Natural Dye Extraction From Teak Leves *(Tectona Grandis)* Using *Ultrasound Assisted Extraction* Method for Dyeing on Cotton Fabric

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Abstract. Synthetic dye waste of textile industry has contaminated the water and aquatic living organism. One of the solution is used teak leaves as the natural dye. Common conventional extraction process is soxhletation which need more time and more solvent. One of extraction method that has been developed is ultrasound assisted extraction (UAE). The objectives of this research were to study the effect of solvent, extraction time, pH, and extraction yield. The conclusion of this research was optimum yield of teak leaves extract about 42,79% at pH 3 and extraction time of 20 minutes with aquadest as the solvent using UAE method. If natural dye of taek leaves was used on cotton, the color will change depend on fixator that was added. In GC-MS analysis, some compound that were detected are decanoic acid methyl ester and methyl tetradecanoate. Both of the compound have chromophore cluster.

1. Introduce

In April 2016, Rancaekek people in Bandung complained about textile industrial waste which contaminated their agriculture lands, the lost was 11,3 trillion Rupiah. Textile industrial waste which was thrown to river contaminated water and aquatic living organism. The contamination of textile industrial waste was shown by physical condition of river surface, the colors changing, turbidity of the water, the smell of the water and agriculture lands that was broken near the river.

One of the solutions to reduce the impact of industrial textile waste so it is not dangerous anymore for the environment or living organism is to use the natural dye for textile. Teak leaves *(Tectona grandis)* can be used as natural dye. Teak leaves contain anthocyanin which is source of brown color [1].

Conventional extraction process that commonly used in research before to extract teak leaves are soxhlet extraction, reflux extraction method, and maseration which is need more time and more solvent. According to those reasons inovation of extraction technology is needed that can save time of extraction process and solvent and also environmental friendly. One of extraction method which be improve recently is Ultrasound-assisted Extraction (UAE). Ultrasound-assisted extraction (UAE] offers an inexpensive, environmentally friendly, less time consuming and efficient alternative to conventional extraction techniques. The enhancement in extraction obtained by using ultrasound is mainly attributed to the effect of acoustic cavitation produced in the solvent by the passage of an ultrasound wave [2,3]. Ultrasound also offers a mechanical effect allowing greater penetration of solvent into the sample matrix, increasing the contact surface area between the solid and liquid phase, and as a result, the solute more rapidly diffuses from the solid phase into the solvent [4,5].

This study was to know about parameters that effect yield of teak leaves extract, those parameters are solvent, extraction time, and pH. After that is to determine the optimum yield, and qualitative analysis of pigment in yield, the chromophore cluster using GC-MS, and will be applied on fabric.

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2. Methods

2.1. Materials

Dry teak leaves with moisture content approximately 10% were purchased from Surabaya, ethanol 60%, HCl, NaOH and aquadest.

2.2. Procedure

First, teak leaves were grinded inside the blender and then sieved until have size 35 mesh – 60 mesh. After that 1 gram of teak leaves and 200 ml of solvent were put into three neck round flask. Then, three neck round flask was put inside the ultrasound cleaning bath. Extraction process was done at temperature 60°C in atmospheric pressure, 200 ml of solvent, 40 Hz frequency, ratio teak leaves with solvent was 0.005 g/ml. The extraction process has done in various variable such as time (5, 10, 15, 20, 30, 40, 50, 60 and 70 minutes), solvent (ethanol 60% and aquadest), and pH (3, 4, 5, 7, 9 and 10). Adding HCl and NaOH to make condition become acid or base. Here is the ultrasound assisted extraction equipment:

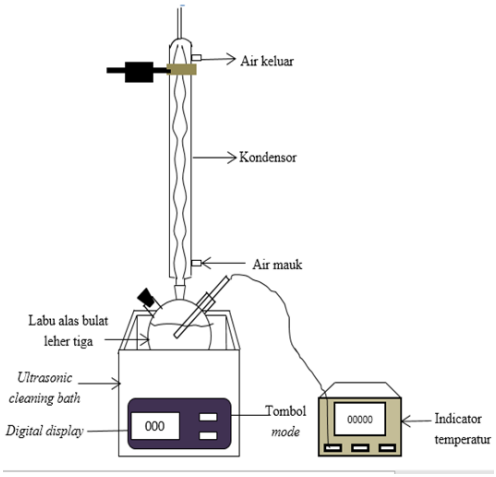


Fig 1. ultrasound assisted extraction equipment

Extraction process will be stopped when the extraction time was completed. After the extraction process then the filtrate will be filtered using vacuum pump to separate it from impurities. Natural dye that mixed with the solvent were separated using hot plate, the solvent will be vaporized and the natural dye powder will stay on the bottom of beaker glass. The natural dye was analyzed by GC-MC and will be applied on cotton fabric.

3. Results and Discussion

3.1 Effect of Solvent

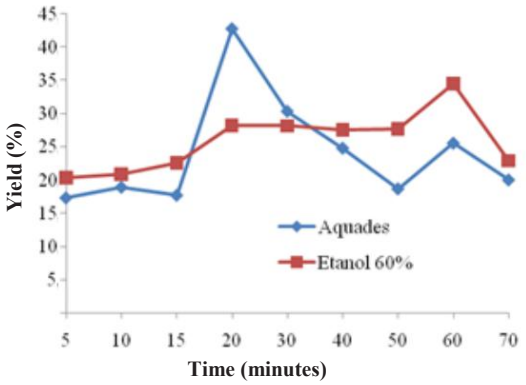


Fig 2. Yield vs Time (Minutes) of Teak Leaves extraction in pH 3

Fig 2 shows the effect of reaction time on teak leaves extract. Extraction using aquadest produce teak leaves extract greater than using ethanol as the solvent. Anthocyanin is polar molecule which has aromatic ring. Anthocyanin polarity influence the extraction process [6]. Commonly polar organic solvent suitable for anthocyanin extraction. The optimum yield of teak leaves extraction with aquadest as solvent in pH 3 was 42,79% at 20 minutes. Meanwhile the optimum yield of teak leaves extraction with ethanol 60% as solvent in pH 3 was occur at 60 minutes. Based on **fig 2**, teak leaves extract was decreasing after reach the optimum condition. It's occur because of vaporization process using hot plate make yield of natural dye also vaporize with the solvent.

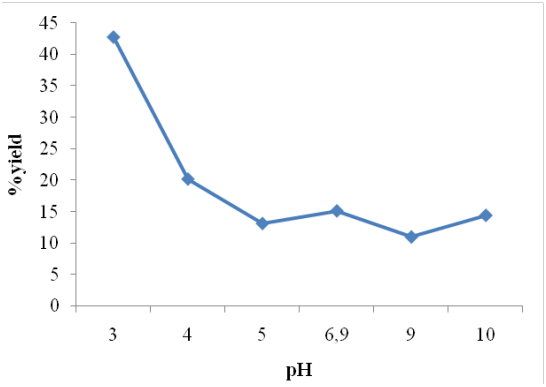
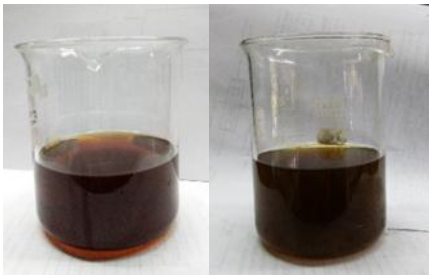


Fig 3. Effect of pH to yield of extract teak leaves

Fig 3. shows that optimum yield of teak leaves in acid extraction condition. The yield of teak leaves extract decreased with increasing of pH. Possibly due to the decreasing of pH makes hydrogen ion density increase and obstruct releasing hydrogen ion [7].

3.3 Effect of Extraction Time



(a) **(b)**

Fig 3. Effect of extraction time to yield of extract teak leaves in minutes (a) 20 (b) 30

Fig 3. shows that color of teak leaves extract in extraction time of 30 minutes was darker than extraction time of 20 minutes, extraction time have effect for the color of the teak leaves extract, from that can be concluded that the more time needed for extraction process, the more material extracted [8].

3.4 GC-MS Analysis

Based on GC-MS analysis, the main compound of teak leaves extract were decanoic acid methyl ester (41,16%), methyl tetradecanoate (22,84%), and hexadecanoic acid, methyl ester (15,17%). Some of that compound consist of chromopore cluster, it is substance which bring colors. **Fig 4.** and **fig 5.** Shows the compound structure of decanoic acid, methyl ester and methyl tetradecanoate. Chromophore cluster can be found in their compound structure which is ketone cluster (C=O) and carboxylate cluster (COOH).

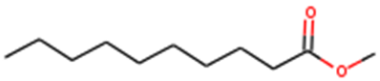


Fig 4. Structure of Decanoic acid, methyl ester

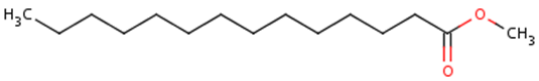


Fig 5. Structure of Methyl tetradecanoate

Table 1. Compounds of Teak Leaves Extract based on area of Teak Leaves Extract

No	Retention Time	% Area	Name
1.	3,076	4,81	*Decanoic acid, methyl ester*
2.	4,894	41,16	*Decanoic acid, methyl ester*
3.	7,020	22,84	*Methyl tetradecanoate*
4.	9,094	15,17	*Hexadecanoic acid, methyl ester*
5.	10,632	0,26	*Heptadecene-(8)carbonic acid-(1)*
6.	10,746	0,56	*Pyridine-3-carboxamide, oxime, N-2-trifluoromethylphenyl*
7.	10.798	8,53	*9-Octadecanoic acid, methyl ester,(E)-*
8.	11,015	4,02	*Heptadecanoic acid, 16-methyl-, methyl ester*
9.	12,284	1,74	*Thiosulfuric acid*
10.	13.124	0,92	*8-Acetyl-3, 3-epoxymethano-6, 6, 7-trimethylbicyclo*

4. Application on Cotton Fabric

Application of teak leaves extract on cotton fabric was done by adding some fixator such as $CaCO_3$ and $Al_2(SO_4)_3$. Some test were done to know the tenacity of the color such as dipped in TRO (Turkey Red Oil) about 24 hours, TRO has function as dispersing agent which can disperse color to the cotton fabric entirely. Besides, TRO can omit chemical substances or impurities which stick on the cotton fabric so the colors can stick on cotton fabric sturdily. After that the cotton fabric was washed by water then dye into liquid of teak leaves extract and dry in the sun.

Fig 6. Application Teak Leaves Extract on cotton fabric

Fig 6. shows the result of teak leaves extract which was applied on cotton fabric. The precipitation of natural dye will be increase after cotton fabric was dipped in fixator, because the metal grained inside cotton fabric can precipitate natural dye in a big amount compared without fixator [9].

5. Conclusion

Natural dye from teak leaves can be extracted using ultrasound assisted extraction at 60°C in atmospheric pressure, 40 Hz frequency, extraction time were 5, 10, 15, 20, 30, 40, 50, 60 and 70 minutes, solvent ethanol 60% and aquadest, and pH 3, 4, 5, 7, 9 and 10. Optimum yield of teak leaves extract is 42,79% at pH 3 and extraction time 20 minutes with aquadest as the solvent. If natural dye of taek leaves are used on cotton, the color will change depend on fixator that be added.

6. References

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