Brachiaria mutica dyes powder for textile application: dyeing quality of cotton fabrics

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Abstract. To minimise water pollution as well as to improve the economic value of some un-utilised natural resources, application of natural dyes have been widely encouraged this past decade. Natural dyes considered to be effective in minimising the use of chemical agents which in turn overcome pollution problems. Despite the advantages of natural dyes, few problems are also encountered. It is therefore production of natural dyes powder becomes another concern. In this article, application of *Brachiaria mutica* as natural dyes powder for textile dyeing was discussed in terms of colour darkness, colour fastness, and colour difference. Experiment was carried out by observing dyeing process using 4 types of mordants, i.e. lime, alum, limestone, and ferrous sulphate. The colour darkness of cotton fabric was in the order of system with ferrous sulphate > limestone > alum > lime juice. The system with alum, limestone, and ferrous sulphate generated colour of green, while system with lime juice resulted in yellowish green colour. In term of colour fastness to washing, the quality was in the order of system with ferrous sulphate > limestone > alum > lime juice.

1. Introduction

Batik is one of traditional fabrics of Indonesia. Literally, batik is a fabric with little dots'. Batik is made by a resist process for dying where the patterns are reserved on the textiles by giving wax to certain areas prior to dying. In Indonesia, batik industry dominated world market that it is able to drive the national economy. Batik industry in Indonesia is available in 19 regions. The name of batik is then labelled based on the region, such as Pekalongan batik, Surakarta batik, Yogya batik, Lasem batik, Cirebon batik, Sragen batik. Each batik from the area has a specific motif. There are three types of batik produced, namely hand-drawn batik, stamped batik and combination batik. The development of batik industry in Indonesia is closely related to the development of batik which began hundreds of years ago. Pekalongan is one of the largest central of batik industry. The potential of small and medium industries of Pekalongan batik is economically sufficient in providing a huge income to the country, both in terms of employment and income of foreign exchange and taxes.

The recognition of Indonesian batik as "Intangible World Heritage" on 2 October 2009 by the UN Agency, i.e. Educational, Scientific and Cultural Organization (UNESCO) received a good response from the community which in turn increasing market demand for both local and foreign consumption to be wide open, thus providing a great opportunity for the development of this industry. It was very helpful in boosting batik popularity and public awareness to preserve batik. The imposition of batik uniforms has been applied in many institutions on certain days.

The development of SMEs Batik is increasing from year to year and the exports value continues to grow. It can be seen from the exports achievement of batik and batik products in 2017, reaching about 58.46 million dollars with the main destination countries of Japan, the United States, and Europe. It was reported that in 2018, there were 136 thousand batik industries or about 20% of national textile small-medium industries. While textile small-medium industries were about 680 thousand units, or about 20% of national small-medium industries which were about 3.4 million units [1].

Central Java dominates about 91.6% of batik industry in Indonesia, in which most of them are available in Pekalongan. Globalization challenges, such as the enactment of ACFTA agreement, require batik entrepreneurs in Pekalongan to compete with batik-likes textile products from other countries, such as China and Malaysia. Local competitors who are also increasingly aggressive in bringing up batik designs with new motifs made competition in batik industry even tighter. The increase level of competition created barriers to market entry and market concentration of entrepreneurs in industry. In turn, it affected the shape of market structure in the Pekalongan batik small and medium industry. Furthermore, to be able to continue to survive in increasingly fierce competition, companies in the industry must perform several behaviours that will ultimately affect the performance of industry.

In spite of the contribution of batik industry towards national economics, its existence is a threat to the surrounding environment. Production process of batik involves the application of dyes and chemical agents which are harmful to the environment. Research of Fajri [2] found that only about 0.6% of batik industries in Pekalongan have their own wastewater treatment installation. The high capital and operational costs forced the others to dispose their wastewater directly to the environment. The dyeing process resulted in the disposal of high BOD, COD, and TSS wastewater. It was reported that batik industries released wastewater contained BOD, COD, and TSS beyond the allowable threshold [3, 4]. The high content of BOD, COD and colour in waters can kill organisms and disturb ecosystem balance. The increase in BOD, COD, and colour content contributes in lowering water quality index.

To minimise water pollution as well as to improve the competitiveness of batik products, application of natural dyes have been widely encouraged this past decade. Textile dyeing using natural dyes considered to be effective in minimising the use of chemical agents which in turn overcome pollution problems. Considering high pollution due to the use of synthetic dyes, batik producers are moved to develop natural coloring that is environmentally friendly by developing the potential surrounding natural resources. It improves the economics value of those natural resources as well as protects the environment from hazardous effects of chemicals that can damage human life sustainability.

Many researches about exploration of natural dyes for textile application have been done [5-7]. However, they focused in the basic extraction of natural dyes. The process is complicated, started from raw material collection, cleaning and extraction. The extraction process is carried out by fermentation [8, 9], cold extraction [10, 11] as well heat extraction [12-14]. The dye extract is then used to dye the fabric. However, in mass production, this process is considered uneconomical. Moreover, it's difficult to generate consistent colour for textile fabric dyeing using natural dyes extract. More serious problems encountered in scale-up process to ensure the dyeing quality as well as optimise time proccesses. Commercially, another problem found in the distribution of natural dyes extract.

Considering the above problems, production of natural dyes powder is absolutely important. A research to produce natural dyes powder that can be used as simple as the application of synthetic dyes must be carried out. In addition, the resulting colour is also unique, with a comparable quality to that of synthetic dyes. Natural dyes powder is also promising due to its stability, easiness of storage, packaging and distribution. Effects of some parameters to rendement of natural dyes powder has been reported in the previous article [15]. Application of natural dyes powder for textile dyeing will be discussed in terms of colour darkness, colour fastness, and colour difference.

2. Methods

2.1. Materials

Brachiaria mutica was used as sample of natural dyes. Deionised water was used for all of the solutions preparation. Teepol, potassium alum, lime, limestone, ferrous sulphate, and cotton fabric were purchased from local markets.

2.2. Procedure

Production of natural dyes powder was done as given in Kusumastuti et al. [15]. The dyeing process involved fabric preparation and fabric dyeing in dye solution. Wetting process was done by immersing cotton fabric in teepol multipurpose detergent (2 g/l) for 24 hours, followed by fabric washing and drying. Pre mordanting process was carried out in concentration of 50 g/L for an hour. The mordanting solution was varied using potassium alum, limestone, ferrous sulphate, and lime. Pre-mordanting method is a mordanting process applied to fabric prior to dyeing. This method is commonly applied
Pre-mordanting is a technique which involves primarily mordanting the fabric before dyeing i.e. the mordant is applied to the fabric prior to dyeing. It's the most common method of mordant given to celluolosic and some animal fibres that don't have affinity to natural dyes. Pre-mordanting is very helpful in preparing fabric before dyeing process. The mordanting bath could be reused many times thus the overall dyeing procees becomes more economical as well as minimising the harmful contaminants. This process is also suitable for large scale application [5]. The pre-mordanted fabric was then immersed in dye solution in ratio of fabric to dye solution of 1:30 at 60ºC for 30 minutes. Dye solution was prepared by dissolving natural dye powder in deionised water at concentration of 2.5 g/L. Fixation process was then applied at potassium alum solution of 50 g/l for 5 minutes. After been washed, the fabric was dried under room condition. The dyed fabric was then examined for colour darkness using spectrophotometer, colour difference using chroma-meter, and colour fastness through washing process.

Colour darkness test resulted in reflectance values which converted into K/S units of colour darkness, i.e. dyes absorption into fabric fibres. Investigation of colour darkness of dyed cotton fabric using *Brachiaria mutica* extract was done using UV-PC Spectrophotometer Model ISR-2200 Shimadzu. Chroma meter is a tool to measure surface colour and surface darkness/lightness of a substance. Data output is in the form of L*, a*, and b* values. L* refers to levels of darkness/lightness between black and white, while a* and b* are chromaticity coordinate. The coordinate a* shows the balance between red/green, and b* between yellow/blue. Colour difference was measured using Chroma meter Konika Minolta CR-400. Colour fastness to washing is one of the most important and common quality parameter of dyeing results. Colour fastness to washing is defined by contacting a specimen of textile in contact with pieces of specified adjacent fabrics and agitated in a soap or soap-soda solution. The change in specimen colour and the adjacent fabrics staining are assessed with the grey scale. The resistance to colour loss of any dyed or printed material to washing is preferred to as its wash fastness. If dye molecules have not penetrate inside the inter polymer chain space of fibre or have not attached to the fibre with strong attractive force, poor washing fastness result. This test determines colour loss and change in washing process and possible staining of other garments or lighter portion that may be washed with it.

Standard assessment of colour fastness testing results for washing use the grey scale standard (Grey scale) and the staining scale standard (Staining scale). The grey scale standard consists of 9 pairs of grey standard plates which are used to assess colour changes in the colour fastness test by determining the degree of difference or colour contrast from the lowest to the highest level. The staining scale standard consists of a pair of white standard plates and 8 grey standard plates which are used to assess the colour staining of white cloth used in colour fastness testing, by determining the degree of colour difference from the lowest level to the highest level.

3. Results and discussion

3.1. Colour darkness

The colour darkness test results can be assessed by the percentage of reflectance (R %) and transmittance (T %). Effect of mordanting agents to colour darkness was investigated using lime, alum, limestone, and ferrous sulphate. Mordanting was done to remove the remaining impurities generated from the fabric processing. Application of natural dyes to textile fibre should be done with the aid of metallic salts as natural mordanting agent thus generates affinity of colouring matter and textile fibre [16]. Textile fibre absorbs mordant solution that allows the formation of metal ion and appropriate functional groups in the structure of the fibre. Interactions of dyes and complex of mordant-fibre could form brightly coloured species. The value of colour darkness given by dyeing of cotton fabric using *Brachiaria mutica* extract is shown in table 1.

Table 1. Colour darkness test results

Sample	No	Value	Wavelength (nm)	R%	T%
Lime	1	43.66	660.50	46.80	53.20
	2	47.52	659.50		
	3	49.22	656.00		
Alum	1	37.29	636.50	37.80	62.20
	2	39.37	649.50		
	3	36.73	639.00		
Limestone	1	17.07	624.00	18.60	81.40
	2	19.87	627.50		
	3	18.87	620.00		
Ferrous Sulphate	1	3.50	419.50	3.41	96.59
	2	3.11	430.00		
	3	3.63	426.00		

Colour darkness can be measured from the maximum wavelength, i.e. the wavelength with the smallest reflectance value (R%). Smaller value of R% resulted in darker colour of fabric, whereas higher value of R% gave lighter colour of fabric. After converted into percentage, the maximum wavelength is obtained in the highest transmittance (T%), it is therefore higher T% resulted in darker colour of cotton fabric. Opponently, lower T% generated lighter colour of cotton fabric.

Table 1 shows that reflectance percentages (R%) were in sequence of ferrous sulphate, limestone, alum, and lime at 3.41; 18.60; 37.80; and 46.80, respectively. The darkest colour was obtained by cotton fabric dyed under ferrous sulphate mordant, while the lightest colour was generated by the dyeing of cotton fabric using lime mordant. The acid pH caused by the addition of lime resulted in lighter colour. Alum mordanting agent contains aluminium ion which formed weak coordination complexes with the dye. It tended to form quite strong bonds with the dye but not with the fibre [17]. It is therefore the dye was released off the fabric. This results are in accordance with the results obtained by previous researchers [18]. They revealed that the increase in colour intensity by the use of Ca and Fe metals was higher than that of Al metals. This is due to Aluminium has a weak complexity ability compared to other metals. The high colour intensity is closely related to the ability of colouring matter molecules to form metal complexes with positive charged mordant.

3.2. Colour difference

Colour difference test parameters was carried out using a chromameter, colours are quantified into L * a * b * notations, which is known as CIELAB. In the CIELAB colour unit, L * indicates lightness, while a * and b * show chromaticity coordinates. Hue was quantified using equation given by

McLellan et. al. [19]. In this study fabric samples were tested using a Konika minolta CR-400 chromameter. Data on chromaticity is shown in table 2.

Table 2. Chromaticity data of the dyeing of natural dyes of *Brachiaria mutica*

Sample	*L	*a	*b	ºHue	Colour
A1	57.88	-15.605	31.415	178,89	Yellowish Green
A2	73.65	-32.09	18.85	179,47	Green
A3	78.27	-35.06	14.65	179,60	Green
A4	74.845	-32.525	17.35	179,51	Green

Data on chromaticity of *Brachiaria mutica* dyes can be seen in table 2. Symbol A1 refers to system with lime mordant, A2 refers to system with alum mordant, A3 refers to system with limestone mordant, A4 refers to system with ferrous sulphate mordant. Brightness levels of fabric that have been immersed in natural dyes of *Brachiaria mutica* are expressed in L*a*b* colour space. L* indicates lightness and a* and b* are chromaticity coordinates. a* and b* are colour directions: +a* is the red axis (from 0 to 60), -a' is the green axis (from 0 to -60), +b* is the yellow axis (from 0 to 60) and -b* is the blue axis (from 0 to -60).

The analysis results show that there were differences between the types of mordanting agent. Dye solution of *Brachiaria mutica* using lime juice as mordanting agent has the lowest brightness level of 57.88. While the *Brachiaria mutica* dye solution with alum as mordanting agent has a brightness level of 73.65. The variation of natural dyes powder with ferrous sulphate as mordanting agent has L value of * 74.845 resulted in darker colour than the dyes powder with limestone as mordanting agent which has an L * of 78.27. Higher L* value indicated that the resulting colour is brighter. L* values obtained in this study ranged from 57.88 to 78.27.

The notation a * and b * are chromaticity coordinates, the value +a * indicates the direction of red and -a * indicates the direction of green. The average value of a* dyes resulted in this study was in the range of -15.605 to -35.06. Cotton fabric dyed using *Brachiaria mutica* dye powder under limestone as mordant has a* value of -35.06 and fabric dyed using *Brachiaria mutica* dye powder under ferrous sulphate as mordant has a * value of -32.525. While fabric dyed using *Brachiaria mutica* dye solution under alum and lime juice as mordanting agents provided a * values of -32.09 and -15.605, indicating green and yellowish green in the sample.

The b * notation represents the chromatic colour in direction of blue and yellow. A positive value of b* (+ b *) from 0 to 60 and a negative value of b* (-b *) from 0 to -60. The average value of b* in this study was in the range of 14.65 to 31.415. *Brachiaria mutica* dye solution with lime juice has the highest b* value of 31.415. *Brachiaria mutica* dye solution with alum mordant has a value of b* which was almost the same as dye powder of *Brachiaria mutica* using ferrous sulphate mordant, while the dye powder of *Brachiaria mutica* with limestone mordant has the lowest b* value of 14.65. The results obtained from the measurement of b* notation revealed the variations of green and yellowish green in the sample. The greater value of b * indicated the more yellow sample colour. Based on the L* value, a * value, and b * value, the ºHue value can be obtained. Based on the data in table 2, it can be seen that ºHue value was in the range of 178.89º to 179.60º. The ºHue value indicated that *Brachiaria mutica* natural dyes generated a green colour.

3.3. Colour fastness

Mordant is also important in increasing colour fastness to washing by improving dye uptake. Chemical bonding of fibre and dyes is able to increase colour fastness. Chemical bonding of metal ion in the mordanting agent and fibre enlarges dye molecules thus causing difficulties to come out of fibre pores, results in high colour fastness to washing. Some transition metals, i.e. stannous chloride, ferrous sulphate, alum, chrome and copper sulphate have been employed as mordant for their strong coordinating power capable of giving weak to medium attraction or interaction forces. The mordant roles as bridging materials thus create substantivity for the subsequent fibre. Combination of metallic salts and fibre is able to form insoluble precipitates that fix both the dye and mordant into fibre, results in better colour fastness. Mordanting agents play an important role in ensuring colour brightness and wash fastness of the fibre as well as determining the final colour obtained [20]. Moreover in case of cotton that is more difficult to dye than wool or silk due to the absence of amino and carboxyl groups that provide attachment sites to dye molecules, the utilisation of mordant is a mandatory [5]. Investigation on characteristics of colour fastness to washing was done to each mordanting agent. The results are presented in table 3.

Table 3. Colour fastness to washing test results

Sample	Colour Fastness to Washing		Colour Staining	
	No	Value	No	Value
Lime	1	3	1	4
	2	3	2	4
	3	3	3	4
Alum	1	3-4	1	4
	2	4	2	4
	3	4	3	4-5
Limestone	1	4-5	1	5
	2	4-5	2	5
	3	4	3	4-5
Ferrous Sulphate	1	5	1	5
	2	5	2	5
	3	4-5	3	5

Fibre absorption to dyes is increased by application of mordanting agent thus leads to the improvement of colour fastness to washing. Colour fastness to washing is also governed by bonding of fibre and natural dyes. Pre-treatment with lime juice had registered fair resistance to colour change with good staining on cotton fabrics. Good resistance to colour change was found in the application of alum as mordanting agent and performed good staining on cotton fabrics, while utilisation of limestone resulted in good to excellent resistance to colour change with excellent resistance to staining on cotton fabrics. The best performance of colour fastness to washing was achieved by system using ferrous sulphate as mordant. Excellent resistance to colour change as well as excellent resistance to staining on cotton fabrics was obtained.

As mordanting agent, limestone gave positive effect by the role of calcium ion (Ca2+). It formed a bond with flavonoid in the fibre that produces a bigger complex molecule. The enlargement of complex dyes molecule resist the molecule to come out from the fibre after dyeing process that improve the colour fastness. In general, mordant can increase colour fastness of fabrics dyed with *Brachiaria mutica*. The ability of metallic mordant to form two or more bonds with dye molecules can increase colour fastness to washing. Application of ferrous sulphate resulted in the best colour fastness to washing. This is supported by Failisnur et.al. [18] who revealed that Fe and Ca tend to form stronger molecule complex than that of Al.

4. Conclusion

A research to produce natural dyes powder has been carried out. The obtained natural dyes powder was then applied in the dyeing of cotton fabric. Under quality parameters of colour darkness, colour fastness, and colour difference, some results are presented. *Brachiaria mutica* generated green colour on cotton fabric. It was found that based on the mordanting agents, the darkest colour were in the order of ferrous sulphate > limestone > alum > lime juice. In the perspective of colour difference, only lime juice mordant resulted in yellowish green colour, while the other mordants, i.e. alum, limestone, and ferrous sulphate produced green colour. In term colour fastness to washing, the best result was obtained by the system using ferrous sulphate as mordant, while lime juice mordant gave the lowest colour fastness to washing indicated by fair resistance to colour change and good resistance to staining on cotton fabric.

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