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The Effects of Material to Solvent Ratio on the Performances of Natural Dyes Extraction

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Abstract. The utilization of natural dyes from natural resources in fashion industry is preferred than synthetic dyes as the natural dyes is considered as more environmentally friendly than synthetic dyes. In addition, dyes from natural resources, such as plant, are renewable resources that can be sustainably used. In recent years dye from Mangifera has been used widely as sources for yellow color in fashion industry. However, there are still more studies need to investigate the effects of solvent to material ratio during extraction process to attain a standard color that can be applied in industry. Thus, this study focused on the extraction process of dye from mangifera leaf to optimize and provide recommendation about the best solvent and the ratio of solvent to material used in the dyeing process. Experimental results showed that the best solvent for Mangifera indica extraction process is methanol. In addition, the best material to solvent ratio to obtain maximum yield was 1 : 7.

Keywords: Natural dyes, extraction, solvent, ratio

INTRODUCTION

Dyes are useful in various fields, including food and beverages, textiles, cosmetics, and household materials. The usage of color could add aesthetic value to any product [1]. Based on the sources, textile dyes could be classified into two categories: natural dyes, which are generated from plants or animals, and synthetic dyes, which are produced from chemical reaction with aromatic hydrocarbon derivatives (benzene, naphthalene, and anthracene) as the raw materials [1-2]. Excess usage of synthetic dyes could damage the environment due to its toxic waste that degraded into carcinogenic compounds [3-4]. Thus, it would also harm the other living creatures nearby. Therefore, natural dye is considered as an alternative because it is less toxic, renewable, biodegradable, and environmentally friendly [5]. Furthermore, mordant substitution by the natural dye eliminates the heavy metal content in the waste. Common technique used for obtaining specific substances from natural plants is extraction [6]. Extraction for dyes can be conducted by using either maceration, reflux or soxhlet extractions.

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The benign utilization of natural dyes instead of synthesized ones, as mentioned earlier increased the demand for natural coloring agents. In Indonesia, batik artisans and the yarn industries commonly use natural dyes conventionally. However, they pay less attention to the method, process condition, and materials-solvents usage. From the previous study by Sutrisna et al. (2020) [7], the optimum method and condition for sappan bark extraction was a Soxhlet with ethanol as the solvent which produced 4.96% of yield and 0.69 of absorbance. As for the mango leaves extraction, the reflux method with water as the solvent was found to be the best way to extract yellow color with 2.52% yield and 0.48 absorbance, and the Soxhlet method with methanol as the solvent produced green color with 6.00% and 0.69 of yield and absorbance, respectively.

In this study, the material to solvent ratio of *Mangifera indica* (mango leaves) was observed with specific methods and conditions. A yellowish-green color from *mangiferin*, which is contained in *Mangifera indica* (mango leaves), was expected and finally shown. In addition to discovering the optimum ratio of material to solvent by yield and absorbance, the dye application for yellow color on fabrics was also investigated in this paper. Hence, further research about natural dye extraction from natural ingredients was needed to obtain a fit material to solvent ratio.

MATERIALS AND METHODS

Materials

Mangifera indica leaves (Gadung species) were gathered directly from the trees planted in Surabaya, Indonesia. The leaves were washed, sun-dried for about 4-5 days, and ground using a 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 ratios of material to solvent 1:7 and 1:15 were used in case of each variable for *Mangifera indica* leaves extraction. The extraction using reflux and Soxhlet set equipment was maintained by TP-101 digital temperature indicator at 100^{o}C on the three-neck rounded flask for distilled water, 64^{o}C for methanol, and 79^{o}C for ethanol following each boiling point. Samples were collected at 30 minutes intervals, filtered using filter paper Whatman Grade 40 (for reflux method), and the absorbance were measured using HP-8453 Double Beam UV-Vis Spectrophotometer at a wavelength of 463 nm for *Mangifera indica* leaves. Time variation was employed in three ways i.e. 2, 4, and 5 hours. The yields at the end of time variation were calculated by taking 5 ml of samples that were dried in an oven for 2 hours at 110^{o}C. Moreover, LC-HRMS analysis was done using Thermo Scientific^{TM}-Q Exactive^{TM} 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 x 1 mm x 1.9 u particle size analytical column. Throughout the analysis process, the column oven is maintained at 30^{o}C. The dye application for both ratios was applied in linen cloth which had a high absorption.

RESULTS AND DISCUSSION

Mangifera indica Spectral Analysis

The absorbance data of *Mangifera indica* leaves extraction for each method are presented in Fig. 1 and Fig. 2, respectively.

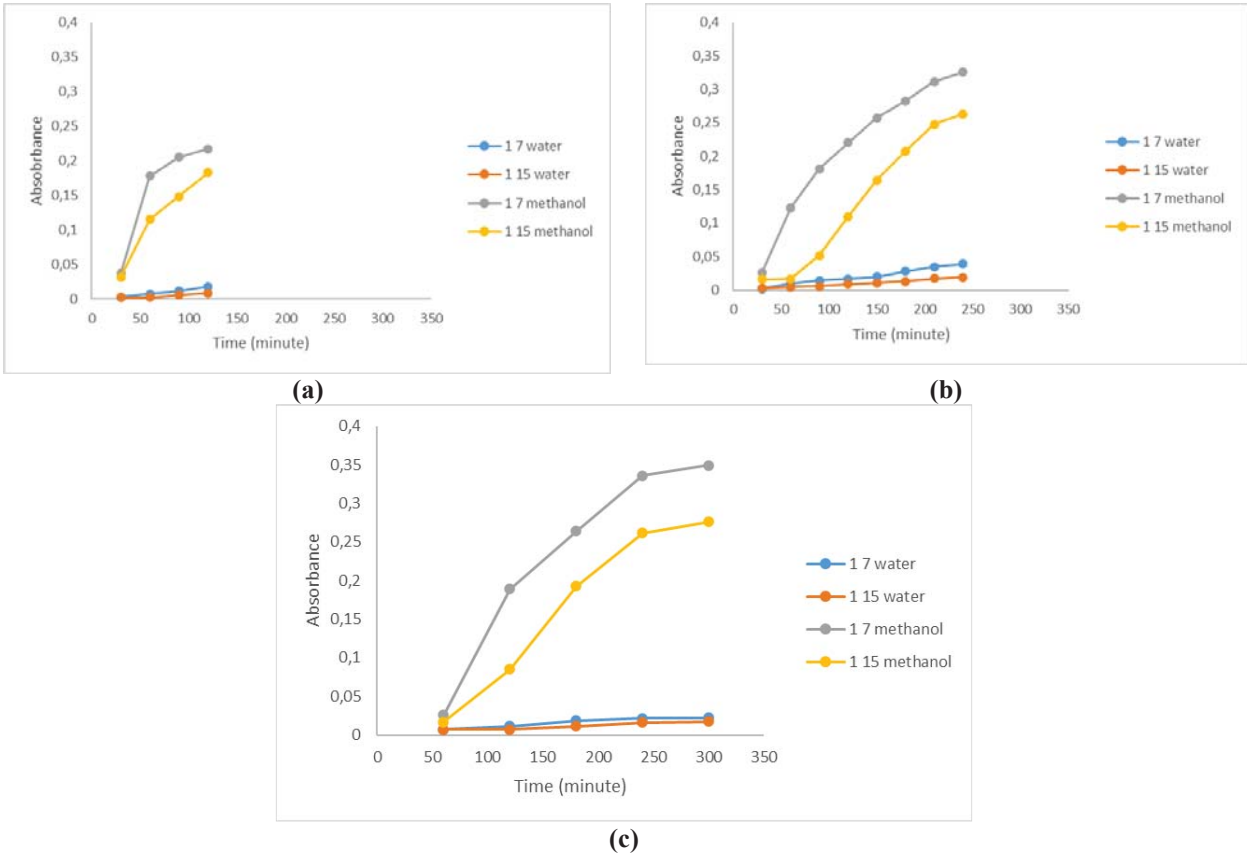
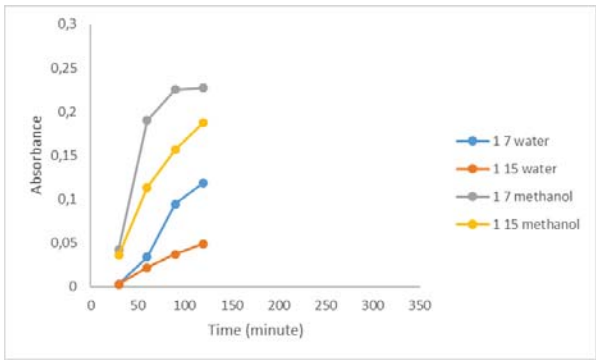


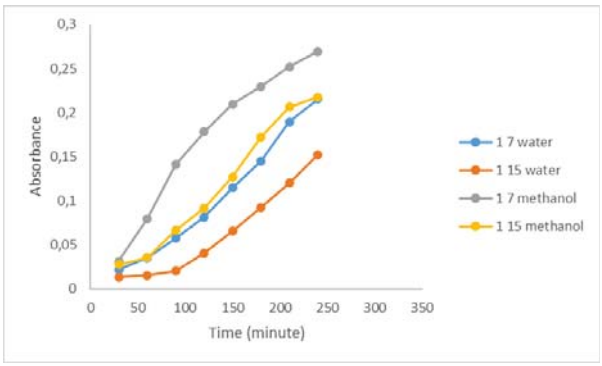
FIGURE 1. Methanol and Water Solvents Absorbance vs Time (min) Relation for **(a)** 2 hours; **(b)** 4 hours; **(c)** 5 hours by Soxhlet Method

The absorbance trend vs time for methanol solvent in Fig. 1 showed an increase then reached a steady state as shown in Fig. 1c for 5 hours extraction. In the case of solvent, the usage of water was not shown any significant absorbance rise compared to methanol. Thus, it is indicated that the Soxhlet method for leaves extraction was more suitable with methanol solvent. Due to methanol's volatile characteristic and lower surface tension than water (20.21 mN/m for methanol and 58.98 mN/m for water), the extraction process in the materials' pore was easier [7]. Furthermore, the functional groups in methanol (-OH and -CH_3) could dissolve either polar or non-polar compounds which could extract the *mangiferin* better than water as a universal solvent [8].

Both of the solvents for *mangiferin* extraction by reflux method generated a high increase of absorbance along the time as shown in Fig. 2. However, methanol solvent still gave a higher absorbance than water solvent. Therefore, it can be concluded from Fig. 1 and 2 that methanol solvent produced a better performance for both extraction methods. This phenomenon happened because of the flavonoid compound that had the -OH functional group in *mangiferin,* which had similar polarity with the solvent. Therefore, the materials to methanol solvent ratio was further discussed.



(a)



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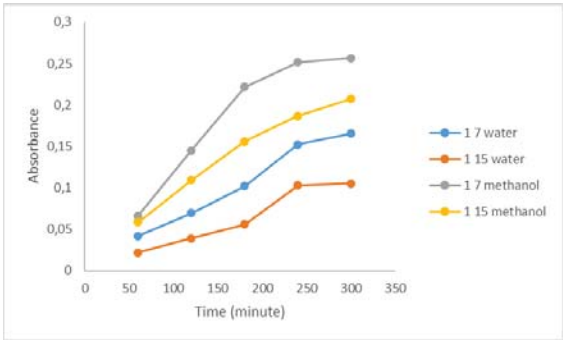
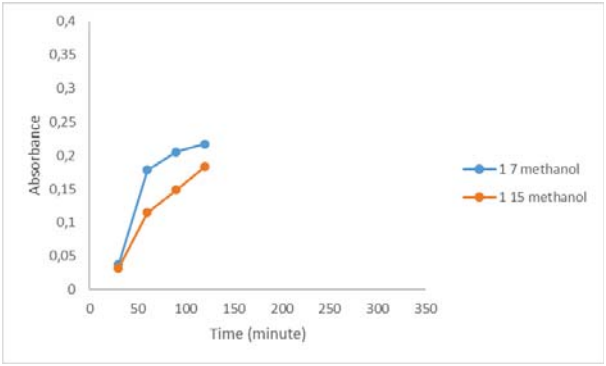


(c)

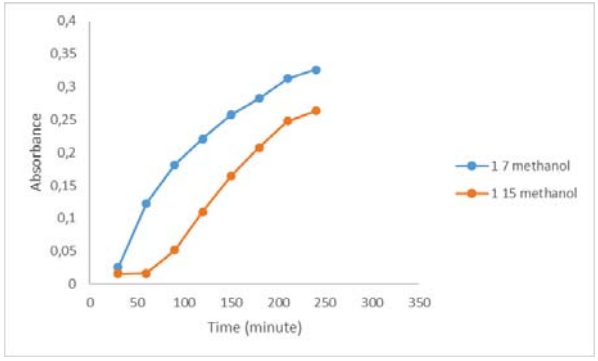
FIGURE 2. Methanol and Water Solvents Absorbance vs Time (min) Relation for **(a)** 2 hours; **(b)** 4 hours; **(c)** 5 hours by Reflux Method

Materials to Solvent Ratio in *Mangifera indica*

The plots of the absorbance with time are shown in Figure 3 and Figure 4 for each extraction method sequentially. The materials to solvent ratio of 1:7 produced a greater absorbance value for all extraction duration as shown in Fig. 3. Moreover, a 1:7 ratio by the Soxhlet method gave an economic advantage where it only needed a small amount of solvent to dissolve the same quantity of materials.



(a)



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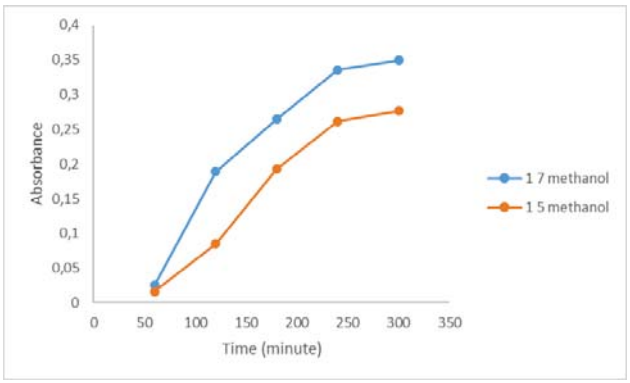
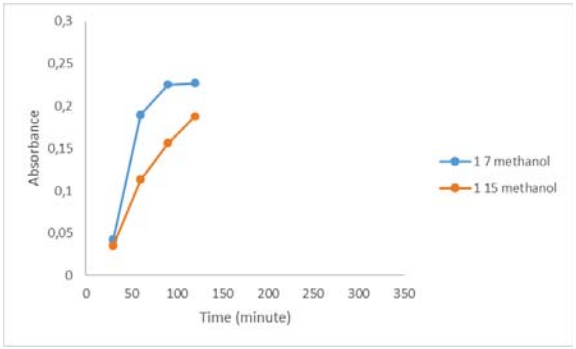


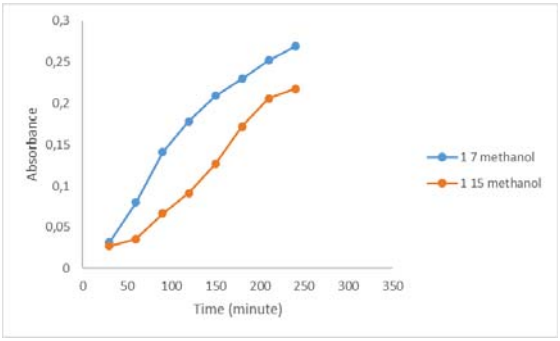
(c)

FIGURE 3. Methanol Solvent Absorbance vs Time (min) Relation for **(a)** 2 hours; **(b)** 4 hours; **(c)** 5 hours by Soxhlet Method

An extraction process of *Mangifera indica* by reflux method with methanol in Fig. 4 performed the same tendency with the Soxhlet method in Fig. 3 where 1:7 materials to solvent ratio gave higher absorbance results than the 1:15 ratio. A greater amount of solvent produced a lower absorbance because a large amount of solvent diluted the sample containing color substances.



(a)



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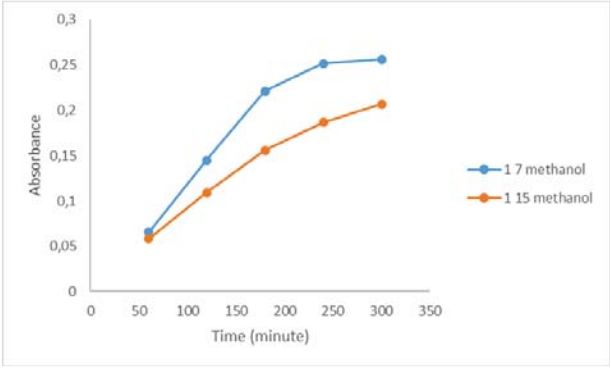


(c)

FIGURE 4. Methanol Solvent Absorbance vs Time (min) Relation for **(a)** 2 hours; **(b)** 4 hours; **(c)** 5 hours by Reflux Method

Comparing the extraction method by methanol solvent, the Soxhlet extraction gave a better extraction result than the reflux one based on the absorbance number. It was due to continuous reflux and siphoning method resulting renewable solvent that Soxhlet method had. In addition, a Soxhlet method was more suitable with a low boiling point solvent which could easily evaporate. Thus, the extraction process by methanol generated a better performance in the Soxhlet method than the reflux method.

Mangifera indica Yield Analysis

The yield of *Mangifera indica* extraction for each ratio was presented in Table 1 and Table 2.

TABLE 1. Yield Percentage of *Mangifera indica* Extraction by 1:7 Ratio

Method	Water Solvent (%)	Methanol Solvent (%)
Soxhlet ; 2 hours	0.89	2.16
Soxhlet ; 4 hours	1.22	4.1
Soxhlet ; 5 hours	1.14	3
Reflux ; 2 hours	1.41	2.51
Reflux ; 4 hours	2.08	2.72
Reflux ; 5 hours	1.71	2.54

TABLE 2. Yield Percentage of *Mangifera indica* Extraction by 1:15 Ratio

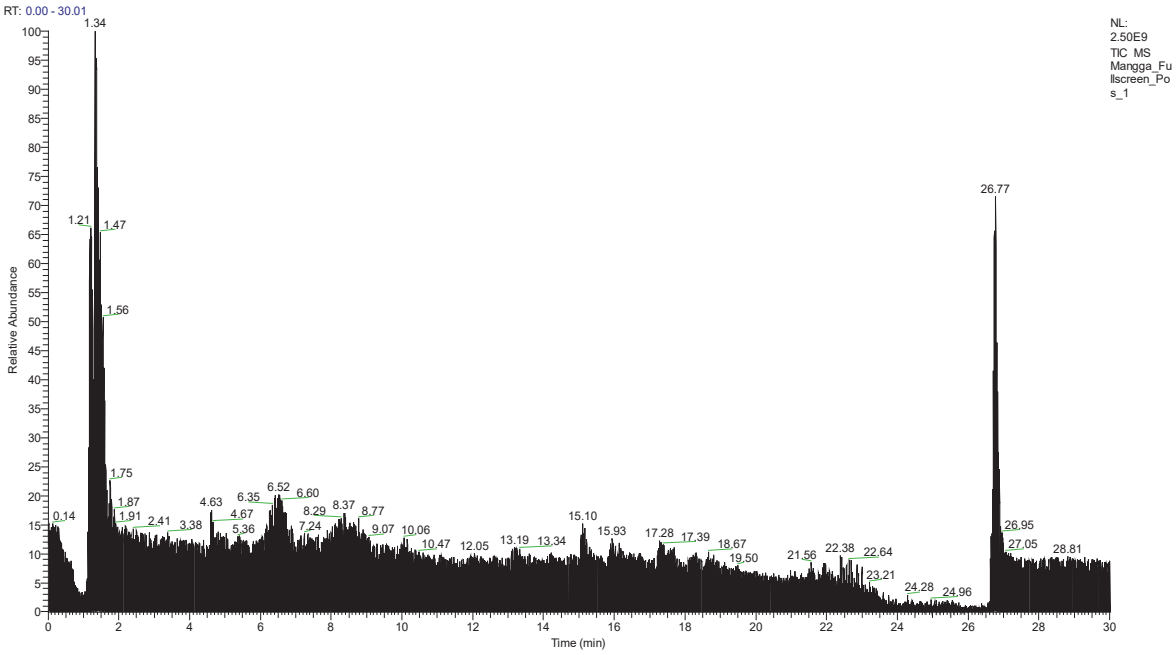
Method	Water Solvent (%)	Methanol Solvent (%)
Soxhlet ; 2 hours	0.15	1.74
Soxhlet ; 4 hours	1.14	3.7
Soxhlet ; 5 hours	0.66	2.61
Reflux ; 2 hours	1.3	1.22
Reflux ; 4 hours	1.46	2.1
Reflux ; 5 hours	1.38	2

As shown in Table 1 and Table 2, yield percentages for both extraction methods by water and methanol solvents were not proportional to the increase of extraction duration where a maximum yield was achieved at 4 hours of extraction. These results were also in line with the spectral analysis. Overall, the 1:7 materials to solvent ratio generated a greater yield value than the 1:15 ratio in every method and time of extraction.

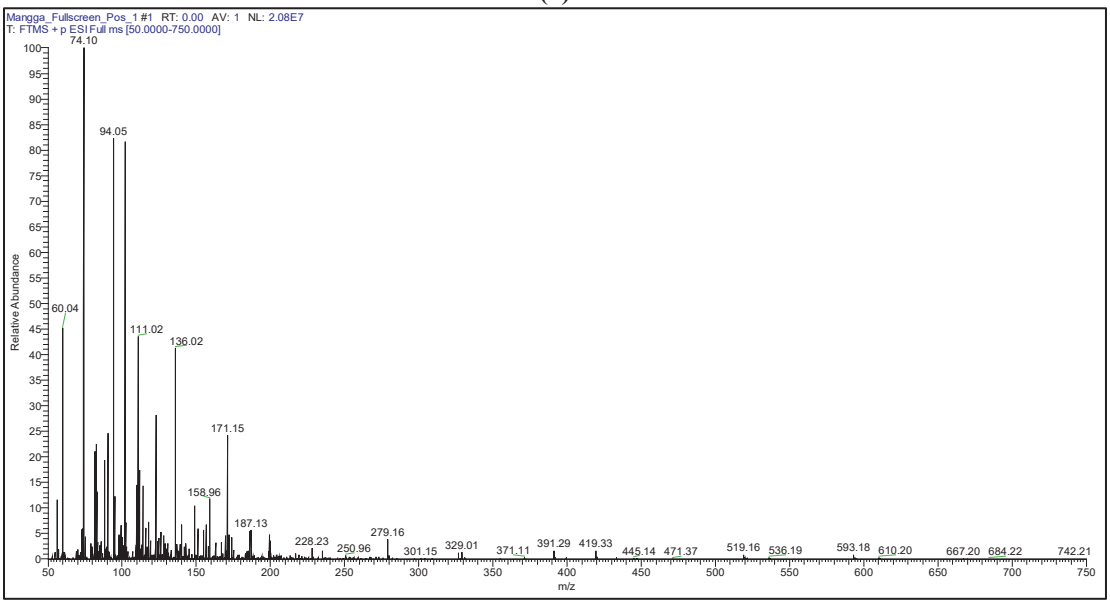
According to Zhang et al., (2018) [9], a similar polarity between materials and solvent played a big role in the extraction success. This statement was proven by the yield percentages in Table 1 and Table 2 which a higher yield was resulted from using methanol solvent.

Mangifera indica LC-HRMS Analysis

The best extraction performance of *Mangifera indica* (both ratios), Soxhlet extraction by methanol solvent for 4 hours, were shown in Fig. 5 and Fig. 6 respectively. LC-HRMS analysis by scan mode was chosen to identify and quantify the compounds containing in the leaves extract.



(a)



(b)

FIGURE 5. LC-HRMS Analysis of 1:7 Ratio. **(a)** Chromatogram; **(b)** Mass Spectrum

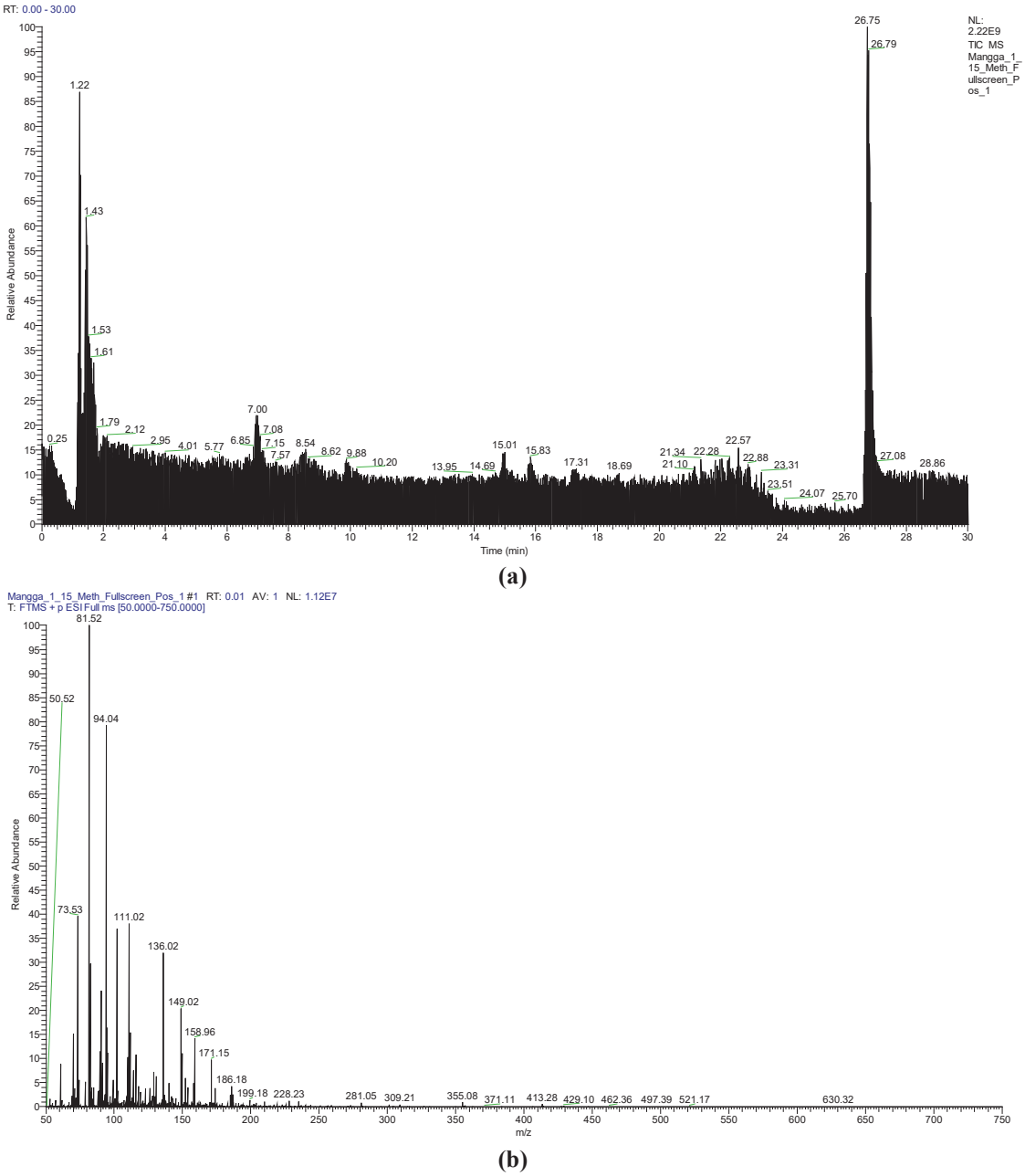


FIGURE 6. LC-HRMS Analysis of 1:15 Ratio. **(a)** Chromatogram; **(b)** Mass Spectrum

The highest 5 peaks in Fig. 5 and Fig. 6 were summarized in Table 3 and Table 4 consecutively.

TABLE 3. *Mangifera indica* LC-HRMS Analysis Summary for 1:7 Ratio

Compound Name	Retention Time	Area	m/z
N-(2,6-difluorophenyl)-2-(4-nitrophenyl) acetamide	1.34	38,054,009.88	73.3
Choline	1.47	3,459,385,491.39	104.02
Trigonelline	1.56	149,947,034.42	136.02
L-Norleucine	1.75	122,181,566.79	158.96
L-Phenylalanine	1.87	123,635,161.53	171.15

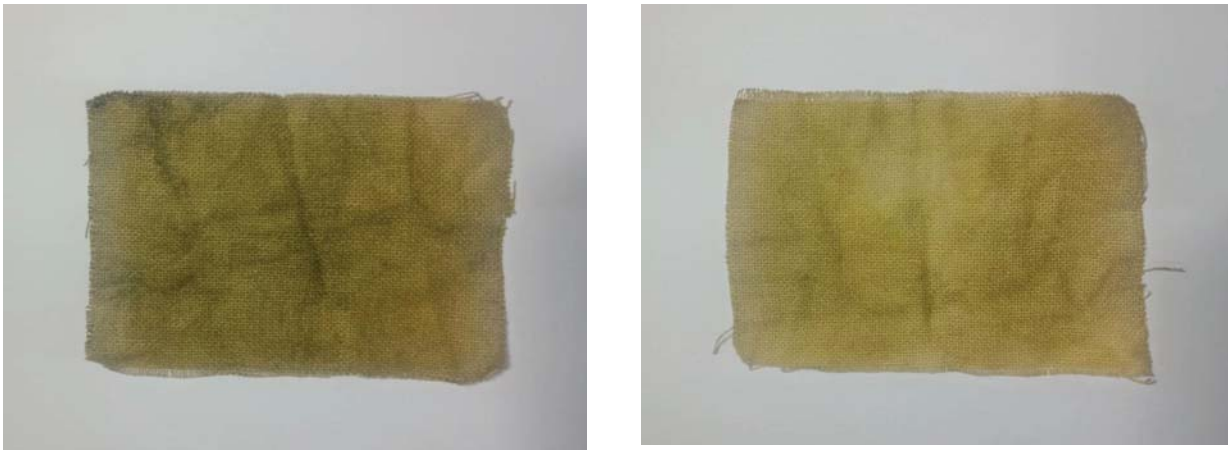
TABLE 4. *Mangifera indica* LC-HRMS Analysis Summary for 1:15 Ratio

Compound Name	Retention Time	Area	m/z
Choline	1.43	274,517,020.11	104.11
3-Hydroxy-2-methylpyridine	1.53	17,978,973.68	136.02
(7E,13E)-9,15-dihydroxy-4,10,16-trimethyl-1,5,11-trioxacyclohexadeca-7,13-diene-2,6,12-trione	1.61	1,237,464,377.44	149.02
L-Norluecine	1.79	29,187,369.42	158.96
Adenosine	2.12	245,360,485.90	171.15

LC-HRMS analysis for 1:7 and 1:15 ratios indicated the same choline compound with a different retention time as shown in Table 3 and Table 4. The late detection time for choline in the 1:15 ratio was due to a minimal compound contained in the sample. The m/z values for both ratios also showed a similarity number with data from Pubchem NCBI respectively by 104.02, 104.11, and 104.10. Thus, it can be concluded that the ratio difference did not affect the content in the sample.

Mangifera indica Linen Cloth Coloring

The coloring results were shown in Figure 7 for 4 hours of Soxhlet extraction by methanol solvent.



(a) **(b)**

FIGURE 7. *Mangifera indica* Coloring Results in Linen Cloth for **(a)** 1:7 Ratio; **(b)** 1:15 Ratio

The coloring result of *Mangifera indica* extraction produced a darker color in thew 1:7 materials to solvent ratio (Fig. 7a) than the 1:15 ratio. The pale color in Fig. 7b might be due to the coloring agent dilution in the 1:15 materials to solvent ratio.

CONCLUSION

This research has conducted the extraction processes of yellow colour from mango leaf. The extraction process required different process conditions to optimize the yield of dyes produced. Methanol was found to be the best solvent for *Mangifera indica* extraction from spectrophotometer and LC-HRMS analysis. Regarding the material to solvent ratio, the 1:7 ratio showed to be the best extraction performance for both materials. The application of color into the fabric showed a promising result and further research are still required to enhance the colouring process of fashion in industrial scale.

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