Biduri (*Calotropis gigantea*) leaves extract as natural dyes and ultraviolet protector applied on silk fabric with an exhaust dyeing method

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Abstract. Biduri (*Calotropis gigantea*) leaves extract can be natural dyes and ultraviolet protector agents on the silk fabric. In this research, silk fabric with ultraviolet protection performance was developed using biduri leaves extract in a simple dyeing process. This study aimed to obtain a colored silk fabric from biduri leaves extract with good UV protection properties. The methodology consisted of the extraction process using an aqueous method, dyeing process with exhaust dyeing method, and post mordanting with several mordant. Evaluation of dyeing results included color strength (K/S), color brightness (L*), color direction (a*, b*), ultraviolet protection factor (UPF), and durability after washing. The result showed brightness was inversely proportional to the color strength, and the direction of the resulting color were greenish and yellowish. Dyeing with dyes from the biduri leaves extract with FeSO₄ mordant were effectively reflected by the improvement of the UPF up to 31 from an initial value of 5.4. UPF value after 5 times washing down to 29.2 and at 26.8 washing 10 times. Biduri (*Calotropis gigantea*) leaves showed a high potential to be utilized as natural dyes and UV protector agents simultaneously on the silk dyeing process.

1. Introduction
Indonesia has huge biodiversity with a high potential for utilization, one of which is biduri. However, exploration and utilization of this plant have not been optimized. Biduri (*Calotropis gigantea*) is one of the shrub plants/reeds plants that could grow widely on the coast, the mountains, and even on hard and calcareous soil [1]. In the textile sector, especially in functional textile, the utilization of biduri plants still limited to the biduri fibres such as material for filler and insulative filler [2], oil-absorbing [3], and composite [4]. Another part that may potentially be used in textiles is leaves. Alkaloids, glycoside, saponin, fats and oil, tannin and phenolic, flavonoids are bioactive which contained in biduri leaves [5]. Flavonoids, tannin, and phenolic are commonly known as a natural pigment for dyeing textile material. Its purposes not only for dyes but also for enhancement of other functional properties such as antimicrobial and UV protector [6,7].

Ultraviolet light contained in solar radiation with wavelength 200-400 nm, is considered to be one of the causes of disease in humans, such as skin cancer, wrinkles and DNA cell damage [7,8]. Protection against ultraviolet radiation can be achieved by using various types of sunscreens and physical barriers, such as clothing. Clothing materials can be chemically treated to make them able to absorb or reflect ultraviolet radiation. One of the fabrics that can be used for ultraviolet protection is
silk fabric, however its ability to protect ultraviolet radiation requires more treatment to enhance the protection performance through the immobilization of such UV protector agents [7].

In previous research, one of genus of biduri, milkweed (Calotropis procera) leaves extract has been used as natural dyes to dye cotton and served as a potential source of natural dyes [8]. Moreover, Gajanan (2018) has reported biduri leaves extract has an ability to absorb in the entire UV range and considered as a proper wide spectrum sunscreen and also in anti-aging cream preparation. In this research, we aim to produce a colored silk fabric with good UV protection properties from biduri leaves extract using a simple and common dyeing method in the textile wet process. Evaluation to the silk fabric was carried out to its color value, UV protection performance, and durability after washing.

2. Materials and methods

2.1. The materials and apparatus

100% silk fabric that was ready to dye produced by Centre for Textiles, Bandung, Indonesia, with EPI 86, PPI 80, weight 67.11 g/m². Fresh biduri leaves were harvested from Bandung, West Java during January-February 2020. Al₂(SO₄)₃, FeSO₄, and CuSO₄ (Merck) were used as mordant. Teepol was used as a wetting agent. Aqua demineralization (Brataco) was also used for boiling, washing and dilution of whole processes.

The apparatus used in this research included: laboratory glassware (Iwaki Pirex), string hotplate (Heidolph), oven (Heraeus), analytical balance (Chyo JL-180), launder-o-meter (SDL Atlas), paddler and dryer (Werner Mathis AG), spectrophotometer (X-Rite Color-i7), spectrophotometer UPF (JASCO V-750). All works on this research were conducted in textile chemistry research laboratory and textile chemistry testing laboratory Centre for Textiles.

2.2. Extraction biduri leaves

Biduri leaves were collected and washed correctly with water. The leaves material was shade-dried for five days, roasted at 105°C for 1 hour, then uniformly ground using the mechanical grinder and passed through the sieve number 60. Biduri leaves powder were boiled by aqua demineralization in 100°C with liquor ratio 1 : 10 for 60 minutes and then filtered. The filtrate was then used in the dyeing process.

2.3. Dyeing procedure

The dyeing process using biduri leaves extract included dyeing, washing, rinsing, and mordanting. The dyeing process was conducted using a laboratory exhaust dyeing machine launder-o-meter at 4°C for 30 minutes with a liquor ratio at 1 : 30. In order to reduce the surface tension of the fabric, 2 mL/L wetting agent was added into dye bath. After 30 minutes process, the fabric washed and rinsed prior to the mordanting process. Furthermore, the fabric immersed into a solution containing 30 g/L mordant respectively and padded with 80% WPU for 3 dips and 3 nips, then drying on 100°C for 2 minutes. Finally, the fabric samples were washed thoroughly with cold water, squeezed, and dried.

2.4. Color measurement

As preparation for colour measurement, fabric samples were scanned with 350 dpi for recording the sample photographs. Color measurement procedures followed SNI-ISO 105-J03 2016 (Textile – Test for colourfastness – part J03; Calculation of colour difference). Color values were evaluated by means K/S and CIELab color difference value on spectrophotometer. K/S value was automatically assessed by kubelka-munk equation 1.

\[
K/S = \frac{(1-R)^2}{2R} \quad (1)
\]
where \( K \) is the light absorption coefficient, \( S \) is the scattering coefficient and \( R \) is the reflectance of the dyed samples at the wavelength of the maximum absorption.

### 2.5. UPF measurement and washing durability

Dyed silk fabric samples were prepared for UV protection factor (UPF) measurement, which was carried out by following a standard procedure AS/NZS 4399: 2017 (Sun protective clothing – Evaluation and classification) on wavelength interval 200 – 00 nm. UPF factor each sample determined by total transmittance spectra and automatically assessed by following equation 2.

\[
UPF = \frac{\sum_{1}^{400} E_\lambda S_\lambda A_\lambda}{\sum_{290}^{400} E_\lambda S_\lambda T_\lambda A_\lambda}
\]  

(2)

Where \( E_\lambda \) is CIE reference erythema dose spectrum, \( S_\lambda \) is the radiation intensity distribution of sunlight, \( T_\lambda \) is the diffuse transmittance spectrum (%), and \( A_\lambda \) is the bandwidth. UPF classification based on AS/NZS standard 4399: 2017 is shown in table 1.

**Table 1.** UPF classification based on AS/NZS standard 4399: 2017 [1,2].

| UPF rating | UV protection | UV blocked |
|------------|---------------|------------|
| 15-24      | Good          | 93.3 – 95.9% |
| 25-39      | Very good     | 96 – 97.4%   |
| 40-50, 50+ | Excellent     | > 97.5%     |

In order to find out the effect of multiple washing to UPF value, washing durability test for sample fabric was conducted following SNI-ISO 105-C06:2010 (Textile – Test for color fastness – part C06; Color fastness to domestic and commercial laundering).

### 3. Results and discussion

#### 3.1. Effect of mordant on color strength and color value

Table 2 shows the \( L^* \), \( a^* \), \( b^* \), and \( K/S \) value of silk fabric dyed with biduri leaves extract dye solution at different mordant. \( L^* \), \( a^* \), and \( b^* \) refer to the three-axis of the CIE-Lab color system. The \( L^* \) value indicated the perceiving of brightness with interval scale value from 0 (black) until 100 (white). The higher the \( L^* \) reading, the lighter the color. \( a^* \) and \( b^* \) value indicating color direction, +\( a^* \) for red, -\( a^* \) for green, +\( b^* \) for yellow, and -\( b^* \) for blue. The color strength value increased with the addition of dyes and mordant, while brightness decreased. Mostly, color direction of resulted silk fabric was greenish and yellowish, addition \( \text{Al}_2(\text{SO}_4)_3 \) and \( \text{CuSO}_4 \) mordant generated color direction to be brighter and darker, respectively. \( \text{FeSO}_4 \) mordant generated darkest color as the effect of conversion from part of ferro become ferri after its reaction with oxygen in the air. Ferro and ferri forms on silk fabric produced overlapping spectrum and changed the maximum wavelength, thus affected the color of silk fabric became darker [7].

Mordant was able to increase the complexity among color molecules and silk fabric, therefore the color intensity would also increase. As the result, mordanting usually generates darker color of fabric compared than that of fabric without mordant [12]. The darkest color was found in silk fabric which using \( \text{FeSO}_4 \) mordant, whilst the lightest was achieved in silk fabric which using \( \text{Al}_2(\text{SO}_4)_3 \) mordant. Silk fabric with \( \text{Al}_2(\text{SO}_4)_3 \) mordant was darker than that of undyed fabric, but lighter compared to fabric without mordant. This phenomenon can be explained as follows, iron and copper ions tend to form coordination complex between dye and amino acid or carboxylic groups, while as aluminum ion
tends to form bonding with dyes rather than fibre, therefore reduce interaction among dyes and silk fabric [7].

**Table 2.** Color strength and color values of dyed silk with biduri leaves extract.

| Mordanting agent | Mordant concentration (g/L) | Color strength (K/S) | Color value | Color obtained |
|------------------|-----------------------------|-----------------------|-------------|----------------|
| -                | Undyed                      | 0.74                  | 88.89       | 0.06           | 12.87          |
| Without          | -                           | 1.85                  | 82.95       | -1.21          | 23.9           |
| Al₂(SO₄)₃        | 30                          | 0.83                  | 88.35       | -1.43          | 14.63          |
| FeSO₄            | 30                          | 6.35                  | 69.75       | 5.06           | 25.17          |
| CuSO₄            | 30                          | 1.51                  | 83.12       | -7.62          | 1.51           |

3.2. **UPF value**

The UV protection factor value are presented in table 3. The UPF value on silk fabric after dyed result with and without mordant increased compared than that of standard fabric (undyed). Silk fabric naturally had a low UPF value and increased significantly after dyeing with biduri leaf extract. This indicated that silk fibre had little resistance to exposure to ultraviolet rays and biduri leaf extract can increase its UPF value significantly. The addition of mordant could relatively increase the UPF value of silk fabric colored with biduri leaf extract. Mordant can react with dye molecules and silk fabric, so that the dye fixed on the fabric. The result showed that the mordant used in this study could reduce the transmission of ultraviolet light through silk. However, based on table 3, the highest UPF enhancement found in the sample fabric which using FeSO₄ as mordant and classified as “very good” UPF.

3.3. **UPF after washing durability**

After the washed treatment, sample fabric run into decrease UV protection performance as shown in table 4. The highest decline occurred in the sample fabric without mordant down to 37.3% from initial UPF value after 5 times washed. This decline indicates the use of mordant is required in dyeing with natural dyes, thus dyes may bind to the fibre perfectly.
Table 3. UPF test result of silk dyed with biduri leaves extract.

| Mordanting agent | Mordant concentration (g/l) | UPF | Class | Enhancement (%) |
|------------------|----------------------------|-----|-------|-----------------|
| -                | undyed                     | 5.4 | No class | -               |
| Without          | -                           | 9.2 | No class | 70.37%          |
| Al₂(SO₄)₃       | 30                          | x6.4| No class | 18.52%          |
| FeSO₄           | 30                          | 31.175| Very good | 477.31%         |
| CuSO₄           | 30                          | 12.05| No class | 123.15%         |

Table 4. UPF test result of silk dyed with biduri leaves extract after washed.

| Mordanting agent | Mordant conc. (g/L) | UPF | UPF 5x washes | Decline (%) | Class. | UPF 10x washes | Decline (%) | Class. |
|------------------|---------------------|-----|---------------|-------------|--------|----------------|-------------|--------|
| without          | -                   | 9.2 | 6.7           | 37.3        | No class | -              | -           | -      |
| Al₂(SO₄)₃       | 30                  | 6.4 | 6.1           | 4.7         | No class | 5.8            | 9.4         | No class|
| FeSO₄           | 30                  | 31.2| 29.2          | 6.3         | Very good| 26.8           | 14.0        | Very good|
| CuSO₄           | 30                  | 12.1| 11.2          | 7.1         | No class | 8.0            | 33.6        | No class|

4. Conclusion

Biduri (*Calotropis gigantea*) leaves can be utilized as natural dyes and UV protector agents simultaneously on the silk dyeing process. The color strength and color values may change according to mordant used, which FeSO₄ mordant showed the most significant influence to color strength of the resulted fabric. Mostly, the color direction produced greenish and yellowish in whole resulted silk fabrics. Moreover, the use of FeSO₄ mordant significantly increasing UPF value up to 31.2 and keeping UPF value on “very good” classification after 10 times washing treatment.

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