Aqueous Extraction and Dyeing Behavior of Areca Nut (*Areca catechu*) Natural Dye for Cotton Fabric

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Abstract*—The objective of this study is to extract and characterize the natural dye from the powder waste of Areca nut (*Areca catechu*), which is a plant that is widely found in tropical Asia, East Africa, and the Central Pacific regions. The study also aims to develop a recipe for dyeing cotton fabric using this natural dye. The aqueous extraction method was used to extract the *Areca catechu* dye. Two different metal mordants (Aluminium potassium sulfate (Alum) and Ferrous sulfate) and two mordanting techniques (pre-mordanting and post-mordanting) were used at 3% and 5% concentrations to set the extracted dye on the cotton fabrics. The extracted dye exhibited a pH value of 4.0, presented a distinct reddish-brown hue, and showed absorption spectra in the UV-A and visible light regions. The presence of functional groups was determined by FT-IR analysis of the extract. Dyed cotton samples displayed good colorfastness properties, particularly in washing and light fastness. Color intensity values were measured in terms of color strength (K/S) and Color space values (CIE Lab values). K/S values indicated that dyeing with ferrous sulfate had the highest color strength values. Using different mordants causes not only the difference in color strength but also the color properties. Thus, the dye extracted from *Areca catechu* can be used with a suitable mordant for the coloration of cotton fabrics. The future study of *Areca catechu* natural dye should focus on the economic viability of producing the dyes commercially.

Keywords—Areca nut, characterization, extraction, natural dye, textile

I. INTRODUCTION

Due to the environmental issues associated with synthetic dyes, there is an increased interest in environmentally friendly coloring options, including safer synthetic and natural dyes. Most importantly, natural dyes offer the advantages of being derived from renewable sources and biodegradability, resulting in minimal environmental impact [1]. Natural dyes are derived from various sources, including plants, animals, and minerals [2]. They are increasingly favored by consumers, having a global demand exceeding 10,000 tonnes annually. Natural dyes are highly valued for their ability to reflect cultural heritage and unique aesthetic qualities that synthetic dyes cannot replicate [3].

Among all natural dyes, those derived from plant sources hold significant potential due to their widespread availability. Plants such as madder, henna, weld, and indigo have been extensively utilized [4]. Areca nut, or betel nut (*Areca catechu*), is a slender palm tree belonging to the Arecaceae family, and it is widely distributed in tropical Asia, East Africa, and the central Pacific regions [5]. *Areca catechu* nuts are rich in tannins, which can be utilized as a natural dye for the textile industry [6].

A substantial amount of waste is generated in *Areca catechu* processing factories such as “Supari,” “pan masala,” or “Sarawita. The waste primarily includes dust, unripe, and molded nuts, which are typically disposed of by burning. Effectively utilizing this agricultural waste would address disposal issues and provide an additional source of income for the farmers and processing industries. The presence of tannin in *Areca catechu* waste offers opportunities for dye extraction, potentially mitigating waste accumulation while providing economic avenues for agricultural communities.

This research aims to extract and characterize the natural dye from the powder waste of *Areca catechu* nuts using aqueous extraction and to explore the feasibility of integrating *Areca catechu* waste-derived natural dye into industrial fabric dyeing processes.

II. METHODOLOGY

A. Materials

In this experimental study, Areca nut (*Areca catechu*) powder, a waste product of *Areca catechu* processing factories, was collected from a factory in Matale, Sri Lanka. A 100% plain-woven bleached cotton fabric (weight of 80 gm^{-2} with ends per inch (EPI) and picks per inch (PPI) of 58 and 72) was purchased from “Janasalu,” Department of Industrial Development, Kurunegala, Sri Lanka. The laboratory-grade chemicals of Ferrous sulfate ($FeSO_4$, Sigma Aldrich, UK) and Aluminum potassium sulfate ($KAl(SO_4)_2.12H_2O$, Alum, Fluka, Germany) were purchased from chemical suppliers in Colombo, Sri Lanka. Distilled water (laboratory water distiller-Bionics Scientific Technologies, India) was used in dye extraction and chemical preparations. All the chemicals were used without any additional purification.

Fig. 1: *Areca catechu* powder waste

Fig. 2: Aqueous dye extract

B. Methods

1) Dye Extraction

Areca catechu powder was grinded and then sieved using a 250 µm sieve to obtain a uniform particle size (Fig. 1). The Sample was oven-dried at 105 ˚C for 5 hours for moisture content determination.

A mixture comprising 10 g of *Areca catechu* powder and 4 g of Sodium Chloride (NaCl) was mixed with 200 ml of distilled water, following a ratio of 1:20. This mixture was left to stand overnight. Subsequently, it was heated to 70 °C and stirred for 3 hours continuously, as shown in Fig. 2. The mixture was then allowed to cool to room temperature and filtered using a mesh fabric. The resulting aqueous dye extract was then used for further dyeing studies with cotton fabric.

2) Mordanting

Dyeing was carried out with different mordants and without any mordants. Two specific mordants, namely Ferrous sulfate ($FeSO_4$) and Aluminum potassium sulfate ($KAl(SO_4)_2$/Alum), were used in this study. Two Mordanting methods, Pre-mordanting and Post-mordanting, were performed at a temperature of 80 °C for 2 hours. The amounts of mordant added were 3% and 5% (the amount of mordant used from the fabric weight), and distilled water was used for the mordanting process.

For the pre-mordanting technique, a solution was prepared by dissolving the mordant in distilled water, using two mordant concentrations. The material-to-liquor ratio (MLR) employed was set at 1:30. Subsequently, the fabric samples were carefully squeezed to remove excess liquid and allowed to air dry for a few minutes.

For the post-mordanting approach, fabric samples were initially dyed and then mordanted using the two above-mentioned mordants at two different concentrations. The MLR remained consistent at 1:30 throughout this stage. The fabric samples were then squeezed to remove excess liquid and allowed to air dry.

3) Dyeing

In the dyeing process, the fabric was dyed at its initial pH value. 5 g of 100% mercerized cotton fabric was dyed using 200 ml of the dye solution at a temperature of 70°C, maintained for 2 hours. The MLR utilized for this step was 1:40.

4) Dye Characterization

a) pH Value Analysis

The pH level of the dye solution influences the dyeing behavior during dyeing, impacting solubility and color outcome. It was measured using a digital pH meter (S-610H pH meter, PEAK Instruments, USA). Three pH readings of the dye solution were taken using the pH meter, and the first reading was removed. The average pH value was calculated using the second and third readings.

b) Fourier-Transformed Infrared Spectroscopy (FTIR) Analysis

The determination of specific functional groups of *Areca catechu* dye was measured by Fourier transform infrared (FTIR) spectroscopy (Vertex 80, Bruker Corporation, USA). The samples were placed in FTIR-ATR mode at 600-4000 cm^1.

c) Ultraviolet-Visible (UV-Vis) Spectral Analysis

The absorbency of the wavelength of the aqueous dye extract was determined using Ultraviolet-visible (UV-Vis) spectroscopy (UV-3600i Plus, Shimadzu Corporation, Japan). The extracted dye was then diluted by dissolving 1 ml in 50 ml of Deionized water to measure its absorption spectrum under a range of wavelengths (200 nm–800 nm).

5) Evaluation of Dyed Fabric Color Characteristics

Color evaluation was done through various fabric color characterization methods on the dyed fabric samples.

a) Colorfastness Assessment

The dyed fabric samples were subjected to rigorous colorfastness assessments, encompassing various conditions such as exposure to light, rubbing, and perspiration. The colorfastness of *Areca catechu* dyed fabric was evaluated in accordance with the respective international standards, including fastness to washing (ISO-105-C06-2010), fastness to rubbing (ISO-105-X12-2016), and fastness to light (ISO-105-B02-2014). The changes in the shade and the staining of the adjacent multi-fiber fabric were assessed using grey scales [7].

b) Evaluation of Color Strength and Color Space of Dyed Fabric Samples

The color strength of the dyed fabric samples, indicated by their K/S values, measures the extent of light absorption at the maximum absorption wavelength. Higher K/S values signify more intense colors. The color strength (K/S) values of the dyed fabric samples were determined using the "Kubelka-Munk" equation [8].

$$K/S = (1-R)^2 / 2R \qquad (1)$$

Where R is the reflectance of the dyed fabric, K is the absorption coefficient, and S is the scattering coefficient.

The reflectance of the dyed samples (R) was measured using the Data Color spectrophotometer (DC 800 Spectrophotometer, Datacolor Technologies Suzhou Corporation, China). The color strength values of the dyed fabric samples were determined using the ratio of absorption (K) and scattering (S) coefficients (K/S) in the Data Color tools plus software (built-in software in the spectrophotometer).

These color strength values served as quantitative indicators to express the color characteristics of the dyed fabric samples. The color properties were further analyzed within the Commission International de l'Eclairage (CIE) color space. The CIE Lab color space values are expressed as three-color values: L* represents the perceptual lightness from black to white, and a* and b* represent the four colors: red, green, blue, and yellow. a* represents the green–red colors within a range of; b* represents the blue–yellow colors within a range of [9].

III. RESULTS

A. Dye Characterization

1) pH Value Analysis

Without mordanting agents, an aqueous solution of *Areca catechu* dye exhibited a pH of 4.0 and a distinct reddish-brown hue.

2) UV–Vis Spectroscopic Analysis

Fig. 3 presents the UV spectrum of *Areca catechu* aqueous extract dye in an aqueous solution. The spectrum exhibits absorptions in the 357 nm and 478 nm regions, indicating the presence of tannin compounds in the dye and yellow color in the *Areca catechu* aqueous extract, respectively [10], [11]. The dye molecules readily absorb radiations in the UV-A region (320-395 nm) and visible light range [12]. UV-A is the long wave UV wavelength that makes up approximately 95% of the UV radiation that reaches the Earth and it is the primary cause of photo aging, wrinkling, skin cancer, and eye damage because of it can penetrate the dermis layer of skin [12]. AS a result, this absorption in the UV-A region can provide excellent protection against harmful UV radiation.

3) Fourier-Transformed Infrared Spectroscopy (FTIR) Analysis

The FTIR spectra obtained for extracted *Areca catechu* (aqueous extract) are shown in Fig. 4. The presence of –OH groups was indicated by the peak at 3376 cm^{-1}. The –CH group stretching corresponding to the peak at 2921 cm^{-1} [13] and a C=O bond at 1615 cm^{-1} were observed [14]. The aromatic C=C bond is found at 1414 cm^{-1} wavenumber [13], while the stretching of the C–O group was observed in the spectrum near 1275–1200 cm^{-1} [14]. Furthermore, the peaks at 1070 cm^{-1} and 795 cm^{-1} indicate fluoroalkane (C-X) and C=C bonds, respectively [14], [15].

B. Color Evaluation

When the fabric is submerged in the dye solution, it quickly takes on a brownish color, and in the presence of a mordant, the color changes accordingly. The dyeing depth can be improved using different metal salt mordants in both methods. When ferrous sulfate is used as the mordant, the fabric turns to a darker shade. The presence of Alum as the mordant caused a brighter yellowish shade on the fabric. The color chart of the dye is shown in Table 1.

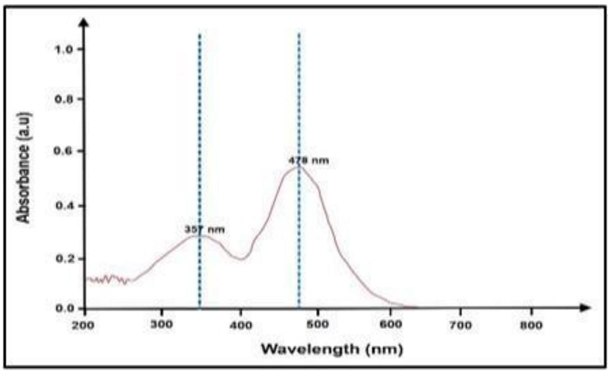

Fig. 3: UV-Vis spectral analysis of Areca catechu-aqueous extract

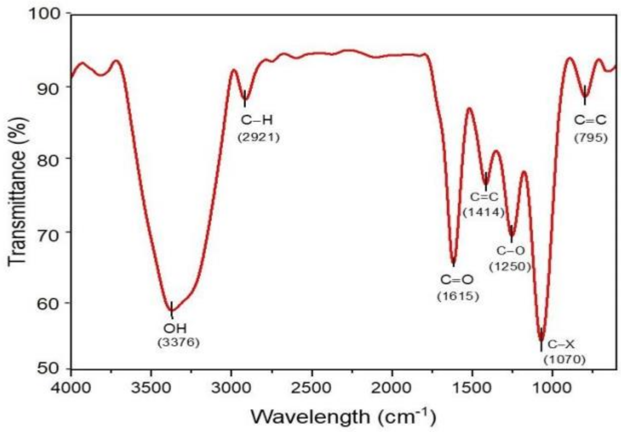

Fig. 4: Fourier transform infrared spectra of Areca catechu dye- Aqueous extract

TABLE 1: COLOR EVALUATION OF *ARECA CATECHU* DYE

Mordant type		Color shade	
Non			
		Mordanting method	
		Pre	Post
$FeSO_4$	3%		
	5%		
Alum	3%		
	5%		

C. Colorfastness Properties

The fastness properties of dyed fabric samples can be altered based on the type of mordants used, the method of mordanting, and the concentration of the mordant. To determine how much the dyed cotton sample can withstand washing, light, and rubbing, the color fastness test must be carried out.

Tab. 2 displays the colorfastness characteristics of cotton fabric dyed with *Areca catechu* dye. In comparison to the mordanted samples, the non-mordanted sample showed poor fastness to washing with a color change of 2-3. The rating for colorfastness to washing for both Alum and Ferrous sulfate appears to be above 4 for color change except for the Alum 3% pre-mordanted sample. According to the wash fastness results shown in Table 2, both substrates showed outstanding color staining values of 4-5 and 5. The method of mordanting appears to have the same effect on the color fastness of washing cotton fabric dyed with *Areca catechu* dye for both the mordanting agents used. This indicates excellent fastness to washing.

Colorfastness to light for cotton dyed with this dye in the absence of any mordanting agent was improved upon the application of ferrous sulfate and alum as the mordanting agents for both the mordanting methods. There is a high light fastness for ferrous sulfate which had excellent color change values of 4-5 or above, and fabric samples mordanted with Alum displayed lower color change values compared to ferrous sulfate. Furthermore, the ratings of post-mordanted fabrics dyed by 3% and 5% of ferrous sulfate are higher than the pre-mordanted fabrics.

With the use of mordants, the rub fastness of the cotton samples has improved slightly. The rub fastness of the cotton samples was better for wet rubbing than for dry rubbing. With very few exceptions, the tested samples displayed a grey scale rating of 4 for wet rubbing. On the other hand, for dry rubbing the grey scale rating was 3-4.

D. The Color Characteristics of The Dyed Fabrics

Color characteristic values of the dyed cotton fabrics resulting from the dyeing process are shown in Fig. 6. It shows the dye uptake of the dyes in terms of K/S values at different concentrations and different mordanting methods. It has been observed that using ferrous sulfate as a mordant significantly improves the K/S values. Among the chemical mordants used, 5% ferrous sulfate has the highest color strength value (K/S = 6.44). An improvement in the K/S value of the dyed samples denotes a higher amount of dye adsorption, interaction, and bridging with the pre-mordanted substrate via different conjugated bonds.

There are variations in hue color as well as significant changes in K/S values, L* values, and brightness index values with the type of mordant used. Fig. 5 shows the CIE L* a* b* values of dyed cotton fabrics with *Areca catechu* aqueous extract. It can be seen that the fabric samples with lighter shades give a higher value of L*, while those with a darker shade have a lower L* value. The L* values were lower for the fabric samples dyed using ferrous sulfate, corresponding to deeper shades. The L* value was higher for non-mordanted dyed samples, corresponding to lighter shades. Similarly, using Alum, the L* values were also higher, leading to lighter shades. Positive values of a* and b* represent shades of red and yellow, respectively. Also, higher values of a* and b* indicates brightness, which denotes the red and yellow hues, respectively.

TABLE 2: FASTNESS PROPERTIES OF COTTON FABRIC DYED WITH *ARECA CATECHU* DYE

Mordant	**Mordant concentration**	**Mordanting method**	**Wash fastness**		**Lightfastness**	**Rubbing fastness**	
			Color Change	**Color Staining**	**Color Change**	**Dry staining**	**Wet staining**
Non	-	-	2-3	4	2-3	3	3
$FeSO_4$	3%	Pre	4	4-5	4	3-4	3-4
		Post	4	5	4-5	4	4
	5%	Pre	4	4-5	4	3-4	3-4
		Post	4	4-5	4-5	3-4	4
Alum	3%	Pre	4-5	5	3-4	3-4	4
		Post	4	5	3-4	3-4	4
	5%	Pre	4	4-5	3-4	3-4	4
		Post	4	4-5	3-4	3-4	4

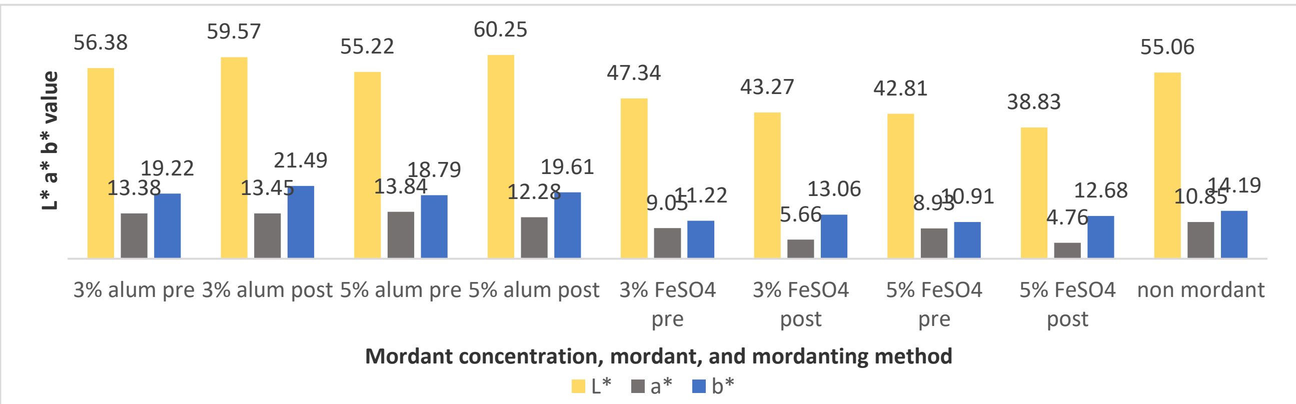

Fig. 5: L*a*b* value comparison of cotton fabric dyed with Areca catechu - Aqueous extract

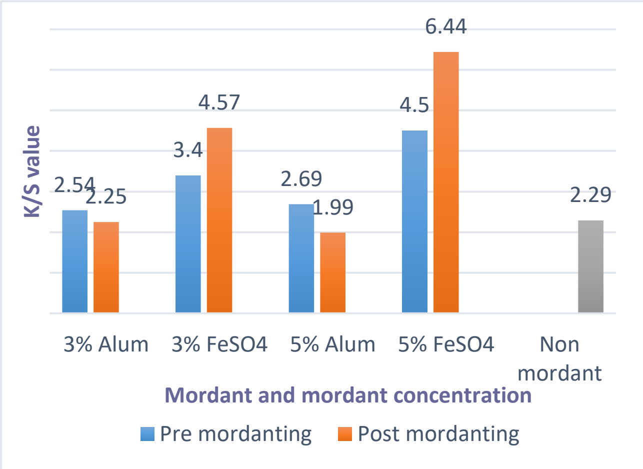

Fig. 6: K/S value comparison of cotton fabric dyed with Areca catechu - Aqueous extract

It can be seen that the b* value of all the dyed fabrics ranged from 10.91 to 21.49, which represents the yellow color. The a* value varied from 4.76 to 13.84, which shows the dyeing colors obtained from all the dyeing processes were mainly red. Using alum as the mordant has increased the quality of the red tone, while ferrous sulfate has caused a reduction in the quality of the red tone.

The mordanting process showed an impact on the quality of the yellow tone, which can be seen from the b* value of the mordanting methods: post-mordanting> pre-mordanting> non-mordanting. The b* values of the dyed cotton fabrics show that the dyeing colors obtained from all the dyeing processes were mainly yellow.

IV. Conclusion

This research is focused on the potential of *Areca catechu* as a natural dye for cotton fabric. *Areca catechu* dye, extracted through aqueous extraction, demonstrated its capability to impart unique color characteristics to cotton textiles. Without mordanting agents, the aqueous solution of *Areca catechu* dye extract exhibited a pH of 4.0, presenting a distinct reddish-brown hue. The dye has absorption spectra in the UV-A region and visible light region. The structural analysis of the functional groups in each extract was established using FTIR spectroscopy. The color evaluation results showed that the addition of mordants significantly influenced the color outcomes. Ferrous sulfate resulted in a darker shade, while Alum produced a brighter yellowish tone. Additionally, the mordanting process positively affected colorfastness properties, particularly in washing and light fastness. K/S values indicated that dyeing with ferrous sulfate yielded higher color strength values. L* values were found to be lower when ferrous sulfate was used, which correlates to deeper shades. Alum gives higher value of L*, which corresponds to lighter shades. The results revealed that cotton fabrics mordanted using 5% ferrous sulfate by post-mordanting gave the optimum color strength with good fastness properties. Further research can explore the optimization of dyeing parameters and investigate the potential for scale-up production of *Areca catechu*-based dyes for commercial applications.

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