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Textile natural dye powder of *Terminalia catappa* leaves

M Krisnawati, I W N Cahyani, O Paramita and A Kusumastuti

Faculty of Engineering Universitas Negeri Semarang, Kampus UNNES Sekaran,
Gunungpati, 50229 Semarang Indonesia

mariakrisnawati@mail.unnes.ac.id

Abstract. This study was carried out to utilize the extract of *Terminalia catappa* leaves as natural dyes powder on cotton fabric using alum mordant and various solvents. *Terminalia catappa* leaves were extracted using solvents of water, 25% ethanol, 50% ethanol, 75% ethanol, and 100% ethanol at a ratio of 1:10 with pre-mordanting and fixation methods. Natural dyes powder was characterized using parameters of colour direction, colour intensity, and colour fastness. It was found that type of solvent affected the colour direction and colour intensity. The results revealed the direction of yellow-green to brown colour. It can be concluded that extract of *Terminalia catappa* leaves could be used for the production of natural dyes powder on cotton fabric.

1. Introduction

In the past, the clothes of the nobility or the rich were dyed using natural dyes. Natural dyes obtained from natural sources of animals, plants can come from roots, stems, leaves, fruit, skin, and flowers. In plants, pigments generates different colours, depend on their chemical structure [1]. Tannins could be found in the leaves, bark, roots, and fruit of plants [2].

Nowadays, synthetic dyes are widely used in textile dyeing process. This is due to the numerous choices of colour and the abundant availability in local market. It promoted the use of synthetic dyes as the substitution of natural dyes. Moreover, the synthetic dyes offer brighter colour, fixed colour composition, as well as the applicability in various types of fibres, both natural and synthetic fibres. The dyeing process using synthetic dyes are also easier and faster. Leucea [3] reported the possibility of allergic reactions induced by the utilisation of disperse dyes. The carcinogenic potential is also threaten the users of azo dyed materials. Along with the awareness to environment and the health hazards associated with synthetic dyes, people in various countries have been concern in using back the natural dyes [4]. The alteration in the use of synthetic dyes to natural dyes is strongly influenced by public awareness of health and environmental safety [5].

Natural dyes generate soft colours, offer matching and harmonious properties. The application of natural dyes in cotton fibres generates a distinctive aroma. Wastewater of natural dye is relatively environmentally friendly. Moreover, the wastewater is safe for health because the substances contained in natural dyes are easily decomposed and harmless to the environment. The fabrics dyed with natural dyes have a higher value than those of chemical dyes.

Utilisation of natural dyes in textile application have been studied by some researchers [6-8]. For textile application, the natural sources should be selected by considering the availability. Non edible materials are highly recommended to be utilised. Most researchers applied the food waste such as fruits peel and fruits seeds, some other researchers utilised bark and dry leaves.



Ketapang (*Terminalia catappa*) is one of the sources of natural dyes. Ketapang leaves contain tannin of about 11%-23% [9]. Previous studies on the application of ketapang leaves as textile dyes have been done [9-11]. Purnama et al. [9] investigated the tannin level as the influence of maceration time and initial conditions of leaves (dried and fresh). The highest tannin level was obtained at immersion time of 6 days using fresh leaves. Similar result was also found by Faisal and Chafidz [8]. Nofitarini et al. [11] found the best extraction time of 30 minutes to produce the highest tannin level.

However, the previous studies focused on the utilisation of fresh extract of *Terminalia catappa* as textile dyes. This impractical process requires long time preparation with inconsistent results. The generated colours could be different among different extraction processes. It is therefore, production of natural dyes powder is of important in enhancing the utilisation of natural dyes in textile processing.

2. Research method

Aqueous extraction was used to extract the colouring matter of *Terminalia catappa*. Prior to extraction, the raw material was broken into small pieces or powdered to enlarge the surface area. The fine material was then brought to boil and filtered to remove the impurities. Despite the process simplicity, this technique suffers from the possibility of dye decomposition due to the application of high temperature. It is therefore; the requirements of high temperature resistance as well as water solubility are inevitable for this method.

2.1. Materials

Terminalia catappa leaves were used as sample of natural dyes. Deionised water was used for all of the solutions preparation. Ethanol 96% was used as solvent. Commercial Arabic gum, maltodextrin, cotton fabric, sodium carbonate, Turkey red oil (TRO), alum, ferrous sulphate, and lime were purchased in local stores.

2.2. Procedure

This research studied the independent variables of solvents and filler types. The solvents used in this research were water, 25% ethanol, 50% ethanol, 75% ethanol, and 100% ethanol. The fillers used were maltodextrine, arabic gum, and no filler. The dependent variable in this study was quality of dye powder in terms of colour direction, colour intensity, and fastness to washing of cotton fabric. This study compared the application of fresh extract and *Terminalia catappa* powder.

Extraction of ketapang leaves was done at ratio of 1:10 with reflux extraction method. Production of natural dyes powder was carried out using spray dryer at speed of 10 ml/minutes. The spray dryer machine used compressor pressure of 3 bar, discharge of 190/22, and heating temperature of 220°C. During the drying process, filler of maltodextrin and gum arabic was added as coating material at concentration of 10%. Dyeing process was done at ratio of dyes powder to water of 1:6. Cotton fabric was used as sample, dyed at a temperature of 65°C using a hot plate to maintain the even and consistent temperature.

The colour intensity (R %) of the dyed fabric was measured using a spectrophotometer (UV-PC). Colour fastness to washing was measured using grey scale.

3. Results and discussion

3.1. Colour intensity

The amount of dye absorbed in the material expressed by K/S. The reflectance (% R) of test sample was measured at a wavelength of 400 – 700 nm with an interval of 20 nm to determine the maximum wavelength at the lowest % R value. The lower value of R% generates darker colour of fabric, on the contrary, higher value of R% indicates lighter colour, towards white. The reflectance value was converted to a color intensity value (K/S) based on the Kubelka-Munk equation as follows [12]:

$$K/S = \frac{(1-R)^2}{2R}$$

Table 1 describes the colour intensity of cotton fabric dyed with ketapang leaves extract powder. Colour intensity test of ketapang leaves dyes powder produced different R% and T% values. The colour strength depended on the process of dye production, type of solvent, and type of filler. The average value of colour intensity in this study was in the interval of 26.68% to 92.42%.

Table 1. Colour intensity












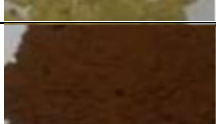
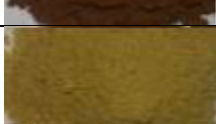
Sample Code	Reflectance (%)
A1	16.17
B1	19.37
C1	7.58
A2	52.06
B2	38.41
C2	19.84
A3	35.85
B3	34.38
C3	24.11
A4	53.18
B4	42.84
C4	52.87
A5	58.93
B5	64.04
C5	35.55

Textile dyeing using fresh extract of ketapang leaves generated lower colour intensity than that of ketapang leaves powder. Colour intensity of ketapang leaves dyes powder without the addition of filler material was significantly different to that of added with Arabic gum and maltodextrine fillers. This study was in accordance with the research conducted by Asih [13] generated colour intensity results of leaves powder with addition of 10% maltodextrin resulted in lower colour intensity values than that of without fillers. Study of Herlina [14] on natural dye of red dragon fruit also found that addition of 30% of maltodextrine resulted in the lowest colour intensity. Gum arabic fillers produced darker colour than that of maltodextrine. The dyes powder of ketapang leaves extract with water solvent have average colour intensity in the range of 80.63% to 92.42%. The highest colour intensity was also provided by system with water solvent. System using 50% ethanol as solvent produced almost the same colour intensity for both maltodextrine and arabic gum fillers.

3.2. Colour direction

The results of colour direction are shown in Table 2. Samples with maltodextrin and arabic gum as fillers generated yellow to light brown colours as peanut, peacan, tortilla, chesnut, dark elm, medium oak, and light walnut. Production of natural dyes powder without filler led to darker colour, in this research, light brown to black colour was obtained.

Table 2. Colour catalogue

Sample Code	Dyes Powder	Colour Catalogue
A1		Peanut
B1		Peacan
C1		Caramel
A2		Tortilla
B2		Chesnut
C2		Sepia
A3		Tortilla
B3		Tortilla
C3		Syrup
A4		Chestnut
B4		Dark Elm
C4		Wenge
A5		Medium Oak

B5		Light Walnut
C5		Brown

Where:

1 = water solvent

2 = ethanol 25%

3 = ethanol 50%

4 = ethanol 75%

5 = ethanol 100%

A = maltodextrin filler

B = Arabic gum filler

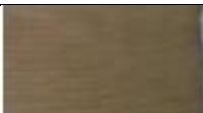
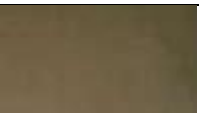
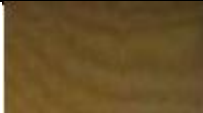





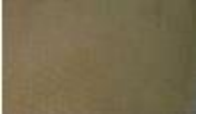
C = without filler

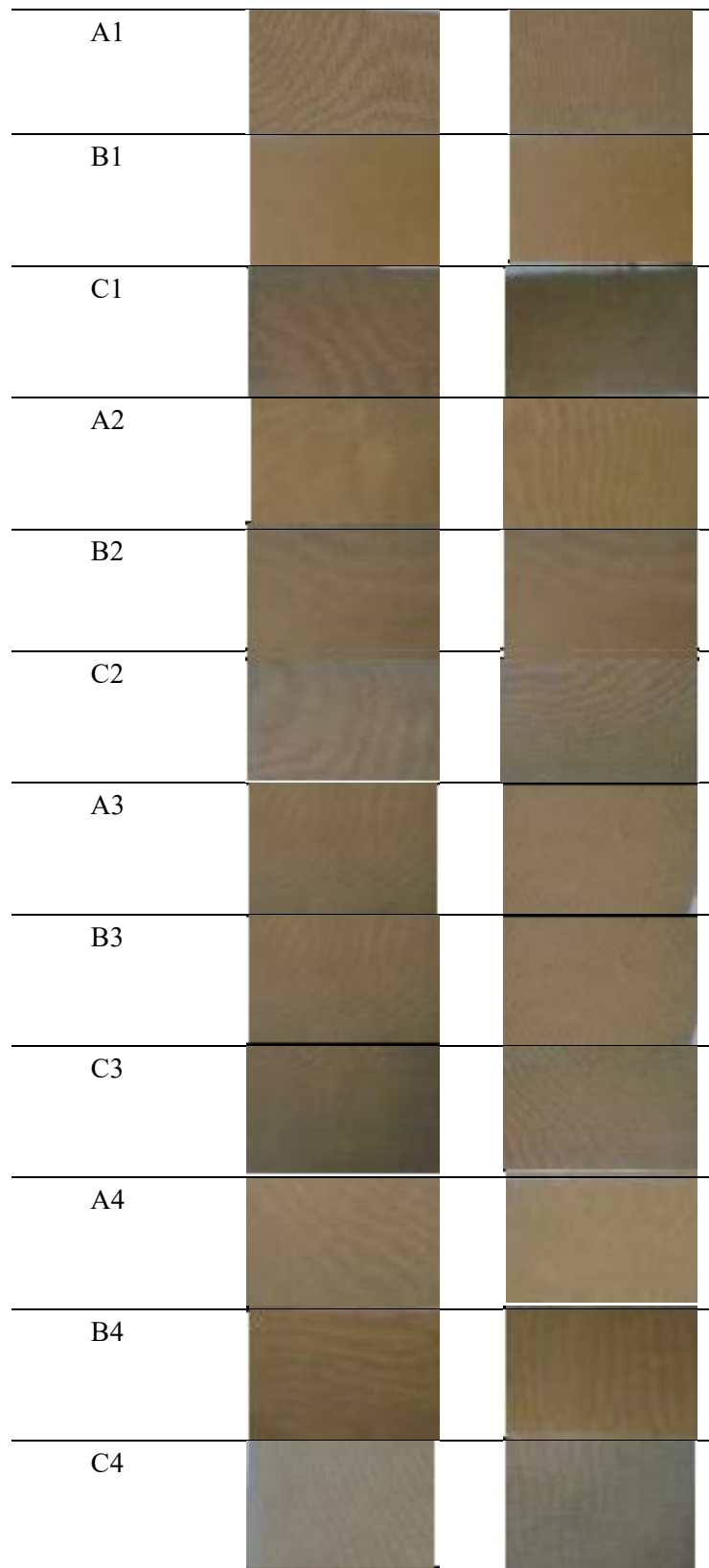
The results of colour direction revealed that ketapang leaves could be used as natural dyes in powder form by generating various colours by applying different solvent and filler types. This study was in line with that of Purnama et al. [9], dried ketapang leaves extracted by maceration method with water solvent for 8 hours produced the most colour direction. The colour produced by ketapang leaves was brownish yellow to dark brown.

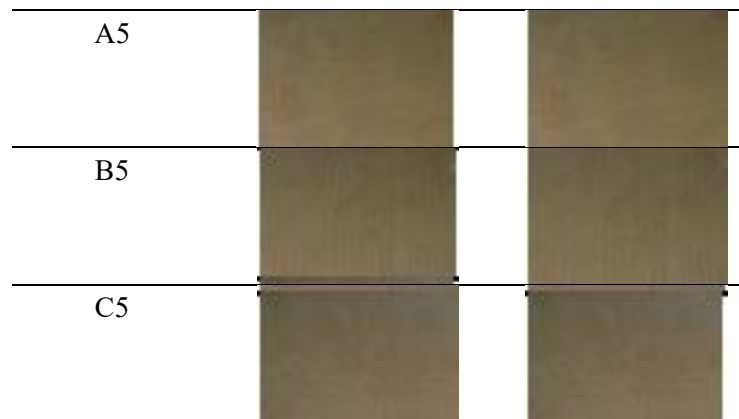
3.3. Colour fastness to washing

Colour fastness test to washing of textile fabrics dyed using natural dyes of ketapang leaves extract powder under various types of solvent and filler were carried out. Fastness test to soap washing could be measured based on the colour difference before and after soap washing. The fastness test to washing was carried out to determine the fastness of cotton fabric dyed using ketapang leaves extract powder. Results of colour fastness to soap washing are shown in Table 3.

Table 3. Colour Fastness to Washing

Sample Code	Before Washing	After Washing
1		
2		
3		
4		
5		





Mordanting process plays an important role in the textile dyeing using natural dyes. This is due to the fact that natural dyes have no affinity with textile fibre, but could form a complex compound with metal salts [15]. The complex compound could react and bond with textile fibre thus the fibre could be dyed and having good colour fastness.

Study in the performance test of ketapang leaves extract powder was also done in colour fastness. It is revealed that after soap washing, all samples were proven to have excellent colour fastness, indicated by no colour change was found in all treatments. This finding conforms the study of Purnama [9] that ketapang leaves with alum fixator was well absorbed in the fibre that the tannins were bound and absorbed very well. Study of Anzani et al. [16] showed that natural dye of sour sop leaves extract combined with alum mordant provided good quality of colour fastness to washing. This finding was also supported by Prabhavathi et al. [17] in the utilisation of some natural dyes in cotton dyeing.

4. Conclusion

Ketapang leaves (*Terminalia catappa*) were proven to be potential as a natural dye powder. The powder production was carried out using a spray dryer. Two types of fillers were varied to minimise water content thus enhance the stability as well as to increase the yield. The colour direction of ketapang leaves extract powder was yellow to brown. The quality of colour intensity resulted from natural dyes powder using water and ethanol solvents were quite good. Colour fastness to washing for all treatments was excellent, indicated by no colour difference in the washed samples. Based on the performance tests, ketapang leaves extract powder is qualified to be utilised as textile dye powder.

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