Utilization of black tea waste as natural batik dyes on cotton and silk

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Abstract. The production of bottled tea beverages generates a large amount of black tea waste, which may cause serious environmental problems without proper handling. Black tea waste has potential as natural batik dyes due to the tannin contains. In this study, the potential of black tea waste to dye batik on cotton and silk has been investigated. Black tea waste was extracted using water as the solvent with ratio 1:5 of material to solvent for an hour at 100 °C. The dyeing performance of the extracts was evaluated by measuring the K/S value and the chromatic values CIELab. The fastness properties of the dyed samples and the effect of different mordant type on dyeing quality were also studied. The fastness properties of the samples were in the range of good to excellent. The results show that there is a strong possibility to utilize black tea waste as natural batik dyes on cotton and silk.

1. Introduction
Batik is a traditional Indonesian textile crafted using wax-resist dyeing techniques. In earlier days, batik was coloured using natural dyes extracted from plants, but after the development and commercialization of synthetic dyes, batik was mainly produced with naphthol (azoic) and solubilized vat dye, because both of them can be applied under cold condition [1]. However, some synthetic dyes, particularly azo dyes which are prepared from -arylamines, are potentially carcinogenic [2]. The developing agents for Naphthol are also on the priority list of harmful chemicals, and their use is forbidden [3].

Moreover, almost all of the synthetic dyes are petrochemical-based and manufactured through hazardous chemical processes. Thus they possess threats to the environment [4]. Those factors prompt the growing interest in the uses of natural dyes, which never erode completely [5], as well as its research and development in batik production processes. The use of natural dyes provides various advantages because of their biodegradability and renewability, as well as their reduced toxicity and allergenicity [6].

Natural dyes can be defined as dyes and pigments obtained from various parts of plants or animals [2]. Almost all parts of plants, including the stem, wood, roots, bark, leaves, flowers, seeds, and fruits, can be extracted to produce dyes. From the animal kingdom, the exudation of dried bodies of a few insects and mollusks can act as colouring substances [6,7]. Besides, some agricultural waste has also been used as raw materials for natural dyes. To reduce the amount of agricultural waste and convert it into added-value products, the research on dye pigments from agricultural waste is of particular interest [6]. Previous studies have been conducted to utilize agricultural waste such as seaweed [8], rambutan peels [9], cacao peels, and palm kernel shell [10] as natural dyes for batik.

Tea (Camellia sinensis) has become the world's most popular beverage after water, and its industry involves more than 13 million people around the world [11]. Black tea is the most consumed tea besides
green and oolong tea, driving around 60% of the production. In recent years, to support the expansion of the demand, diversification into other segments of the market have been widely encouraged, including canned and bottled tea-flavored beverages [12]. During the manufacturing process of bottled tea beverages, large amounts of black tea waste are generated, providing an abundant source of raw materials for natural dyes production. Previous studies show that approximately 30% of dried tea leaves weight are made up of flavonoids, flavonols, and phenolic acids. Most of the polyphenols present are flavonols, commonly known as catechins, which are valuable natural pigments. Catechins in tea leaves include epicatechin (EC), epigallocatechin (EGC), epicatechin gallate (ECG) and epigallocatechin gallate (EGCG) [4,6]. These innate natural chemicals can be used as colouring agents in textile dyeing.

Some studies demonstrate the use of tea leaves and its waste as natural textile dyes on cotton [4, 6, 13-16], jute [13], wool [16, 17], silk [16, 18, 19] and linen [20]. However, to the best of our knowledge, little attention has been paid to the use of black tea leaves to colour textile with batik techniques. Therefore, the objective of this study was to investigate the feasibility of the utilization of black tea waste from the bottled tea beverage industry as natural batik dyes. The dyeing performance of the extracts was evaluated by measuring the K/S value and the chromatic values CIELab. The fastness properties of the dyed samples and the effect of different mordant type on dyeing quality were also studied. The dyeing experiments were conducted on cotton and silk as the most used fabrics for batik production.

2. Methods

2.1. Materials

Wet black tea waste (Camellia sinensis var. assamica) was received as a gift (ca. 10 kg) from PT. Sinar Sosro (Ungaran, Central Java, Indonesia). The wet black tea waste was obtained from the extraction of fermented tea leaves with water at 90-100 °C for 30 min. Commercially bleached cotton and silk as well as alum, lime, ferrous sulphate as mordants were purchased from Prawoto shop, Yogyakarta, Indonesia, and batik wax, bee wax, anionic wetting agent (TR Oil, i.e., sulphonated castor oil), non-ionic detergent of commercial-grade obtained from locally available sources were used as and when required.

2.2. Methods

2.2.1. Extraction of natural dyes. About 10 kg of black tea waste was air-dried under shade until its weight remained constant. They were then stored in black plastic bags in a dry place. To an extraction pan, 5 kg of the black tea waste was added and then extracted using 25 L water. The mixture was stirred, boiled and kept at boiling point for 60 min, allowed to stand for another 15 min and finally filtered to remove the impurities. The filtrate was then used as natural dyes.

2.2.2. Preparation of the fabrics. Cotton and silk fabric were soaked in 2 g/L non-ionic detergent solution for 12 h. The fabrics were then rinsed and air-dried (indoors) at room temperature. To optimize dye adsorption into the commercial-grade, they were boiled and stirred with a mixture containing 2 g/L soda ash and 5 g/L alum for 60 min and allowed stand for another 12 h. They were then rinsed and air-dried under shade.

2.2.3. Batik wax application. Batik wax was heated on special stove until melted at 80 °C. The liquid wax was applied onto the fabrics using the batik stamp. The fabrics were then cut into 25 cm × 25 cm pieces. This process can be seen at Figure 1 – 3.

2.2.4. Dyeing. The dyeing experiments were conducted in cold temperatures to prevent the wax melting. In each experiment, the dyeing process was conducted for both sides of fabrics. Batik pieces were soaked in dyes solution for 15 min for each side to ensure that the dyes were distributed evenly into all parts of the surface of the fabric. The fabrics were then air-dried under shade. These steps were repeated five
times to make dyes molecules penetrate into the fabrics strongly. Three mordants were used in this study, namely alum, lime, and ferrous sulphate. 5 g/L of each mordant was added into warm water to make mordant solutions. After dyeing process, fabrics were soaked in a mordant solution for 10 min then rinsed and air-dried under shed.

**Figure 1.** Stamp batik stove to melt the wax.  
**Figure 2.** Batik Stamp.  
**Figure 3.** Wax application onto fabrics.

2.2.5. *Wax Removal.* Fabrics were boiled and stirred in hot water until the wax was removed. Soda ash was added into the water as an emulsifying agent. The fabrics were then rinsed and air-dried under shed.

2.2.6. *Assessment of colour fastness.* Assessment of colour fastness was conducted in Textile Testing Laboratory in the Center for Handicraft and Batik, Yogyakarta, Indonesia. The dyed batik samples were tested using Indonesian national Standard methods. The specific tests were: colour fastness to washing (SNI ISO 105-C06: 2010), colour fastness to light (SNI ISO 105-B01: 2010), and colour fastness to rubbing (SNI ISO 105-A03: 2010). The changes were related to the standard gray scale (marks 1–5, 1 = very poor, 5 = excellent).
2.2.7. CIELab. The CIELab values of the dyed batik samples were measured with a colorimeter. The colors are given in CIELab coordinates, \( L \) corresponding to the brightness (100 = white, 0 = black), \( a \) to the red–green coordinate (positive sign = red, negative sign = green) and \( b \) to the yellow–blue coordinate (positive sign = yellow, negative sign = blue).

3. Results and discussion

3.1. Colour fastness to washing, light, and rubbing

Data for colour fastness to washing, light, and rubbing of cotton and silk fabric dyed with natural dyes from black tea waste in presence of different mordants are illustrated in Table 1. Mordants are usually a metallic salt which has affinity for both the colourants and fibers. Therefore mordants are required in textile colouring with natural dyes because they help the dyes get fixed on the fabrics [5]. In this study three most common used mordants in the batik industry were employed, namely ferrous sulphate (\( \text{FeSO}_4 \)), lime (\( \text{Ca(OH)}_2 \)), and alum (\( \text{Al}_2(\text{SO}_4)_3 \)).

| Fabric | Mordants         | Washing fastness | Light fastness | Rubbing fastness |
|--------|------------------|------------------|----------------|------------------|
| Cotton | \( \text{FeSO}_4 \) | 4-5              | 1-2            | 4-5              |
|        | \( \text{Ca(OH)}_2 \) | 4                | 1              | 4-5              |
|        | \( \text{Al}_2(\text{SO}_4)_3 \) | 4-5              | 1              | 4-5              |
| Silk   | \( \text{FeSO}_4 \) | 4                | 4-5            | 4-5              |
|        | \( \text{Ca(OH)}_2 \) | 4                | 4              | 4-5              |
|        | \( \text{Al}_2(\text{SO}_4)_3 \) | 4-5              | 4-5            | 4-5              |

3.1.1. Fastness to washing. The washing performance of dyed textiles depends on numerous factors, such as dye chemistry, size, and solubility of dye, nature of dye–fiber attachment, dye–solvent interaction, location of dye on fiber structure and detergent formulation used in washing [21]. As can be seen from the data, both cotton and silk samples had good to excellent washing fastness for the three mordants. It might be caused because tea extracts have low molecular weight and have tendency to aggregate inside the fibers [20], so they show good washing fastness. The fastness improvement might also attributed to the formation of the large insoluble complex by the colouring component present in those colourants and the metal ions within the fiber [1]. The affinity of terms component caused by hydrogen bonding and Vander Waals forces for the dyed samples might also help in term of washing fastness [22]. There is no significant difference of fastness between different mordants for both fabrics.

3.1.2. Fastness to light. The light-fastness is the resistance of dyestuffs to the influence of light energy, especially the ultraviolet (UV) part of the electromagnetic spectrum. It is subdivided into UVA (320–400 nm), UVB (280–320 nm), and UVC (100–280 nm). The shorter the wavelength, \( \lambda \), the higher is the energy \( E \) and the dyestuff damage [23]. The data in Table 1 shows that the cotton samples had very poor to poor light fastness for all mordants. In contrast, silk samples displayed good to excellent value of lightfastness. The lightfastness is related to the materials on which dyes are applied. Fading on cellulose fiber like cotton are caused by an oxidative process, while on protein fibers such as wool and silk, the process has a reductive nature. The chromophore in tea leaves is resistant to photoreduction thus it showed high light fastness in silk. The reverse occurs in cotton.

3.1.3. Rub Fastness. Rubbing fastness means a change in colour of the rubbed textile (by bleeding and fading) [23]. From table 1, it can be observed that. Moreover, the overall rub fastness parameters are rated very good to excellent. These results can be explained by the presence of tannins in the black tea.
extract. Combined with the strong ionic bonds established between the dye and mordants on, they produced the observed fastness levels [24].

![Figure 4. Change of CIELab coordinates obtained from different mordants on cotton dyed using natural dyes from black tea waste.](image1)

|          | FeSO4 | Ca(OH)2 | Al2(SO4)3 |
|----------|-------|---------|-----------|
| L*       | 79.75 | 83.05   | 89.65     |
| a*       | 2.96  | 3.24    | 1.12      |
| b*       | 7.44  | 7.82    | 5.39      |

![Figure 5. Change of CIELab coordinates obtained from different mordants on silk dyed using natural dyes from black tea waste.](image2)

|          | FeSO4 | Ca(OH)2 | Al2(SO4)3 |
|----------|-------|---------|-----------|
| L*       | 68.79 | 78.37   | 76.33     |
| a*       | 6.15  | 6.04    | 6.32      |
| b*       | 13.10 | 14.82   | 15.49     |

3.1.4. **Color Shade.** In a dyeing process, the formed group of dyes enables the dyer to produce a wide variety of shades. The use of different mordants results in variation of shade and color depth. The colors of samples with different mordants were evaluated in terms of L* a* b* values using CIELab. L* values represent lightness/darkness, a* signifies redness/greenness, and b* value indicates yellowness/blueness [5].
Figure 4 displays the change of shade and color depth obtained on cotton with different mordants. By variation of the mordant from iron sulfate to alum on cotton, the lightness increased from 79.75 to 89.65, indicating that mordanting with iron sulfate provides the darkest shade, followed by lime and alum. The positive value of a* shows that all mordants gave red shade rather than green, with the highest red shade obtained by lime, followed by iron sulfate and alum. A similar trend was observed for b* value, indicating yellow shade.

Figure 5 illustrates the results of the CIELab test of the silk samples. The L*, a*, and b* values with different mordants show positive value of L*, which signifies lighter shades, while positive a* and positive b* represent red and yellow. The lightest shade was obtained by lime, followed by iron sulfate and alum. The three mordants obtained similar value of a*. The b* values shifted from the lowest to the highest for iron sulfate, lime to alum, respectively.

4. Conclusions
The feasibility of utilization of black tea waste as natural batik dyes has been investigated. Based on the results of colour fastness properties, black tea waste can be applied as natural batik dyes on silk. However, some measures need to be taken for its application on cotton to improve the lightfastness. The use of different mordants did not affect colourfastness significantly but provided different shades of color. The color shades obtained are red-yellow.

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