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Research Article

Application of Purified Lawsone as Natural Dye on Cotton and Silk Fabric

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The color which is obtained from the leaves of Henna, that is, *Lawsonia inermis* L., is used frequently in hair coloring. It is the chemical lawsone that is responsible for the reddish brown color. Its content makes it a substantive dye for dyeing the textile materials. This work concerns with the extraction and purification of natural dyestuff from a plant *Lawsonia inermis* L. and dyeing of cotton and silk fabric in exhaust dyeing method. The dye portion is isolated from the total extract by column chromatography and is evaluated by dyeing cotton and silk under different dyeing conditions. The color strength and fastness properties of the dye are undertaken by changing mordant and techniques of mordanting. The changes of colors have been noticed by using different types of mordant. The dye exhaustion percentage, wash, rubbing, and light fastness results reveal that the extract of henna can be used for coloration of cotton and silk fabric.

1. Introduction

As compared to natural dyes, synthetic dyes have been widely used due to their lower prices and wide range of bright shades with considerable improved color fastness properties. The use of synthetic dyes has gained higher acceptability in the field of food [1], cosmetic [2], and more importantly textile industries [3] after the invention of synthetic dyes by William Henry Perkin in 1856 [4]. However, recently the concern for the environment has created an increasing demand for natural dyes. The ancient people used these dyes in cosmetics, food, leather, and also in medicine [5–7]. The use of natural dyes is found severely, when the weaving technique for dyeing textile materials is developed [8]. In the situation of environmental concerns, it is time to reconsider the use of renewable, nontoxic, and gentle colors from nature. Eco-friendly natural dyes are familiar substances that spark creative ideas. They could serve as a viable option for our dye needs. Indigo, the blue dye still used in jeans, is chemically the same as the dye extracted from the indigo plant. Natural dyes are very old, perhaps when people found that something changed the color of white fibers.

The worldwide demand for natural dyes is nowadays of great interest due to the increased awareness on therapeutic properties of the dyes. Natural dyes are derived from naturally occurring sources such as plants, insects, animals, and minerals. Several synthetic colorants have been banned because they cause allergy-like symptoms or are carcinogens [9–11]. Although known for a long time for dyeing as well as medicinal properties of the dye, the structures and protective properties of natural dyes have been recognized only in the recent past. Many of the plants are used for dye extraction.

The present study focuses on the development of the optimum extraction conditions of coloring component from the natural material Henna, that is, *Lawsonia inermis* L. and then the major component of the dye, that is, lawsone, is purified. The chemical structure of lawsone is shown in Figure 1. Finally the purified dye is applied on cotton and silk fabric by exhaust dyeing method. The color strength value, dye exhaustion %, and color fastness properties of the dyed fabrics are evaluated and compared with each other.

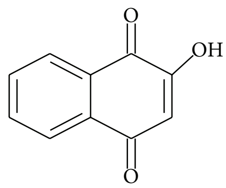

Figure 1: 2-Hydroxy-1,4-naphthoquinone.

2. Materials and Experimental Methods

2.1. Dyestuff Used. Henna, that is, *Lawsonia inermis* L., is used to obtain the dyestuff of lawsone, which is reddish brown in color. Henna is collected from the local market of Bangladesh.

2.2. Fabrics Used. Cotton and silk fabrics selected for the present study are collected from Sinha Textile Group, Bangladesh. Cotton fabric is scoured-bleached and degumming is done for silk fabric before dyeing.

2.3. Chemicals Used. Ethanol (C_2H_5OH), chloroform ($CHCl_3$), acetone (C_3H_5OH), and n-hexane (C_6H_{14}) are used as solvent to extract the dye from *Lawsonia inermis* L. The chemicals are collected from Sigma- Aldrich GmbH and Carl Roth GmbH. They were pure grade chemicals used for laboratory purposes.

2.4. Mordants Used. The following mordants are used: potassium aluminium sulphate ($AlK(SO_4)_2{\cdot}12H_2O$), copper sulphate ($CuSO_4$), and tartaric acid ($C_4H_6O_6$). They were pure grade chemicals used for laboratory and are collected from Carl Roth GmbH.

2.5. Solvent Extraction of Dye from Henna Powder. Plant tissues are difficult to extract because of active lipases which hydrolyze rapidly phospholipids and glycolipids and increase the amount of free fatty acids in the extract. Thus a solvent extraction is preferred to inhibit these enzymes.

Fresh leaves of henna are washed and cleaned with distilled water. They are then dried in the sunlight for one day and again dried at 80°C for one hour in a hot air oven. Dried leaves are made in powder form for getting proper extraction result. 10 gm of henna powder is taken into a thimble and placed in a soxhlet apparatus. 200 mL of solvent is added in soxhlet for 10 gm of sample and the extraction is carried out according to the boiling point (B.P.) of solvent for 3 hours. After extraction the brown extract is cooled, filtered, and concentrated by evaporation. Finally, a crude dried extract is obtained, which is brown in color. The dye is extracted from henna using different solvent according to their boiling point by the same method and yield is calculated. Finally color strength % of extracted dye is compared for getting the best extraction result.

2.6. Purification of Lawsone Dye from Extract by Column Chromatography. The column chromatography consists of a glass tube with a diameter of 30 mm and a height of 70 cm with a tap at the bottom. The column is clamped vertically. The stopcock of the column is closed and it is fitted with fritted disk. The silica gel is weighed out in a flask and enough solvent (eluent) is added with stirring to form slurry. Then, slurry is slowly transferred into the column using a glass funnel until the silica gel level is about 30–40 cm. When the solvent reached the silica gel surface, we slowly added the mixture solution of henna extract into the column using a dropper. After the sample is loaded, a small layer of sand is added to the top of the column. This helped to keep the top of the column level when added solvent eluent. Once the mixture is added to the column and the protective layer of sand is placed, the solvent (eluent) is added continuously. Further the small fraction of the eluent is collected from the bottom of the column. The components are retained by the stationery phase and the lawsone part is separated through the column with the eluent.

2.7. Optimization of Dyeing Conditions for Cotton and Silk Fabrics. Cotton and silk fabrics are dyed with the lawsone dye at a liquor ratio of 1 : 40. For optimizing the dyeing conditions, at first, experiments are carried out to optimize the dyeing pH. Dyeing processes are carried out with 2% (o.w.f) concentration of purified dye at pH 3, 5, 7, and 9. To understand the effect of dyeing temperature and dyeing time on the color strength, another set of experiments is carried out in optimized dyeing pH at 60, 75, 90, and 100°C for different time periods, that is, 30, 45, 60, and 90 minutes. Another set of experiments is also carried out at different dye concentrations such as 2%, 4%, 6%, and 8% in optimum dyeing conditions. Based on the color strength (K/S) values of the dyed samples, optimum dyeing pH, temperature, and time are selected and taken for further study. To study the effect of different mordants and their varying concentrations on color strength, mordanting is carried out with three metallic salts such as aluminium potassium sulphate, copper sulphate, and tartaric acid. Mordanting is carried out through two ways: (1) premordanting and (2) postmordanting. The sample is taken as 1 gm for every experiment.

After dyeing, the dyed samples are rinsed with cold water and washed in a bath with a liquor ratio of 1 : 50 using 1 gm/liter of the soaping agent at 60°C for 10 minutes and then they are rinsed and finally dried in a dryer.

2.8. Evaluation of Color Strength. Estimation of color strength of the dyed fabrics is carried out by determining the K/S values using a computer color matching system (CS-5, Applied color system, USA). The reflectance value (R) in the visible wavelength region is measured by means of the ACS spectrophotometer. The value of reflectance (R) of the dyed fabric is measured at the wavelength of 420 nm and also the K/S value of the sample is found directly from the instrument. Every dyed sample is measured in the same way and the K/S values are obtained directly from the instrument, which followed the Kubelka-Munk theory as

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

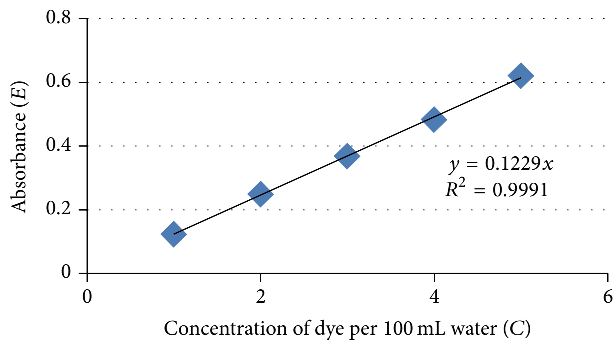

FIGURE 2: Calibration curve of lawsone dye.

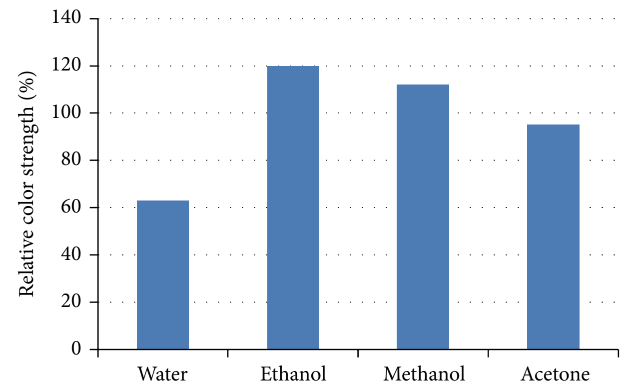

FIGURE 3: The relative color strength of henna extract with different solvent.

where K is absorption coefficient of the sample, S is scattering coefficient of the sample, and R is the degree of reflection. The value of K/S is directly proportional to the concentration of dye in the dyed fabric.

2.9. Measurement of Exhaustion of Dyes by UV/VIS Spectroscopy. Degree of exhaustion is the amount of dyestuff, which is diffused in the fiber from the dye bath at the time of dyeing. UV/Vis spectrophotometer (DT A 01, Perkin Elmer, Singapore) is used to measure the exhaustion and fixation of the dyestuff. By measuring the concentration of dye bath before and after the dyeing process, the percentage of exhaustion can be estimated with (2) considering the color concentration of mordants:

$$E\% = \frac{C_1 - C_2}{C_1} \times 10, \tag{2}$$

where C_1 and C_2 are the concentrations of the dye bath before and after dyeing process, respectively.

The concentrations of the dye solution before and after dyeing are measured using UV/VIS spectrometer at the wavelength of 420 nm. Before measuring the absorbency, the wavelength of maximum absorbency is determined for the dye by using the calibration standard solutions. For the calibration standard solutions, a dye stock solution of 20 mg in 20 mL water is prepared. By pipetting 1 mL to 5 mL of this stock solution is taken and diluting to 100 mL, 5 different calibration standard solutions are prepared. The concentrations of the calibration standard solutions ranged from 0.01 g/L to 0.05 g/L. The dye solution is taken into a cuvette with a width of 1 cm and subsequently the absorption spectrum is recorded, which are presented in Figure 2.

The value of extinction coefficient is obtained from the linear regression line ($y = mx$), where the value of "m" corresponds to the extinction coefficient of the dyes. The extinction coefficient of the dye is determined as per Beer-Lambert Law, based on (3):

$$\log \frac{I_0}{I} = E = \varepsilon c d, \tag{3}$$

where I_0 is intensity of the initial light, I is intensity of the light after passing through the dye liquor, E is extinction at a specific wavelength of λ, ε is extinction coefficient, c is concentration of the dye liquor, and d is cuvette width.

2.10. Evaluation of Color Fastness Properties. Wash fastness of the samples dyed under the optimized conditions is tested according to ISO 105-CO3 method. ISO 105-X 12 test methods are followed to measure the rubbing fastness. Color fastness of textile material to day light is of considerable importance to the consumer. The specimen should be tested according to ISO test method. The light fastness of the dyed test samples (1 × 3 cm) is exposed to UV light of a Xeno tester for 40 hours. The temperature of the machine was 30°C, R.H. 65%, and irradiation dose 980 KJ/m^2. The fastness is assessed by comparing the fading of the specimen with that of blue wool patterns.

3. Results and Discussion

3.1. Optimization of Extraction Conditions and Purification of Lawsone Dye. Soxhlet extracts are weighted after drying and color strength percentage of lawsone dye is calculated; those are shown in Figure 3. The relative color strength of dye extracts obtained in distilled water is not appreciable, which is also shown in [12]. Maximum amount of lawsone dye is obtained by using ethanol. So the ethanol extract is used to purify the dye and then dyeing of cotton and silk fabric is performed with the purified dye.

The ethanol extracted dye is subjected to column chromatography by using the mobile phase of chloroform and n-hexane and the fractions are collected and tested by thin layer chromatography (TLC). The absorbance of the purified lawsone dye is analyzed by UV-visible spectroscopy at 420 nm. The test showed that the lawsone dye is separated perfectly by the mixture of chloroform and n-hexane in the ratio of 4 : 1.

3.2. Effect of Dye Concentration on Color Strength of Cotton and Silk Fabrics. The color strength of the dyed cotton and silk fabrics is dependent on the concentration of the extracted lawsone dye as shown in Figure 4. The color of the dyed cotton and silk fabrics is reddish, when the percentage of dye (o.w.f) is less than 2. When the percentage of dye is more than 2, the fabrics showed reddish brown color. From the results, it is seen that the color strength, expressed as percentage,

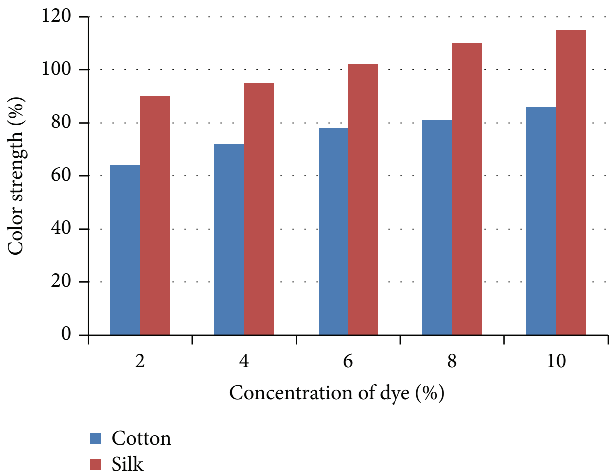

FIGURE 4: Effect of concentration of lawsone dye on color strength of cotton and silk fabrics.

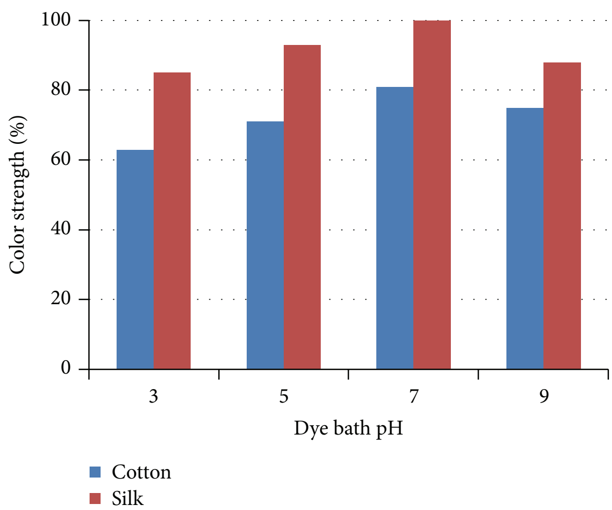

FIGURE 5: Effect of pH on color strength of cotton and silk fabrics dyed with lawsone (4% shade).

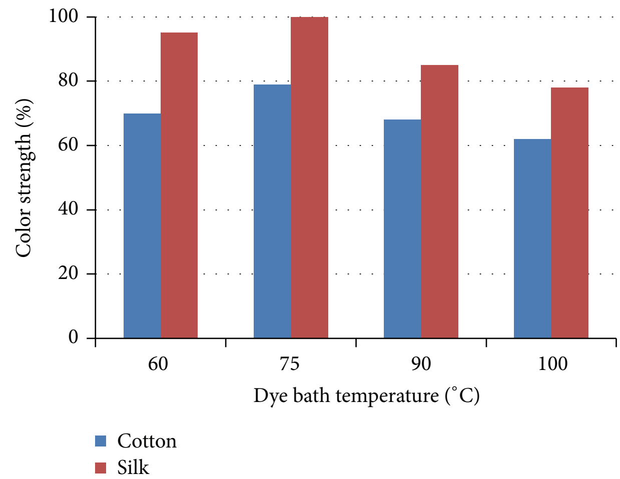

FIGURE 6: Effect of temperature on color strength of fabrics dyed with lawsone dye (4% shade).

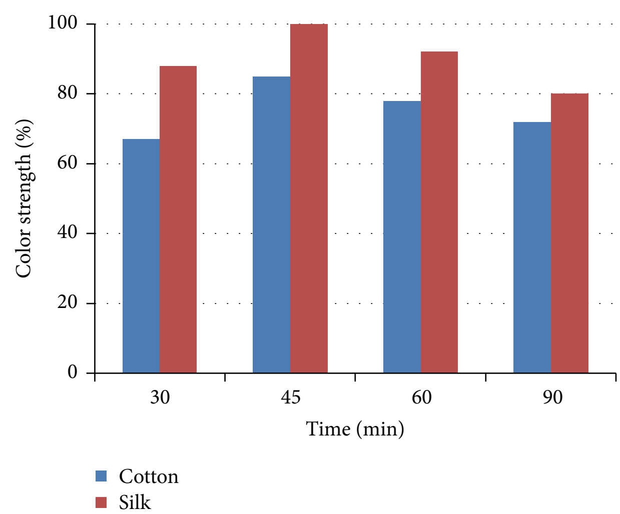

FIGURE 7: Effect of time on color strength of fabrics dyed with lawsone dye (4% shade).

increased with increasing concentration of lawsone dye up to a certain limit. It is also observed from the result that there is a significant difference in the depth of shade between cotton and silk fabrics, though the dyeing condition and concentration of dye are the same. It is found that the color strength of the silk fabric is higher than the cotton fabric at the same concentration. The color strength of silk is more because of its functional groups in its structure.

3.3. Effect of pH on Color Strength of Fabric. Figure 5 showed that the color strength is increased with the increase of pH value in the dyeing bath from pH 3 to 7 for both the fabrics. The maximum dye uptake is obtained at pH 7 for both. The maximum dye uptake of cotton fabric is also found at neutral pH in [12], which is almost similar to the obtained result, though the dye extraction condition is not the same.

3.4. Effect of Temperature on Color Strength of Fabric. The effect of dyeing temperature on color strength of cotton and silk fabric is demonstrated in Figure 6. As evident, the maximum color strength is obtained at 75°C for both cotton and silk fabrics. The shade is also very uniform. The maximum color strength of cotton dyed with henna dye is found at the temperature of 70°C in [12], which is very much similar to the obtained result. If the temperature is increased to more than 75°C, then the depth of shade is decreased. The result is also not good, if the fabrics are dyed less than 75°C. The same result is found in [12]. The depth of shade is more in case of silk fabric than cotton fabric at the same concentration and temperature because of the structure of the fiber.

3.5. Effect of Time on Color Strength of Fabric. The effect of dyeing time on color strength is shown in Figure 7. The best results with respect to time for dyeing cotton and silk fabrics

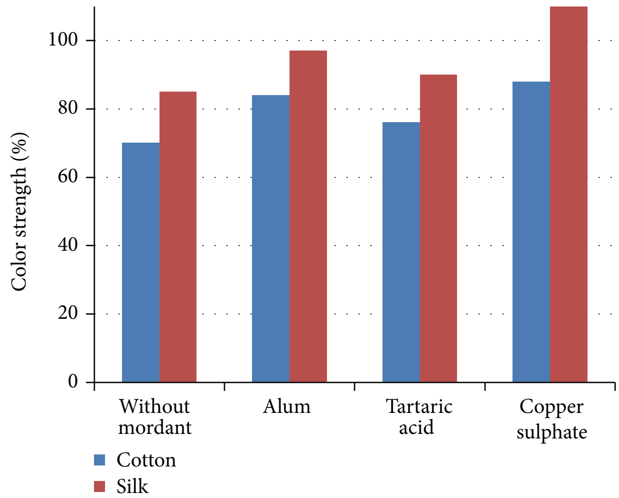

Figure 8: Effect of premordanting (2 gm/L) on color strength of fabrics (4% shade).

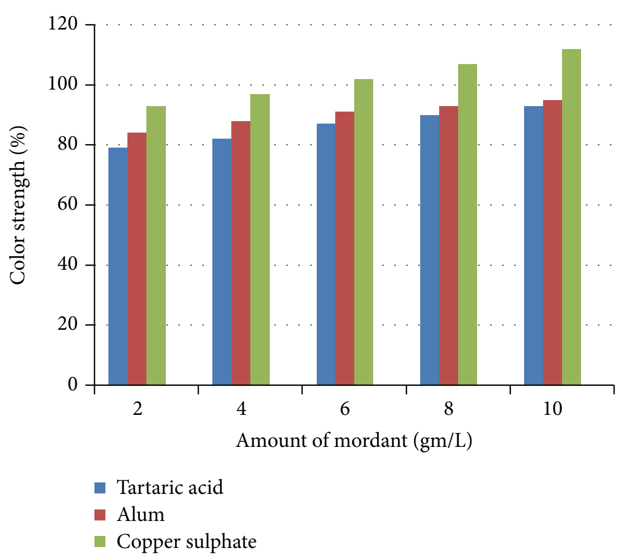

Figure 9: Effect of premordanting on color strength of cotton fabric (4% shade).

are obtained at 45 minutes. The color strength is decreased, when the fabrics are dyed one hour or more than one hour. The decrease in color strength may be attributed to the fact that the desorption of the dye molecules could happen as a consequence of over dyeing, if the process is carried out for more than 45 minutes. So, it can be said that the time for maximum exhaustion of dyes depends on some factors such as shade % and dyeing conditions.

3.6. Effect of Mordanting on Color Strength of Cotton and Silk Fabrics. The effects of mordant on relative color strength for the premordanting method are shown in Figure 8. It is observed that relative color strength values are higher for samples dyed with premordanted fabric at 100°C in comparison with samples dyed without mordanting. The premordanted fabric dyed with henna dye showed better results than unmordanted fabric, which is also demonstrated in [12]. Since the mordant has affinity for lawsone, it attracts more coloring component from dye bath to fabric. It is also observed in Figures 9 and 10 that the relative color strength is increased with increasing the mordant concentration and reached a maximum at 10 gram per liter. It is also seen that the effect of different mordants is not the same. The results of alum and copper sulphate are very good in terms of color strength value. But the deviations in shade tone and brilliancy among the abovementioned dyed sample cannot be measured in terms of color strength value. It is seen that different brilliant shade can be produced on cotton and silk fabrics with lawsone dye by using same amount of different mordants, which is also discussed in [12].

It is clearly seen in Figure 11 that the relative color strength is decreased by the application of mordant. The similar result is observed in [12], when the postmordanting technique is applied to dye the cotton fabric with henna dye. Thus, the premordanting method showed a higher depth of shade in comparison with the postmordanting method.

Copper sulphate, alum, and tartaric acid are well known for their ability to form coordination complexes and to readily chelate with the dye. As a result, these mordants helped to increase the color strength of the dyed fabric. It is also seen that tartaric acid ions form weak coordination complexes with dye than copper sulphate and alum; so the color strength of the dyed fabrics mordanted with tartaric acid is less than the dyed fabrics mordanted with copper sulphate and alum.

Table 1: Maximum exhaustion of lawsone dye by cotton and silk fabrics.

Fabric dyed with lawsone	Dye exhaustion (%)
Cotton without mordant	52
Cotton premordanted with tartaric acid	61
Cotton premordanted with $CuSO_4$	72
Cotton premordanted with alum	68
Silk without mordant	71
Silk premordanted with tartaric acid	77
Silk premordanted with $CuSO_4$	82
Silk premordanted with alum	80

3.7. Result of Dye Exhaustion. The highest degree of exhaustion of lawsone by cotton and silk fabrics, which are dyed in exhaust method, is determined by UV-Visible spectroscopy and the results are shown in Table 1.

The degree of exhaustion of silk fabric dyed with lawsone dye is found higher than that of cotton fabric as shown in Table 1. This is because of the molecular structure of the fiber. Silk fiber has more functional groups in its structure than cotton fiber. As a result, the dye exhaustion is more in case of silk fiber than the cotton. The results also showed that higher dye exhaustion occurred in mordanted cotton and silk fabrics than unmordanted samples.

Table 2: Fastness properties of cotton and silk fabrics dyed with lawsone dye.

Dyeing technique	Shade% (o.w.f)	Fastness rating: Wash fastness: Color change: Cotton	Color change: Silk	Staining (cotton): Cotton	Staining (cotton): Silk	Rubbing fastness: Dry rubbing: Cotton	Dry rubbing: Silk	Wet rubbing: Cotton	Wet rubbing: Silk	Light fastness (40 hours): Cotton	Light fastness (40 hours): Silk
Without mordant	2	3	3-4	3	3	3-4	3-4	3-4	3-4	3-4	3-4
	6	3-4	3-4	3	3-4	3	3-4	3-4	3	3	3
With tartaric acid	2	4-5	4-5	4	4-5	4	4-5	3-4	3-4	4	4
	6	4	4	3-4	3-4	3-4	4	3-4	3-4	3-4	3-4
With alum	2	4-5	4-5	4-5	4	4-5	4-5	3-4	3-4	4	4-5
	6	4	4	4-5	4	4	3-4	3	3	3-4	3-4
With copper sulphate	2	4-5	4-5	4-5	4-5	4-5	4-5	3-4	3-4	4-5	4-5
	6	4-5	4-5	4-5	4-5	4-5	4-5	3	4	4-5	4

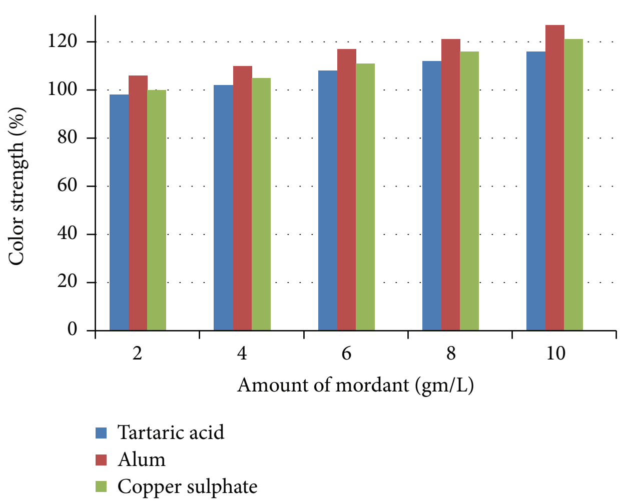

Figure 10: Effect of premordanting on color strength of silk fabric (4% shade).

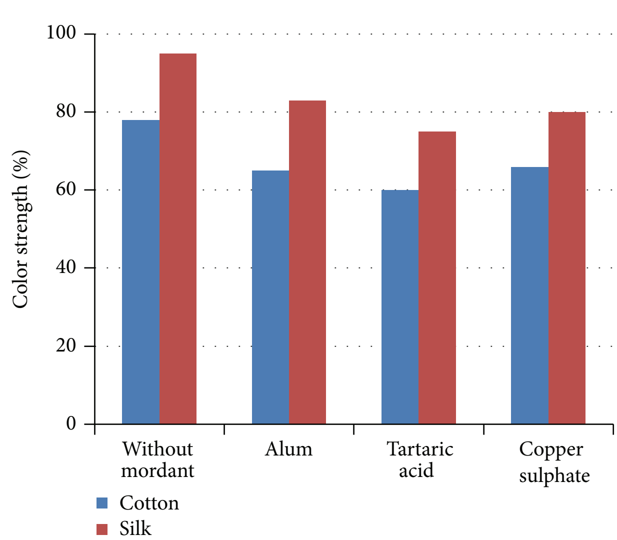

Figure 11: Effect of postmordanting (2 gm/L) on color strength of cotton and silk fabrics (4% shade).

3.8. Result of Fastness Properties. The wash fastness rating of cotton and silk fabrics dyed with or without mordants at the dye concentrations of 2% and 6% (o.w.f) is presented in Table 2. The result showed that the color fastness ratings of the cotton and silk dyed fabrics without mordanting are not good. The fastness properties have improved significantly, when different types of mordants are used before dyeing. The mordanting with copper sulphate demonstrated better result than the others. It is due to the formation of strong bond between the dye and fiber. It is observed that the light fastness of lawsone dyed fabrics is lower than other fastness properties. It is because of the chromophore group structure of the lawsone dye.

3.9. Economical Aspects for Dyeing with the Dyes of Natural Purified Lawsone. The cost of natural dye, that is, lawsone dye, is more than synthetic dye. But the auxiliaries cost and dyeing process cost of fabric by natural lawsone dye are less than synthetic dye. Moreover the natural dye is not harmful for environment, so effluent treatment plant is not necessary for such type of technology. So the technique can be applied commercially for dyeing protein and cellulosic fabric like silk and cotton.

4. Conclusion

The main goal of the work is to extract and purify the lawsone dye from the plant *Lawsonia inermis* L., which is used to dye the cotton and silk fabric to compare the properties of the dyed fabric. The result shows that the color strength values of silk dyed fabrics are higher than the cotton dyed fabrics, though they are dyed in same method. The dye exhaustion percentage of the silk fabric is also higher than cotton fabric. It is observed that mordant has a great effect to increase the color strength of the fabric and the effects of different mordants are not the same. Copper sulphate shows the better result than the other two. Depending on the dyeing procedure and the types of mordant used, a variety of colors also can be produced using the same percentage of lawsone dye. At the same time, the dye processing cost and the cost for effluent treatment plant (ETP) are also lower than that of other synthetic dyes. So, extraction and purification of natural dyes and their application can be of great significance in the future of the commercial and domestic dyeing industries.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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