed to be disposed of that can accumulate on the body of water. Therefore, the textile industries interested to use natural sources for dyeing their fabrics. The traditional textile industries have been using those natural sources, yet with a lack of research on the optimization. Owing to this, research conducted to burgeon the textile industries' productivity.

One of the sources of natural dyes is *Mangifera indica* leaves. In Indonesia, there are many types of *Mangifera indica*; this research particularly studying 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 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) [1]. On the other hand, the extraction upon Rajapuri leaves conducted by Shinde gave extract percentage of 60.71%(w/w) and yield of 3.035%(w/w) [2]. 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 time extraction with consideration of the closeness to the real industries' circumstances which does not include tannins and fatty acids removal.

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 combined with two common solvents, methanol and water, which are the best solvent [1] and practical solvent, respectively.

2. Materials And Methods

2.1. Materials

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

2.2. Methods

Each variable using ratio materials to solvent 1:10. Leaves powder were 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 taken each 30 min or 1 h depends on the extraction duration, filtered using filter paper, and the absorbances measured using HP-8453 Double Beam UV-Vis Spectrophotometer using a wavelength of 361 nm. Time variation employed in three kinds: 2, 4, and 6 h. Moreover, LC-HRMS analysis 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 are used along with Hypersil GOLD aQ 50 × 1 mm x 1.9 u particle size analytical column. Through the analysis process, column oven is maintained at 30˚C.

3. 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, this process directly get the natural dye in aqueous condition and with appropriate mordant can be applied on fabrics. In the preceding process (Fig. 1), leaves powder 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 if the goal is big dyes industry which one of its products is exporting the natural dyes powder as a product. However, this research was conducted to help traditional textile industries which lack with the optimization process.

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

3.1. Spectral Analysis

The absorbance data of Mangifera indica leaves extraction for each solvent presented in Fig. 2 and Fig. 3. Both solvents are using the same wavelength (λ = 361 nm) for spectral processing due to the highest absorbance from wavelength seeking process.

Reflux method produces high absorbance due to its material mixed with the solvent and those face the high temperature. The reasons for high-temperature excellence is because of the increased of 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 that increases the mangiferin's solubility [1]. Based on this evidence, it was observed that the maximum time for *Mangifera indica* extraction using the reflux method and water for its solvent would happen in 60 min.

Despite the optimum time for the extraction process, each variable indicates degradation through its absorbation. This results match with the previous studies about degradation of mangiferin [3]. Degradation happens since the process running for 60 min. In this study, distilled water was used for the extraction process. The preceding experiment indicates that by using pH 7 wil affects the thermal stability of mangiferin to be less stabile. Also, it indicates that the degradation happens after mangiferin being observed in 100˚C using pH 7, therefore, this results match with the preceding experiment.

Figure 3 explains that the absorbance value using methanol as a solvent is not as high as using water. The maximum absorbance is 0.3639 in 360 min. 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.

3.2. Color Differences

In addition to the efficiencies water solvent that has been explained before, the usage of water in extracting natural dyes also produces pale yellow color as figured in Fig. 4(a) below. However, methanol produces the green color which figured in Fig. 4(b) 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 in Bhatia's research [4]. This research intended to direct users to 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 the water, only mangiferin 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, in the methanolic extract, it contains mangiferin. This claim is supported by the LC-HRMS analysis as figured in Fig. 5. that produce m/z value of 423.09 which match with the mangiferin mass spectrometry data [5].

4. Conclusions

The best time for extracting natural dye from *Mangifera indica* (Gadung) leaves using water as a solvent is 60 min and using methanol as the solvent is 360 min. Directly extract *Mangifera indica* leaves to powder using methanol will produce green color because it dominantly contains chlorophyll instead of mangiferin substance.

Declarations

Availability of data and materials

All data generated or analyzed during this study are available.

Competing interests

There is no competing interests among the authors toward this work.

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Authors' contributions

RPH and JJV did the whole research and wrote this article. All authors read and approved the final manuscript.

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Figures

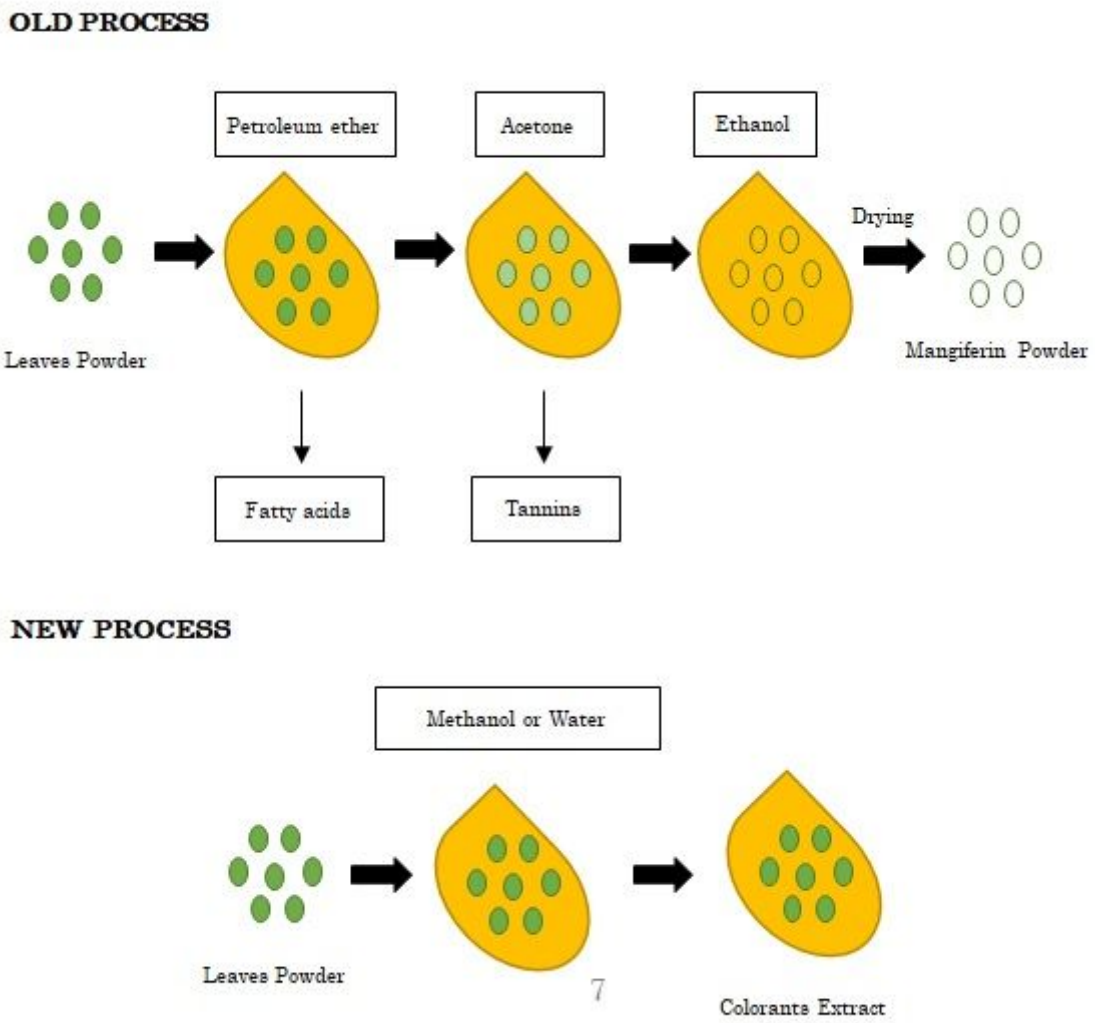

Figure 1

Old and New Process Comparation

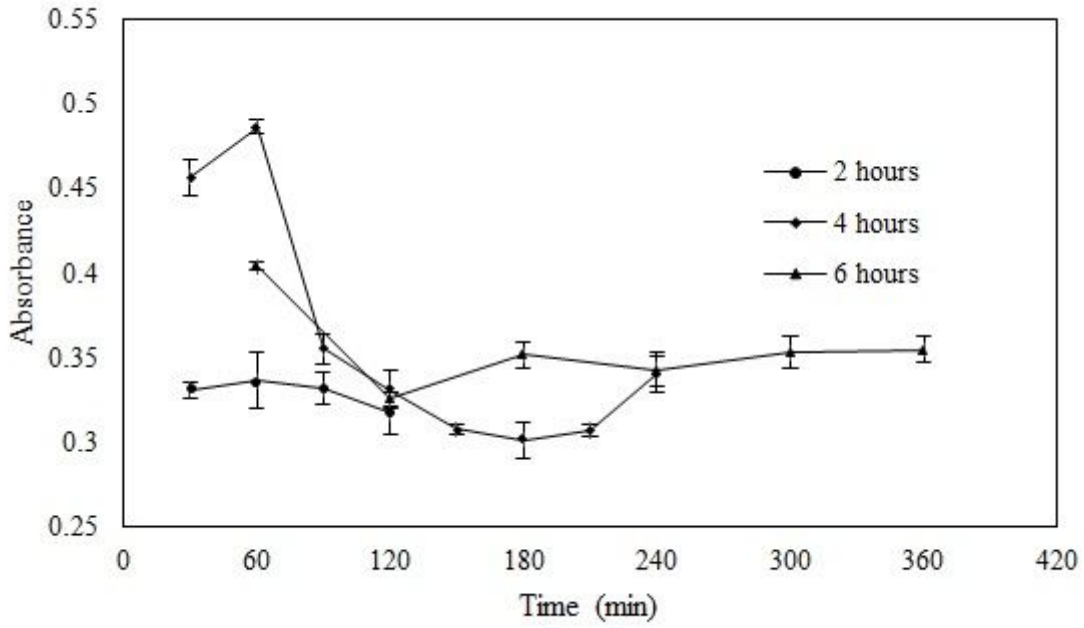

Figure 2

Water Solvent Absorbance vs Time (min) Relation

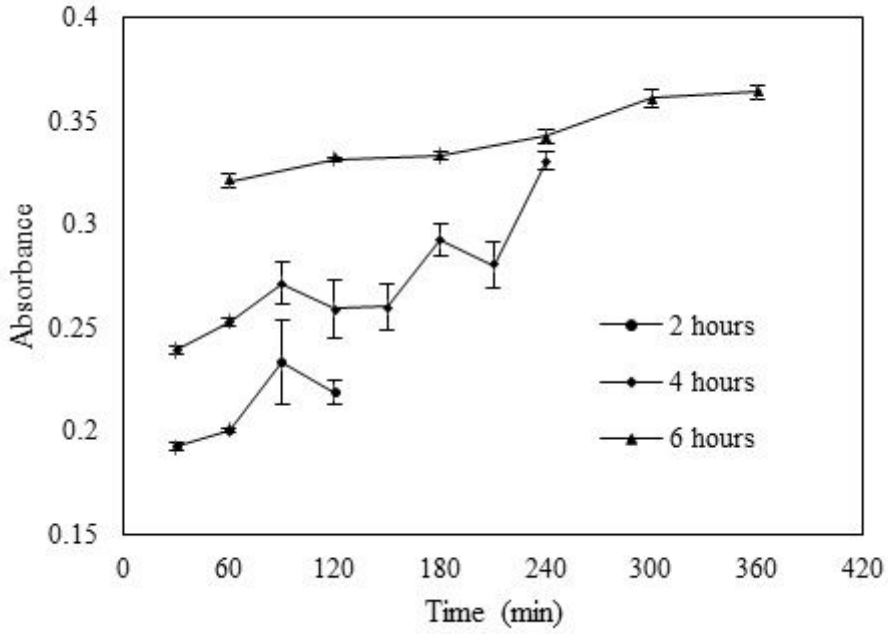

Figure 3

Methanol Solvent Absorbance vs Time (min) Relation Figure 3 explains that the absorbance value using methanol as a solvent is not as high as using water. The maximum absorbance is 0.3639 in 360 min.

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.

(a) Pale Yellow Color (b) Green Color

Figure 4

Color Differences

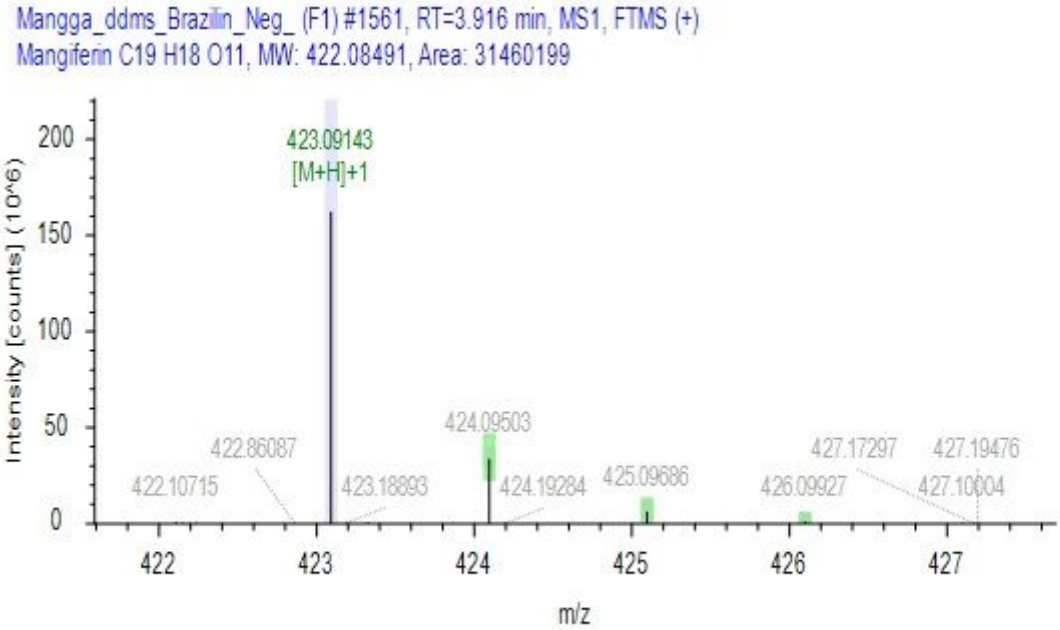

Figure 5

LC-HRMS Chromatogram on Methanolic Extract of Mangifera indica (Gadung) Leaves

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- Data.xlsx
- Data.xlsx