colorants

Article

Dragon Fruit (*Hylocereus polyrhizus*): A Green Colorant for Cotton Fabric

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Abstract: The textile industry has been exploring sustainable chemicals and natural alternatives to replace harmful and carcinogenic substances used in different stages of textile production for dyeing textiles. Natural dyes are gaining popularity, as they are environmentally friendly and less harmful. Betacyanin, a type of pigment obtained from red pitahaya, commonly known as red dragon fruit (*Hylocereus polyrhizus*), has peels that are available as agricultural waste and can be used to meet the demand for natural dye production. This study aimed to explore and utilize dragon fruit's peel as a natural colorant for dyeing 100% cotton knit fabric (scoured and bleached single jersey plain knit) of 170 g/m^2, which could transform a low-value material into a valuable product. However, cotton's phenolic nature and oxidation process result in negative charges on its surface, making natural dyeing challenging. Cationization with cationic agents (ForCat NCH, a mixture of cationic polyamine and 1,3,dichlori-2-propanol) and mordanting (potassium alum or potassium aluminum sulfate) were carried to improve dye exhaustion and enhance colorfastness properties. Spectrophotometer 800 was used to measure color strength (*K/S*), and several fastness tests, including wash, perspiration, and rubbing were conducted to assess the final product's performance. The process parameters, such as temperatures, times, pH levels, and dye concentrations were varied to understand better the optimum conditions.

Keywords: dragon fruit; pitahaya; natural dye; eco-friendly; cotton; cationization; mordant

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1. Introduction

The recognition of environmental issues and the implementation of strict regulations have stimulated investigation into sustainable chemicals that can replace harmful and carcinogenic substances used in different stages of textile production, such as pretreatment [1], dyeing [2–4], printing [5,6], and finishing [7–9]. As an extension of this work, researchers are exploring natural alternatives for dyeing various textile materials that have traditionally employed toxic synthetic colorants [10]. Synthetic dyes damage the environment, as they are non-biodegradable and can cause pollution during their processing and application [10,11]. In contrast, natural dyes are viewed as more environmentally friendly and less harmful. Their use in textile production is becoming increasingly popular, as natural dyes can produce unique and pleasing shades that are often gentle and soft on the eyes [11]. In addition, plant dyes sourced from renewable and sustainable sources can be easily disposed of, and their extraction process does not involve chemical reactions [12]. Researchers worldwide strive to introduce agricultural residual sources that abundantly provide natural dyes to manage the growing demand for these dyes and mitigate potential threats [13]. Such attempts include dye extracted from Java Plum [11], Amur Cork Tree Bark [14], Bougainvillea flowers [15], Lotus Seedpod Waste [16], Pomegranate Peel [17], Red Tea [18], etc.

Apart from carotenoids, chlorophylls, and anthocyanins, betalains are a prevalent pigment derived from natural betalamic acid and contain an immonium group. Nitrogen-containing betalains pigments are water-soluble. The primary sources of betalains can be red violet betacyanins (Figure 1) or yellow betaxanthins (Figure 2) [19,20], which are typically from the Amaranthaceae family (such as *Beta vulgaris* L. and *Amaranthus* sp.) and the Cactaceae family (such as *Opuntia* sp. and *Hylocereus* sp.), respectively. Red pitahaya, scientifically known as *Hylocereus polyrhizus*, is a type of fruit popularly known as red dragon fruit due to its aesthetically pleasing deep purple pulp and abundance of small, soft seeds. This fruit is highly sought after in the European and United States markets and is predominantly cultivated in Asia and Australia [20]. Plenty of dragon fruit peels are available as agricultural waste in various nations, such as Indonesia, where approximately 150,000 tons of dragon fruit are produced annually, which can meet the demand for natural dye production [19].

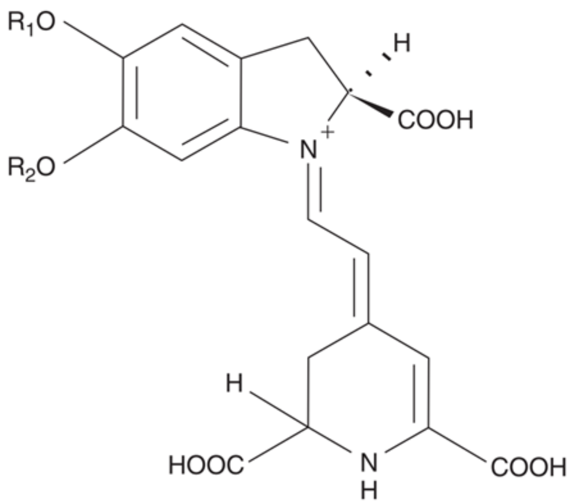

Figure 1. Molecular structure of betacyanin, where R_1 = 3-methyl-3-hydroxy methyl glutaryl and R_2 = H for Hylocerenin (*Hylocereus polyrhizus*). Reprinted with permission from [20]. 2019, Springer Nature.

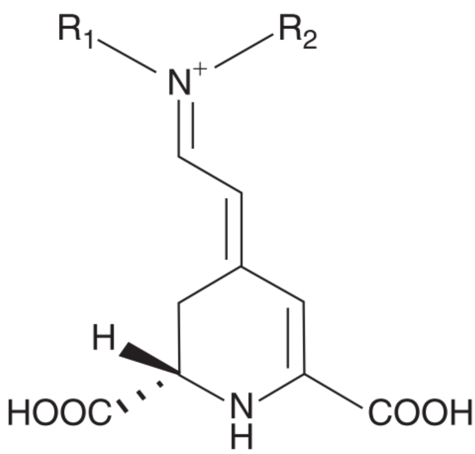

Figure 2. Molecular structure of betaxanthins. Reprinted with permission from [20]. 2019, Springer Nature.

Cotton is the most widely used natural fiber, with the quality of making attractive and comfortable clothing due to its hydrophilic properties [21]. Dyeing the cotton with natural sources is often challenging due to negative charges on its surface. As cotton grows [12], due to its phenolic nature [22], hydroxymethyl groups are partially oxidized into carboxylic acid groups, which results in the cotton carrying negative charges during the dyeing process. This can significantly reduce the efficiency of dye fixation on cotton substrates [12,13]. To address this issue, methods are being utilized to improve dye exhaustion and enhance colorfastness properties on cotton; these include introducing cationic sites into the cotton or employing pretreatment processes, such as chitosan and mordanting with, for example, potassium alum or potassium aluminum sulfate. [12,22,23].

Dragon fruit has been used in various sectors, including the coagulation and flocculation of effluent treatment [24,25], such as other natural substances, i.e., agro-biomass (leaves, seed/flower, stem, and root) and oil palm waste [26,27], which are being used as adsorbent to remove dyes from wastewater waste. Other sectors, including food [28,29], fisheries [30], dye-sensitized solar cells [31,32], and cosmetics [33], are using dragon fruit as raw materials. There is only one work in the published literature where colorants extracted from dragon fruit peel were applied to dye textile substrate [34]; this involved silk fabrics that were dyed using the dragon fruit peel (*Hylocereus costaricensis*). Sliced dragon fruit peel was combined in a 1:10 ratio with water. Then, 2.5%, 5%, 7.5, and 10% lime juice and hot water pre-mordanted silk fabric were dipped in the dye solution before being immersed in a fixer solution of alum (10%). The outcome was light-pink- or baby-pink-colored silk fabric. However, this study uses dragon fruit peel to dye untreated and treated cotton fabric. At first, the optimum dyeing conditions (dye concentration, temperature, time, and pH) were investigated. The treatments used in the study include the use of salt (Glauber's salt) in dyeing, mordanting (potassium aluminum sulfate), and cationizing (ForCat NCH) the cotton fabric before dyeing. The result was a baby pink color for the untreated sample and an off-white or yellowish-white to brownish color for treated samples.

To the best of the authors' knowledge, until now, there has been no research on using dragon fruit's peel as a potential colorant for cotton fabric. The current study explores and utilizes fruit waste as a cost-effective natural dye source for cotton dyeing. The study aims to transform a low-value material into a valuable product by investigating cotton dyeing using dragon fruit peel extract as a natural colorant.

2. Materials and Methods

2.1. Materials

Scoured and bleached single jersey plain knit fabric (100% cotton) of 170 g/m^2 was obtained from Fakir Apparels Ltd., Narayanganj, Bangladesh. Raw dragon fruits were procured from a local market to extract pigment from the fruit's peel. Potassium alum or potassium aluminum sulfate (dodecahydrate), $KAl(SO_4)_2 \cdot 12H_2O$ from Research-lab fine chem. Industries, India, was utilized as a mordanting agent. ForCat NCH, a mixture of cationic polyamine and 1,3, dichloro-2-propanol was employed as a cationizing agent, collected from Fortune Top Pte Ltd., Taiwan. Soda ash (Na_2CO_3) and glacial acetic acid (100% anhydrous, CH_3COOH) were used to maintain varying pH levels. Glauber's salt or sodium sulfate anhydrous (Na_2SO_4) was used to determine whether it was essential to be added to this natural dyeing process. These chemicals were from Merck, Germany. A soaping agent was used for washing purposes. Greyscale was used to evaluate staining and shade change. DW (diacetate-wool) multifiber fabric was employed as an adjacent fabric to assess the staining regarding diacetate, bleached cotton, polyamide, polyester, acrylic, and wool.

2.2. Methods

2.2.1. Extraction of Dragon Fruit Peel

As shown in Figure 3, dragon fruits were first stripped of their peel and diced into small pieces. The peel pieces were then shade-dried. The dried peel was ground into powder using a household blender. The blended peel was then extracted in distilled water (Figure 4) at 10 g/L concentration for 14 h at room temperature (20 °C), 1 h at 40 °C, 1 h at 60 °C, and 1 h at 80 °C with continuous stirring. The extracted solution was then filtered using Whatman filter paper.

(**a**) (**b**) (**c**) (**d**)

Figure 3. (**a**) Separating dragon fruit pulp from the peel, (**b**) cutting the peels into pieces, (**c**) shade-dried peel, and (**d**) crushed peel powder.

(**a**) (**b**) (**c**)

Figure 4. (**a**) Mixing dragon fruit's powdered peel with distilled water for aqueous extraction, (**b**) filtration of the extracted dye, and (**c**) dyeing of the specimen using extracted dye.

2.2.2. Dyeing

Dyeing was carried out using 5 g scoured bleached cotton fabric in a lab dyeing machine (ECO-24, Xiamen Rapid Co., Ltd., Xiamen, China) with a liquor ratio of 1:8 under various dye concentrations, times, temperatures, and pH conditions. A typical time–temperature diagram used in the dyeing process is shown in Figure 5. Here, 15 of the 18 samples were dyed to optimize the conditions for dying cotton fabric with the dragon fruit peel extract. After reaching the proper conditions, one of the fabric samples was dyed along with Glauber's salt, another was dyed while mordanting, and the final one was cationized prior to dyeing with the optimum conditions. After dyeing, the dyed samples were washed properly, and soaping was conducted with 2 g/L soap solution at 60 °C for 10 min and dried at room temperature. Table 1 presents the dyeing experiments carried out to locate optimized dyeing conditions. Once the best dyeing conditions were determined, three more dyeing experiments were performed to investigate the effect of salt addition to the dyeing bath (Table 2), mordanting effect (Table 3), and cationization effect on dye yield (Table 4).

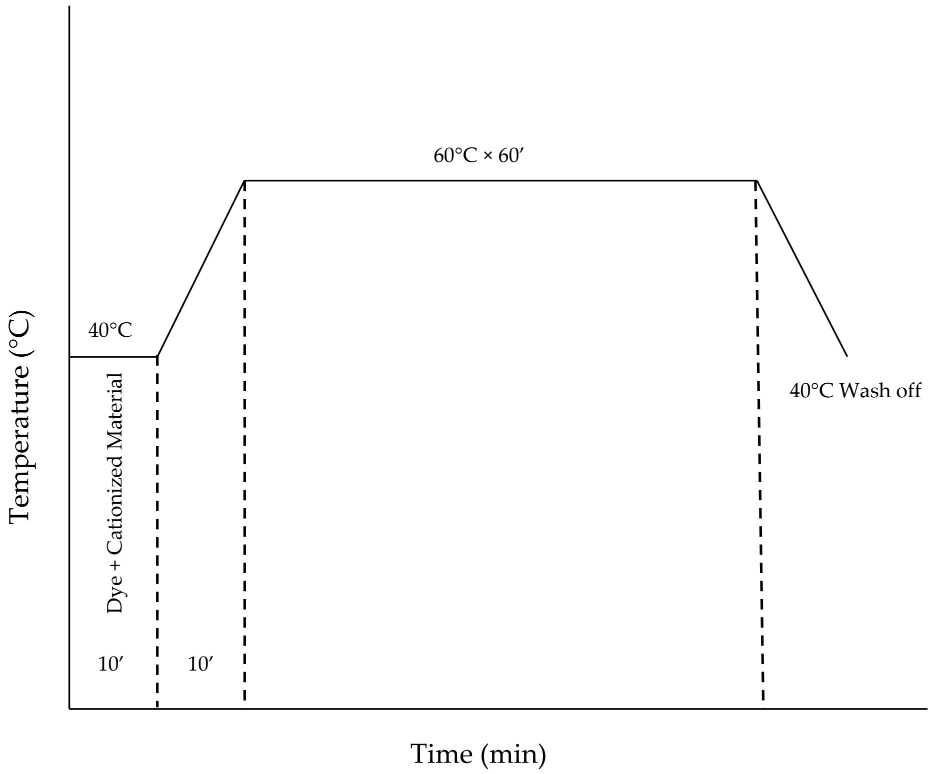

Figure 5. Time–temperature profile of dyeing of cationized cotton knit fabric with dragon fruit peel's extract.

Table 1. Dyeing under various conditions.

Title	Sample No	Dye Concentration (g/L)	Temperature (°C)	Time (min)	pH	Reflectance (%)
Temperature variation	1	30	30	60	7	61.52
	2	30	60	60	7	57.07
	3	30	80	60	7	63.11
Time variation	4	30	60	30	7	58.87
	5	30	60	45	7	62.38
	2	30	60	60	7	57.07
	6	30	60	75	7	64.30
	7	30	60	90	7	62.13
pH variation	8	30	60	60	6	59.70
	2	30	60	60	7	57.07
	9	30	60	60	8	58.70
	10	30	60	60	9	59.89
	11	30	60	60	10	61.78
Dye concentration variation	12	10	60	60	7	62.87
	13	20	60	60	7	58.05
	2	30	60	60	7	57.07
	14	40	60	60	7	56.43
	15	50	60	60	7	53.42

Table 2. Dyeing under appropriate conditions with the addition of Glauber's salt.

Sample No	Dye Concentration (g/L)	Temperature (°C)	Time (min)	pH	Glauber's Salt (g/L)	Reflectance (%)
16	50	60	60	7	10	66.09

Table 3. Dyeing under appropriate conditions with mordanting agent.

Sample No	Dye Concentration (g/L)	Temperature (°C)	Time (min)	pH	$KAl(SO_4)_2{\cdot}12H_2O$ (g/L)	Reflectance (%)
17	50	60	60	9	2	63.21

Table 4. Dyeing under appropriate conditions of cationized fabric.

Sample No	Dye Concentration (g/L)	Temperature (°C)	Time (min)	pH	Reflectance (%)
18	50	60	60	7	37.81

2.2.3. Cationization

One of the cotton fabric samples was treated with 1 g/L soda ash at 30 °C for 5 min in a ratio of 1:8 (cotton fabric weight/dye volume). ForCat NCH was added to the same bath to cationize the fabric surface before dyeing. The temperature was increased to 60 °C after adding ForCat NCH, and the treatment was carried out for 30 min.

2.2.4. Mordanting

During the dying process, the simultaneous mordanting approach was employed. One of the fabrics was dipped into a 2 g/L mordant solution infused with dye extract with a liquor ratio of 1:8 at pH 9, and the dye concentration was 50 g/L. The treatment was carried out at 60 °C for 60 min.

2.3. Testing

2.3.1. Color Strength (K/S) Values of Dyed Fabrics

The DataColor® 800 (Datacolor, Trenton, NJ, USA) dual-beam spectrophotometer installed with a pulsed xenon lamp filtered to approximate D65 light was used to measure the absorbance value of the extracted dye solution and color strength values (*K/S*) of the dyed samples at their respective maximum absorption wavelengths. The dyed sample's color coordinate values (L*, a*, and b*) were also measured by measuring the reflectance curve between 400 and 700 nm using the same instrument. A diffuse illumination with an 8° viewing measurement geometry was adopted during all measurements.

K/S values were calculated using the Kubelka–Munk equation (Equation (1)) and the appropriate software.

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

K is the coefficient of absorption, S is the coefficient of scattering, and R is the reflectance value of the sample.

2.3.2. Colorfastness Determination

Colorfastness to washing of dyed cotton fabrics was assessed according to ISO 105 C06 (C2S): Colorfastness to domestic and commercial laundering [35]. According to ISO 105 E04: 2008 [36], colorfastness to perspiration was determined. Colorfastness to rubbing was evaluated according to ISO 105 X12:2016 [37]. Greyscale was used to assess staining [38] and shade change [39]. DW (diacetate-wool) multifiber fabric [40] was employed as an adjacent fabric to assess the staining toward diacetate, bleached cotton, polyamide, polyester, acrylic, and wool.

3. Results and Discussion

3.1. Effect of Extraction Temperature

Different temperatures were used during the traditional extraction method to select the optimum temperature for extracting the pigment from dragon fruit peel. The maximum *K/S* value was attained at 60 °C, as shown in Figure 6. The *K/S* value grew as the temperature rose from room temperature up to 60 °C, at which point a decline in the *K/S* value was seen. Perhaps 60 °C is the ideal extraction temperature which enables the most dye molecules to be freed from the peel to be available in the bath. Therefore, for cotton dyeing, dragon fruit peel was extracted at 60 °C for 1 h.

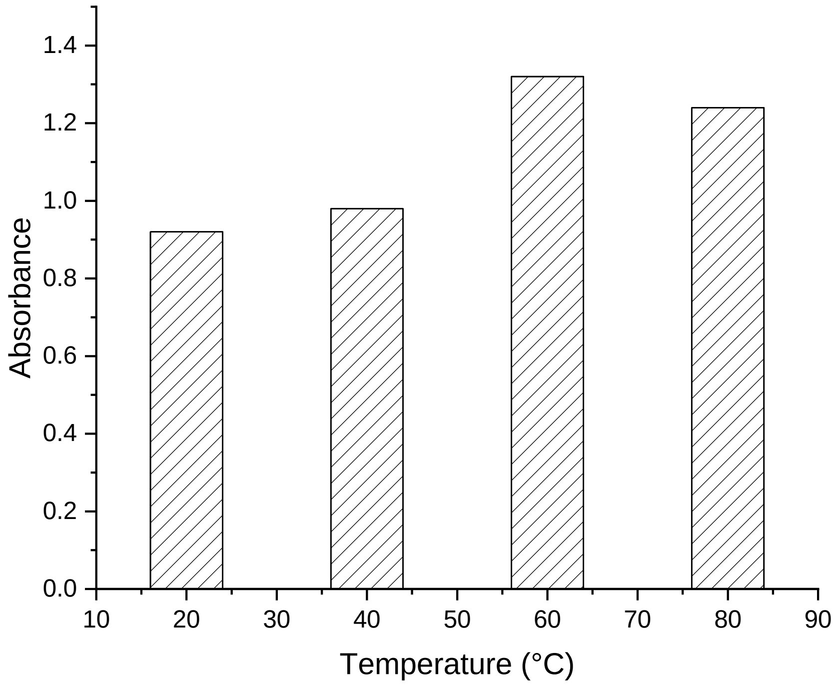

Figure 6. Determination of appropriate temperature for dragon fruit peel extraction.

3.2. *Effect of Dyeing Temperature, Time, pH, and Dye concentration*

Figure 7 provides information on the dyeing process for three samples at different temperatures to compare the K/S values for each sample and choose the appropriate temperature for dyeing. According to the figure, a greater temperature allows for more molecule–diffusion energy, which promotes dye absorption and a higher *K/S* value. The *K/S* value indicates how well the sample material takes up the dye and how deep is the resulting color. In general, higher *K/S* values indicate deeper, more intense colors. In this figure, the sample dyed at 60 °C had the highest *K/S* value of 0.1615, suggesting that the dyeing process was most effective at this temperature. This might be caused by the dye molecules' maximal diffusion rate into the substrate. The energy needed to dye was optimum, and desorption was minimum at this temperature. However, as the temperature increased, the *K/S* value decreased, indicating that temperatures over the ideal range promoted desorption. No other changes in color were observed during the study due to the temperature change other than the variation in color intensity.

K/S value for different dyeing times is shown in Figure 8, from which the appropriate dyeing time can be determined. Here, the sample dyed for 60 min has the highest *K/S* value of 0.1615, which suggests that the dyeing process was most effective at 60 °C for 60 min. The dye absorption improved with time, and more dye molecules could diffuse into the materials, improving the *K/S* value. The desorption rate increased and the *K/S* value gradually decreased as the time passed beyond 60 min, despite the expectation that it would exhibit a similar behavior toward the longer period. This showed that dye desorption is encouraged by times and temperatures higher than the ideal level.

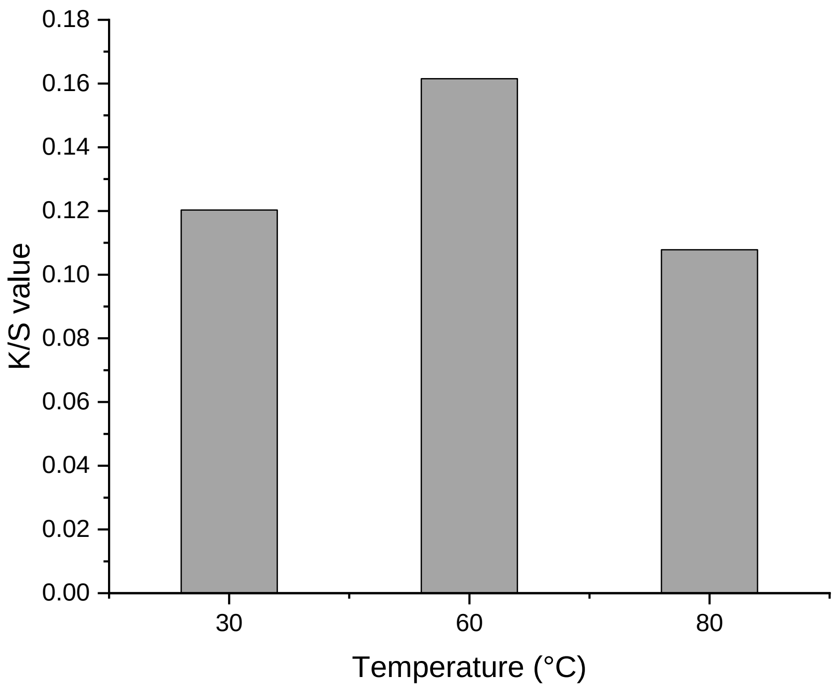

Figure 7. Determination of appropriate temperature for dyeing.

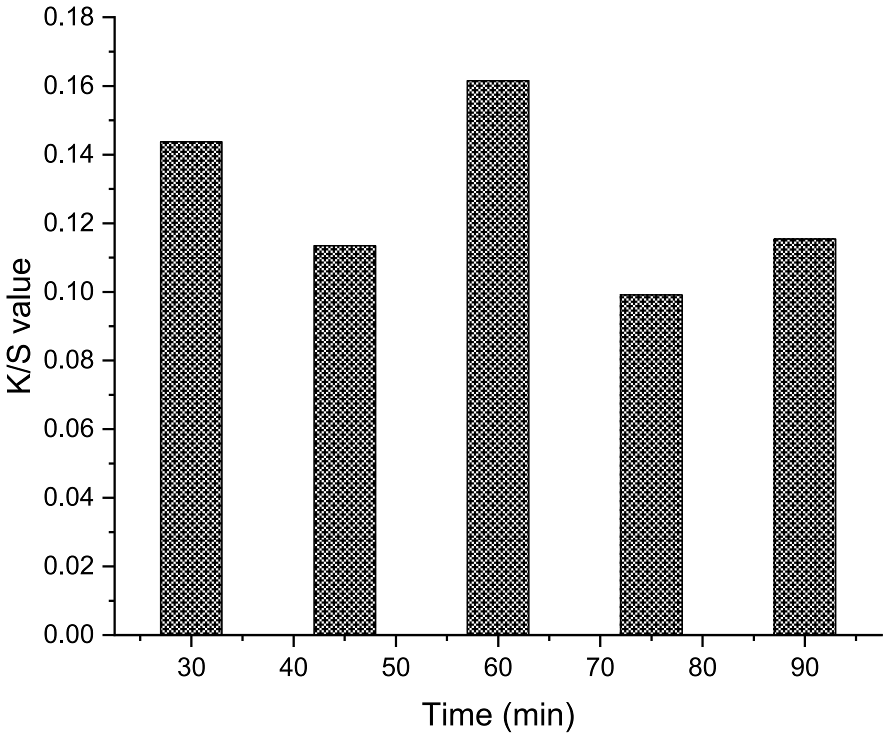

Figure 8. Determination of appropriate time for dyeing.

The proper pH for dyeing was determined by contrasting the *K/S* values for each sample at various pH levels using the information in Figure 9. Here, the sample dyed at a pH of 7 had the highest *K/S* value of 0.1615; after that, it showed a gradual decrease in an alkaline pH range. As the pH increased, cotton fibers became negatively charged by producing cellulosate ion and repelled the available negatively charged betacyanin dye molecules. This resulted in a decreased *K/S* value at pH values higher than 7. The dyed sample's color did not alter or demonstrate any shift due to the pH change.

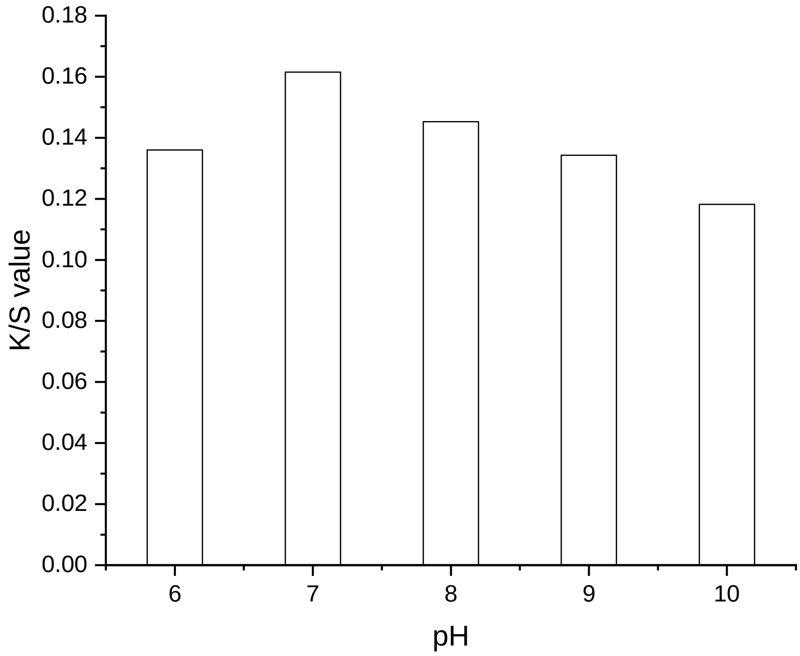

Figure 9. Determination of appropriate pH for dyeing.

Figure 10 is intended to assist in determining the proper dye concentration for a certain dyeing procedure. To determine the ideal dyeing concentration, the crushed dragon fruit peel was dissolved in different concentrations (10, 20, 30, 40, and 50 g/L) and used to color the samples. Here, the *K/S* value generally increased as dye concentration increased. A dye concentration of 10 g/L was too low to generate a deep and rich hue, as seen by the sample treated, where the lowest concentrations of 10 g/L had the lowest *K/S* values (0.1096). On the other hand, the sample treated with the highest concentrations of 50 g/L had the highest K/S values of 0.2031, signifying more dye molecules were available in the bath and could participate as the dye concentration increased, which resulted in the deepest and richest hue.

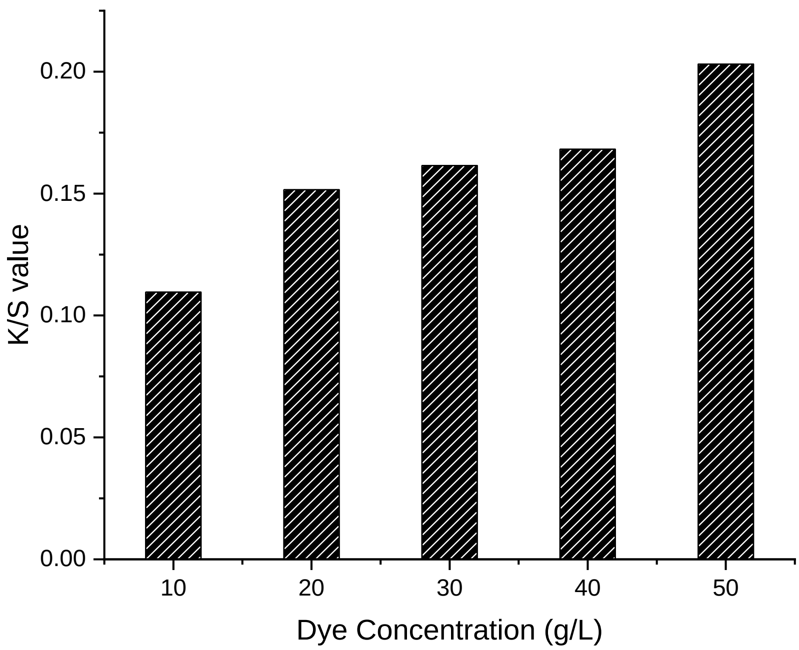

Figure 10. Determination of appropriate concentration for dyeing.

3.3. Comparison between Untreated, with-Salt, Mordanted, and Cationized Samples

The data of the four samples in Figure 11, which were dyed to examine their *K/S* values over 60 min at 60 °C with 50 g/L dye solution at pH 7, are presented in Figure 12. The untreated sample was not subjected to Glauber's salt, mordanting, or cationizing processes. Glauber's salt was added to the with-salt sample. However, the pH of mordanting bath did not remain at 7 due to the addition of potash alum but instead increased to pH 9.

Figure 11. Dyed samples, from left; untreated, salt dyed, mordanted, and cationized.

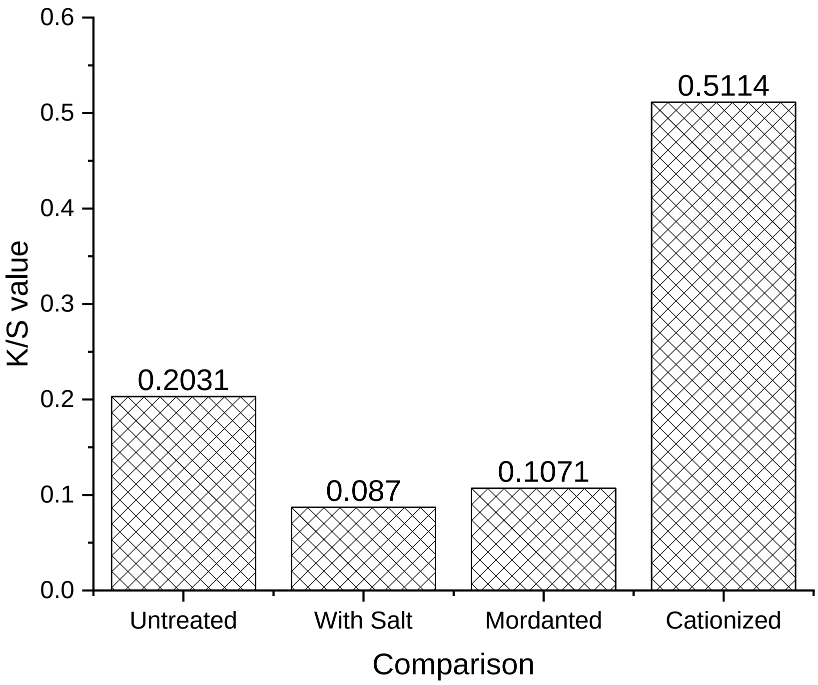

Figure 12. Comparison of *K/S* values among differently dyed fabrics.

The untreated sample appeared to have a *K/S* value of 0.2031, followed by the mordanted sample (0.1071) and with-salt sample (0.087). These variances in *K/S* ratios show that, even at the same concentration (50 g/L), Glauber's salt and the mordanting agent performed less efficiently. The cationized sample appeared to have a *K/S* value much greater than the other samples (0.5114). This implies that the cationizing ForCat NCH significantly changed the solution's dyeing performance. This signifies that the cationized agent enabled more active sites for the dye molecules to be attached to, and this bond between dye molecules and cotton's cellulose through the cationizing agent helped attain the maximum K/S value compared with other samples.

The dyed sample's color changed from baby pink (untreated sample) to yellowish-white (salt-treated and mordanted) and brownish (cationized), indicating a hypsochromic or blue shift. This could result from a molecular change in the electronic structure, increasing the energy needed to excite its electrons, which is responsible for light emission towards shorter wavelengths.

3.4. Colorfastness Analysis

The cationized sample was analyzed only to evaluate colorfastness properties, as it obtained the highest color yield in *K/S*. Figure 13 illustrates that the fabric's wash fastness ranged from poor to fair. This means that taking high dye concentrations may result in a significant loss in color depth. Acid's good perspiration fastness resulted in a minimal loss of color depth, while alkali's outstanding perspiration fastness resulted in no change in color depth. In addition, both the dry and wet rubbing fastness were excellent, indicating no loss in color depth. The poor to fair wash fastness indicated the dye molecules' desorption in the washing condition, which may have broken the bonds made by the cationizing agent and resulted in a color loss.

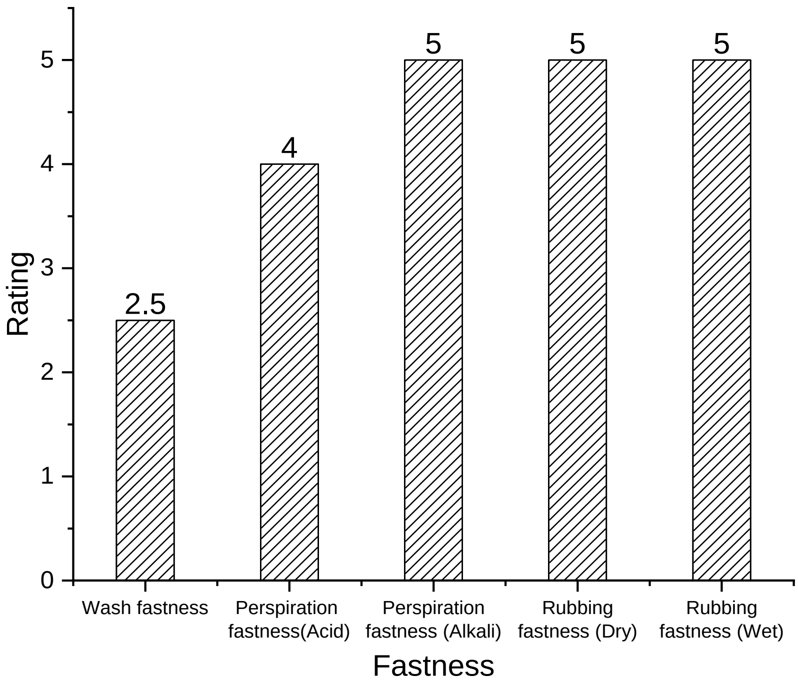

Figure 13. Shade change rating of colorfastness tests.

The DW fabric seemed to have a greyscale rating of 4.5 or higher for wash fastness as shown in Figure 14, indicating that they retain their color well after washing. This fabric also has a perfect rating of 5 for perspiration fastness (acid), which means they are resistant to discoloration when exposed to acidic sweat.

For perspiration fastness (Figure 14) (alkali), all the parts of the DW fabric except for wool have a rating of 5, indicating excellent colorfastness when exposed to alkaline sweat. Wool has a slightly lower rating of 4.5, suggesting it may experience discoloration when exposed to alkaline sweat. Overall, this figure provides useful information for selecting fabrics with good colorfastness properties, depending on the specific application and end use.

When compared with the Natural Organic Dye Standard (NODS) [41], our results for the tests on sweat (both alkaline and acidic) and rubbing fastness (both dry and wet) complied with the criteria given in the NODS. This study used a more vigorous method to test wash fastness, and the results were satisfactory. This study did not use any prohibited or restricted substances listed in the NODS standard. Inorganic element content was not examined, which might be the future scope of this work.

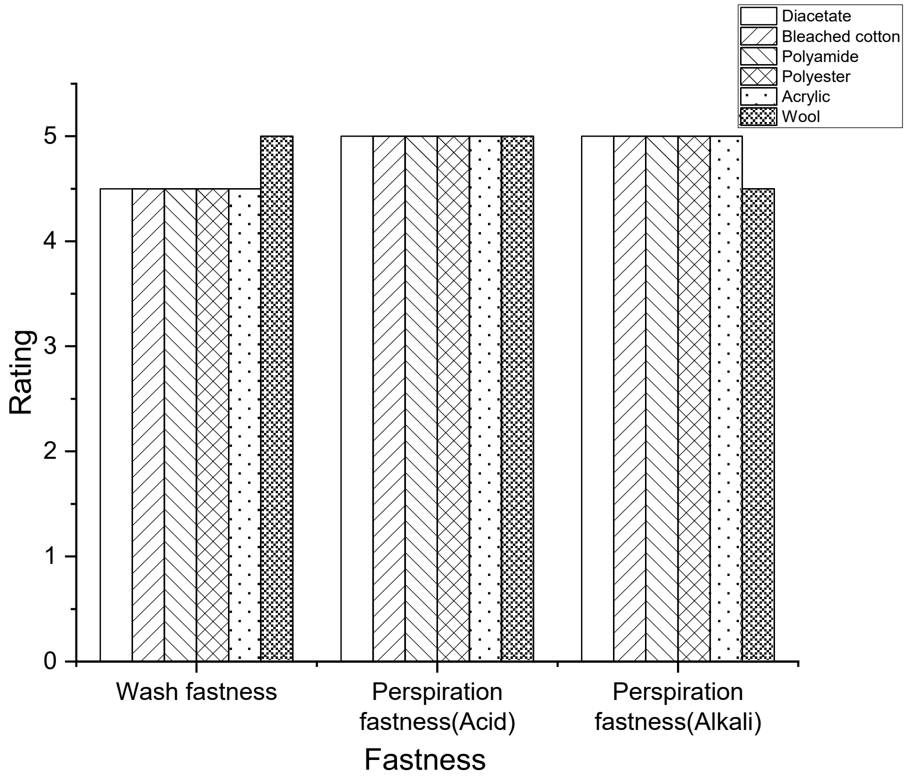

Figure 14. Staining rating of colorfastness tests.

4. Conclusions

The textile industry has been exploring sustainable chemicals to replace harmful and carcinogenic substances used in different stages of production. Natural dyes have gained popularity as a more environmentally friendly and less harmful alternative to synthetic dyes. Red dragon fruit (*Hylocereus polyrhizus*), predominantly cultivated in Asia and Australia, is a potential source of betalains (betacyanins) for natural dye production, as there is an abundance of dragon fruit peels available as agricultural waste. On the other hand, dyeing the cotton with natural sources can be challenging due to opposing charges on its surface. To address this issue, methods are being utilized to improve dye fixation and colorfastness properties on cotton; these include introducing cationic sites into the cotton or employing pretreatment processes, such as chitosan and mordanting with potassium alum or potassium aluminum sulfate. This study aimed to determine the optimal conditions for extracting pigment from the dragon fruit peel and using it for dyeing fabrics. Different temperatures, times, pH levels, and dye concentrations were tested to determine the *K/S* value, indicating the resulting color's depth and intensity.

The maximum *K/S* value was obtained at 60 °C, and the dyeing process was most effective at this temperature for 60 min; pH of 7 was found to be most suitable, and dye concentration 50 g/L produced desirable color results while maintaining good color fastness. Further experiments were conducted to compare the *K/S* values of different chemical solutions with samples using a cationizing agent, ForCat NCH. This chemical significantly improved the dyeability and retention of the dye on the fabric, resulting in a much higher *K/S* value compared with non-cationized samples both untreated and mordanted.

Although the *K*/*S* value was higher for the sample dyed at 50 g/L dye concentration, initially the study was conducted at 30 g/L dye concentration to investigate the ideal dyeing conditions, such as temperature, time, and pH. As a result, the obtained *K*/*S* values were very low. The *K*/*S* value would supposedly rise proportionally if a greater concentration could be used. Colorfastness analysis was also conducted to evaluate the fabric's ability to retain its color over time, where the cationized sample had poor to fair wash fastness properties. Finally, the test results were compared with the NODS standard and complied very well and, in some cases, left areas to explore for future scope of study.

Overall, the study provided insights into the optimal conditions for using dragon fruit peel pigment as a natural dye for fabrics. The findings can be used to guide future research in this area and promote the use of sustainable and eco-friendly dyeing methods.

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