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The use of natural *Areca catechu* dyes for silk and nylon and its halochromic effect

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ABSTRACT

In this study, we have investigated the use of betel nut (*Areca catechu*) extract solution as a natural dye for the use of dyeing for silk and nylon. Chemical structure of betel nut extracted solution was confirmed by using FTIR and ^{1}H NMR spectroscopy and thermal properties also measured for determining the proper dyeing temperature. In order to optimise the colouration properties, variable dyeing parameters (temp, pH and mordant type) was studied. The halochromic effect of synthesised natural dye was analysed by using UV–Vis spectroscopy. Additionally dye build-up, colour fastness properties of both fabrics were compared.

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KEYWORDS
Areca catechu; dyeing properties; halochromic effect; colour fastness

Introduction

Since the prehistoric times, natural dyes were used for colouring of food substrates, leather and natural fibres like wool, silk and cotton. After the invention of first synthetic dyes 'Mauvine', the situation has been changed and the use of synthetic dyes became popular due to a wide range of applications in various fields because of its wide range of hues and good stability of colour (Christie, 2015; Fox, 1987; Narayanaswamy, Ninge Gowda, & Sudhakar, 2013; Samanta & Agarwal, 2009; Shahid, Islam, & Mohammad, 2013; Shukla & Vankar, 2013). Nowadays, a great awareness on the impacts of toxic chemicals on the environment and human health has turned down on the use of synthetic dyes and in these circumstances; a higher demand is put towards the greener alternative substances. Recently, the demand for natural colourants is increasing due to majority of the sources are safer, more environmental friendly, good antibacterial, deodorising and ultraviolet protection properties (Bhat, 1978; Glover 1995; Chen, Wang, Cui, Pan, & Millington, 2015; Ibrahim, El-Gamal, Gouda, & Mahrous, 2010; Prusty, Das, Nayak, & Das, 2010; Sionkowska & Planecka, 2011; Zhou & Tang, 2016). Among all the natural dyes, plant based dyes are mostly potential because of their easy availability. *Areca catechu* is the scientific name of betel nut, also well known as single-trunked palm which is found in east Africa, tropical Asia and the central pacific. Even though chewing the betel nut stimulates the flow of saliva to aid for digestion of food however it caused cancer cells for human being (Duke, 1989; Jayamani, Hamdan, Rahman, & Bakri, 2014). Betel nut fruit also produced fibres, which is used for non-woven fabrics and possesses excellent dyeing behaviour, good strength, drape and air permeability. Furthermore, some researcher reported that betel nut fibre used for filler for polymer composites with polypropylene (Chakrabarty, Hassan, & Khan, 2012; Najm, Mohammad, & Ludin, 2017). *A. catechu* nut contains tannin, gallic acid, catechin, alkaloids, fat, gum, etc. The predominant pigment of *A. catechu* is gallotannic acid, which is used for dye sensitised solar cells, anti-aging effects for skin and good antimicrobial activity (Cyriac, Pai, Varghese, Shantaram, & Jose, 2012; Lee & Choi, 1999; Sarker, 2004; Zhang, Li, Han, & Zhang, 2009).

In the present study, *A. catechu* (betel nut) colourants were extracted, the chemical characterisation of dyes were checked by FTIR, ^{1}H NMR spectroscopy and halochromic effect in various pH conditions were determined by using UV–Vis spectroscopy. The extracted natural dye was applied to silk and nylon fabric, so the dyeing properties, colour fastness properties were examined.

Experimental

Materials

In this experiment, 100% muslin silk woven fabric (60 g m^{-2}) was kindly supplied by Bangladesh sericulture research institute, Rajshahi (Bangladesh) and 100% nylon single jersey fabric (75D) also obtained from Beximco Synthetic Ltd. (Bangladesh). All other chemicals were purchased as chemical reagents from Sigma-Aldrich Chemical Company Ltd. (Bangladesh). Betel nut (*A. catechu*) used as a natural dye was collected from local market in Dhaka (Bangladesh). The reason for choosing this natural dye was because of its easy accessibility, good antibacterial and UV protection ability. The chemical structure of betel nut (*A. catechu*) is 1-methyl-1,2,5,6-tetrahydro-pyridine-3-carboxylic acid methyl ester also known as arecoline, given in Figure 1.

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Methods and procedures

Dye extraction

Betel nut (32 g) were mixed with 800 ml methanol in a round bottom flask with a condenser and placed on hot plate for boiling for 4 h. After aging for 18 h at room temperature, solution was filtered by Bruckner funnel. Dye was obtained (Table 1) from the filtered solution by rotary evaporator at 120 °C and dried in vacuum oven for 60 min.

UV–Vis spectroscopic measurement

Extracted dye (0.5 g) was dissolved in 50 ml methanol, and the solution was transferred in a flask. A 1 ml dye solution was added with 9 ml distilled water in a vial. The maximum absorption of the wavelength (λ_{max}) of dye was determined using UV–Vis spectroscopy (Alginet 8453, Made in USA). The absorbance of the wavelength range is 200–700 nm for known concentration could be measured.

FTIR spectroscopy

The determination of specific functional groups of betel nut extracted dye was measured by FTIR (Perkin Elmer) from Frontier, USA. The samples were placed in ATR mode, the number of scanning was 50 times and the resolution power was 4 cm^{-1}.

^{1}H NMR spectroscopy. Betel nut extracted dye were analysed by ^{1}H NMR spectroscopy. The samples were prepared by dissolving 1.0 mg of dye in DMSO. The ^{1}H NMR spectra were recorded on a 500 MHz Bruker Avance machine with a temperature control unit.

Differential scanning calorimetry

The general thermal properties of natural *A. catechu* dye were examined using differential scanning calorimeter (STA 449F3 Model Jupiter, NETZSCH, Germany). About 10 mg of samples were preheated to 650 °C for 5 min and rapidly cooled down to 30 °C. Subsequently, samples were heated up to 650 °C with a heating rate of 10 °C/min, and melting temperature (T_m) was determined.

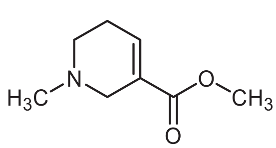


Figure 1. Chemical structure of *Areca catechu* (1-methyl-1,2,5,6-tetrahydro-pyridine-3-carboxylic acid methyl ester).

Analysis of suitable dyeing condition for A. catechu dye

Dyeing's were carried out in Starlet Dyeing Machine DLS-6000, Infrared at different temperature, time and pH for analysing the dyeing performance. Dye stock solution (0.4%) was prepared by mixing with methanol. A 5 ml stock solution of dye was added with 95 ml distilled water (Liquor ratio 100:1) and dyeing were carried out for 1.0 g of nylon and silk fabric by increasing the temperature (50–90 °C) for 50 min. For checking the appropriate temperature (50–90 °C), pH (pH 2, 4, 7, 9 and 11) and suitable mordant for natural *A. catechu* dyeing for silk and nylon fabric, less amount of dye solution (0.4%) was used. Dyeing was carried out without mordant and with different mordant. In case of mordanting, $KAl(SO_4)_2$, $FeSO_4$ & $CuSO_4$ (the amount of 10%) were added to the dye bath. Simultaneous application of the dye and the mordant (Meta mordanting process) was applied during dyeing in exhaust method.

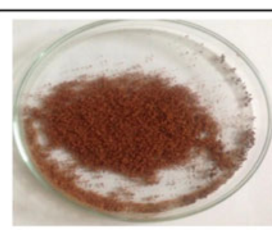
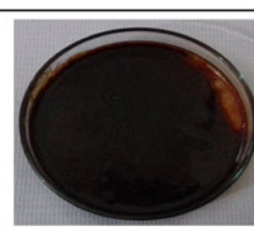
Build-up properties measurement

A 50 ml dye bath, suitable for nylon and silk fabrics (liquor ratio 20:1), containing extracted dye (0.5%, 1%, 2%, 4% and 6% owf) was prepared. Measurement for the build-up properties, dyeing was performed with acetic acid (2.0 g/l) for adjusting pH 4.0, $FeSO_4$ (10%) for mordant and temperature was raised at 70 °C for 50 min in a laboratory (Starlet Dyeing Machine DLS-6000, Infrared) dyeing machine (Figure 2). After dyeing, all the dyed samples were rinsed with cold tap water and dried at room temperature.

Measurement of colour strength

The colour strength (f_k) and the colour difference (ΔE) values of the dyed fabrics were measured using a spectrophotometer (X-Rite 8000 Series, standard light D_{65}, 10° standard observer, specular component included; X-rite, USA) that was interfaced with a personal computer. Here, f_k represents the colour strength (Kabir & Koh, 2017), value as the sum of the weighted K/S values in the visible region of the spectrum, as follows in Equation (1):

Table 1. Betel nut used in the experiments.

Before dye extraction	Before dye extraction	After dye extraction	After dye extraction
			

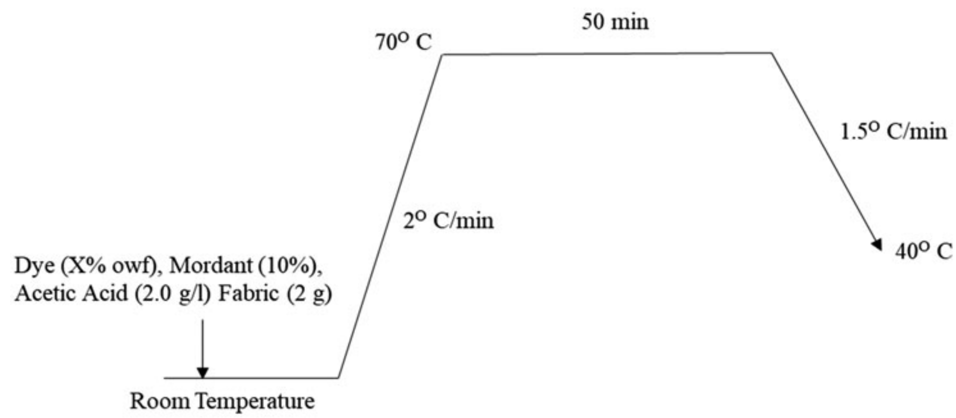


Figure 2. Dyeing profile for *Areca catechu* dye.

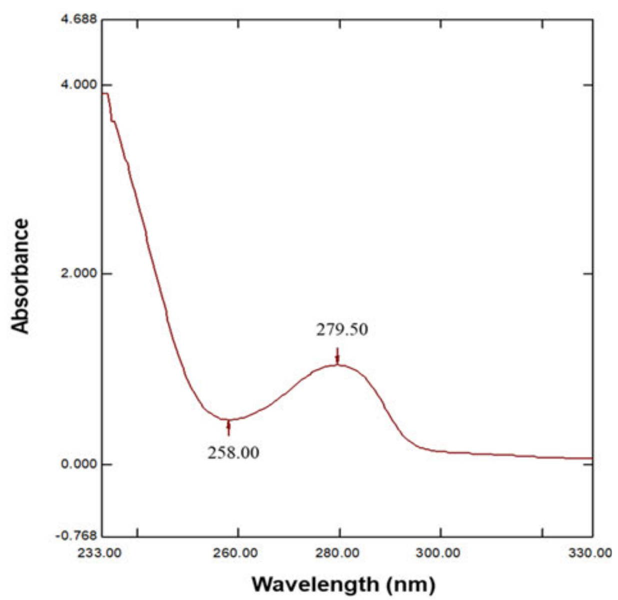


Figure 3. UV–Vis spectrum of *Areca catechu* dye.

$$f_k = \sum_{\lambda=400}^{700} \left(\frac{K}{S}\right)_\lambda \left(\bar{x}_{10,\lambda} + \bar{y}_{10,\ \lambda} + \bar{z}_{10,\ \lambda}\right) \quad (1)$$

where $x_{10,\lambda}$, $y_{10,\lambda}$ and $z_{10,\lambda}$ are the colour-matching functions for a 10° standard observer at each wavelength (ISO 7724/1: 1984). The depth of shade of the dyed fabric was evaluated according to the AATCC test method 173-2006 under illuminant D_{65}, large area view and CIE 10° standard observer(Basel: ISO, 1984) AATCC Technical Manual (2006).

Measurement of colour fastness properties

In order to evaluate the colour fastness, we used 4% owf, because betel nut (*A. catechu*) extracted dyed silk and nylon produced a higher colour strength than other dye concentration under the influence of mordant and acidic condition. *A. catechu* dyed silk and nylon fabric were assessed according to the corresponding international standards, that is, fastness to washing (ISO 105-C06 A2S: 2010), fastness to perspiration (ISO 105-E04:2013), fastness to rubbing (ISO 105-X12:2016) and fastness to light (ISO 105-B02:2013). The changes in the shade and the staining of the adjacent multifibre fabric (Multifibre DW, adjacent fabric, BS ISO 105-F10:1989) were assessed using grey scales (Basel: ISO, 2010, 2013, 2016, 2013).

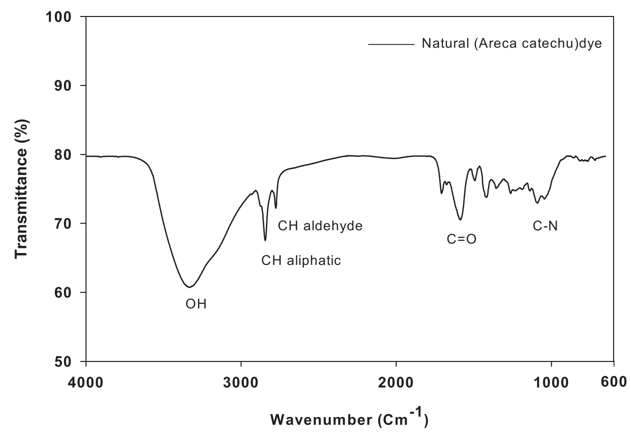


Figure 4. FTIR spectra of natural *Areca catechu* dye.

Results and discussion

UV–Vis spectroscopic analysis

The UV spectrum of the crude betel nut extract dye in an aqueous solution is presented in Figure 3. The characteristic spectrum has shown the absorptions in 230 nm and 260–300 nm regions. Absorption in the 230 nm region owing to $\pi \rightarrow \pi^*$ transitions, may be attributed to various chromophores, including the C = C bond of various compounds, the C = O bond of carbonyl compounds, and the benzene ring. Absorption in the 260–300 nm regions owing to $n \rightarrow \pi^*$ transitions, may be attributed to the electronic transitions of benzene ring and its derivatives. It can be observed that the dye molecules can easily absorb radiations in the UV-C region (230–280 nm) and the UV-B region (280–310 nm). Absorption in the UV-B region can be expected to offer good protection from harmful UV radiation.

FTIR analysis

The chemical structure of natural *A. catechu* dyes has been investigated by the change of the peaks from the

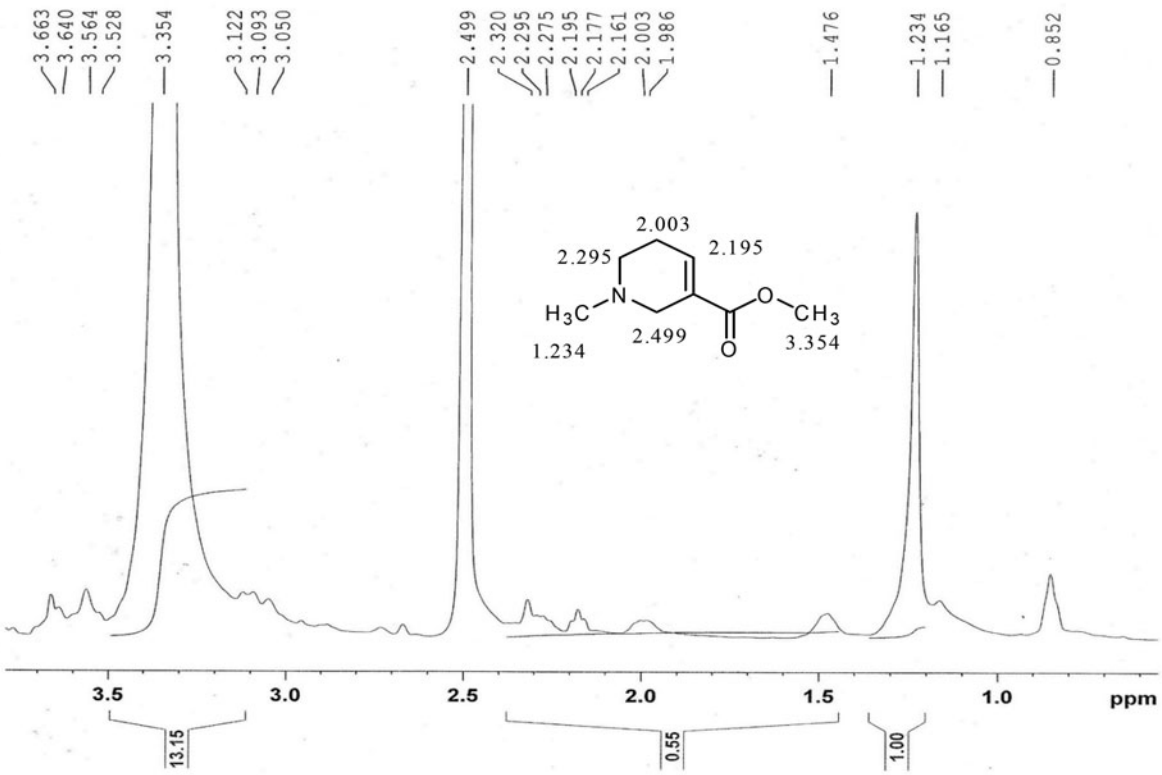


Figure 5. [1]H NMR spectra of natural *Areca catechu* dye.

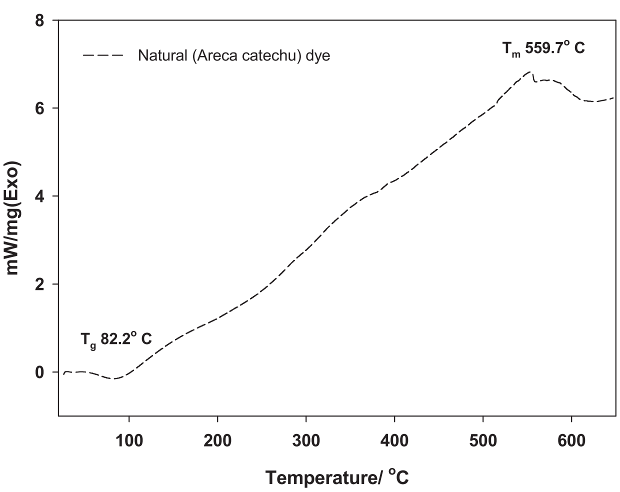


Figure 6. Differential scanning calorimetry of natural *Areca catechu* dye.

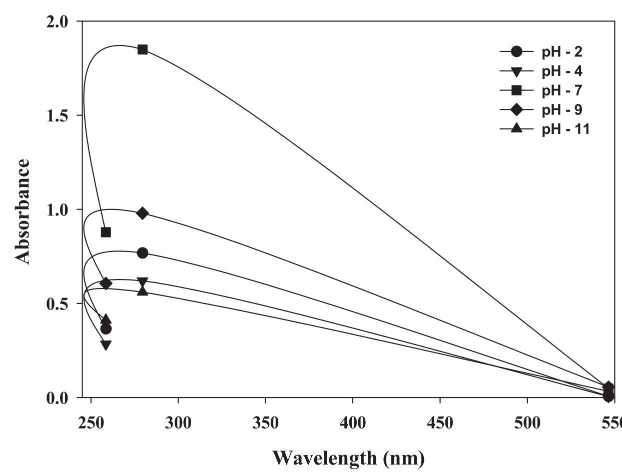


Figure 7. Halochromism of *Areca catechu* dyes.

characteristic spectra shown in Figure 4. The broad absorption band which appeared in the range of 3500–3100 cm^{-1} indicates the presence of –OH groups.

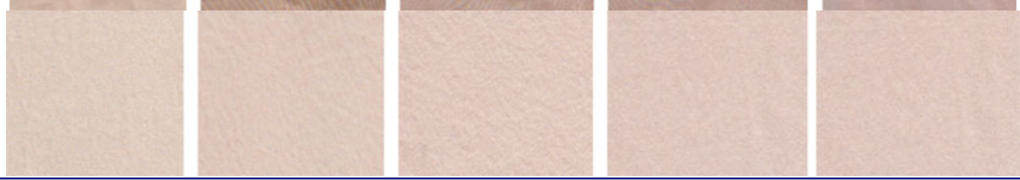
Again, the spectrum near 2900–2800 cm^{-1} corresponding to the symmetric stretching of methylene ($–CH_2–$) groups which is included with aliphatic or aldehyde groups and 1735 cm^{-1} related to the C = O stretch of ester groups were found. The absorption peak appeared at 1200 cm^{-1} for the formation of (C–N bonds) in pyridine structure of the dye.

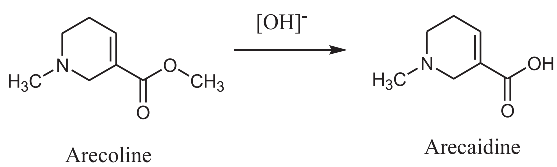
[1]H NMR & DSC analysis

The [1]H NMR spectra of *A. catechu* dyes is shown in Figure 5. For the chemical shift of methyl ester group ($–COO–CH_3$) signals were found at 3.354 ppm, and pyridine group at 2.499 ppm. For chemical shift of methyl group, a new peak appeared at 1.234 ppm which is due to the proton of pyridine ring of betel nut crude dye.

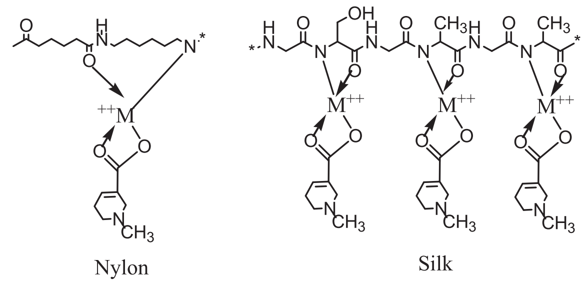
The DSC diagram of natural *A. catechu* dyes is shown in Figure 6. The glass transition temperature of dyes appeared at 82.2 °C and the melting point has been found at 559.7 °C. The higher melting point ascribed to the existence of a 1,2,5,6-tetrahydro-pyridine group, which was characterised by FTIR and [1]H NMR measurements. The meta-linkage and the presence of carboxylic acid methyl ester groups enhanced the higher crystallinity of the dye so it increased the higher melting point. The phenomenon of halochromism was investigated at different pH solution of *A. catechu* dyes.

Table 2. Areca catechu dye solution and dyed sample in different pH condition.

Dye solution	
Silk	
Nylon	



Scheme 1. Preparation of arecoline to arecaidine.



Scheme 2. Coordination of metal complex with *Areca catechu* dyes and protein fibre.

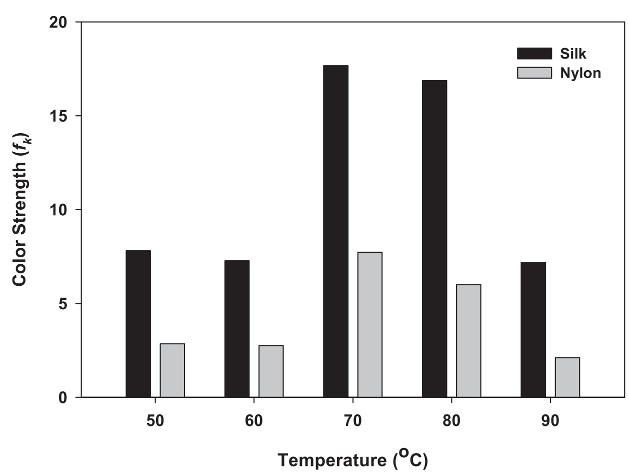


Figure 8. Effects of dyeing temperature on colour strength (f_k) for silk and nylon (dye stock solution 0.4%, without mordant, pH 7.0, time 50 min and material to liquor ratio 1:100).

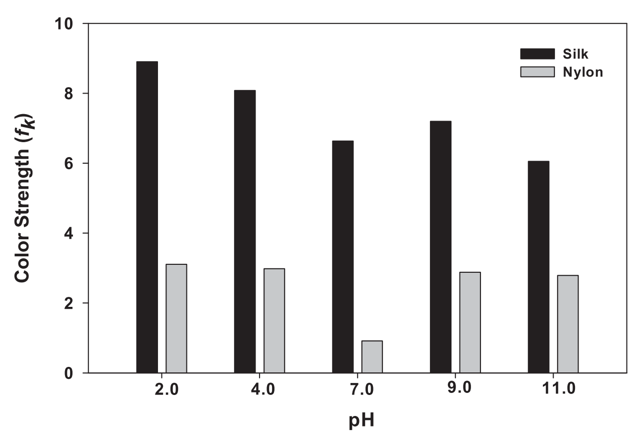


Figure 9. Effects of dyeing pH on colour strength (f_k) for silk and nylon (dye stock solution 0.4%, without mordant, temp 70 °C, time 50 min and material to liquor ratio 1:100).

Halochromic effect

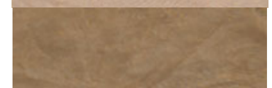
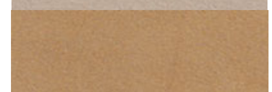
Natural *A. catechu* dyes exhibit a sensitive absorption in different pH condition arising from internal charge transfer, as shown in Figure 7. The electronic character of the dye substituents affects the absorption spectra.

The electron donating ability of methyl group ($-CH_3$) for arecoline at neutral condition shows higher absorption, however in alkaline condition methyl ($-CH_3$) groups shifted to hydroxyl (–OH) groups and formed arecaidine so electron accepting groups effects the low absorption spectra. The colour change of natural dye solution in different pH condition has been depicted in Table 2, it can be shifted yellow to red colour when the pH shifted from pH 2 to 11.

It can be explained that the chemical bonds can be converted hydrogen from hydroxyl ions in solution shown in (Scheme 1).

The conjugated bonds for the carbonyl group of arecoline can be shifted towards hydroxide ions in the range of electronic flow. Various shades of nylon and silk fabric in different pH have demonstrated that the result of halochromic effect of the dye molecules illustrate in Table 2. The ground and excited states in *A. catechu* dyes has shown closer in energy, so absorption illustrates as positive halochromism.

Table 3. CIEL*a*b* and f_k values of dyed samples with extract solution of betel nut (dye stock solution: 0.4%, pH: 4.0, temp: 70 °C, time: 50 min).

Material	Mordant type	Colour strength (f_k)	L^*	a^*	b^*	Dyed sample
Silk	$KAl(SO_4)_2$	7.064421	75.79	4.36	20.15	
	$FeSO_4$	34.44966	58.89	3.88	29.25	
	$CuSO_4$	22.74093	60.09	3.23	21.54	
Nylon	$KAl(SO_4)_2$	6.116173	78.14	4.4	18.31	
	$FeSO_4$	21.61802	67.93	5.28	37.13	
	$CuSO_4$	10.56554	69.7	3.49	17.05	

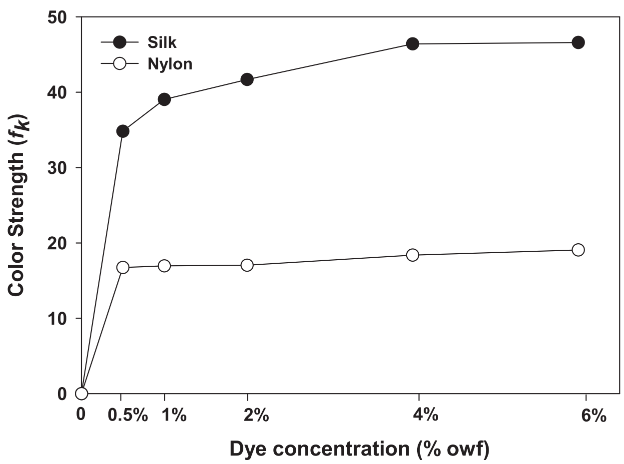


Figure 10. Build-up properties of silk and nylon fabrics (Mordant: $FeSO_4$ (10%), Temp: 70 °C, pH: 4.0 and Time: 50 min).

Dyeing properties

As shown in Figure 8, colour strength was greatly influenced by the dye bath temperature. When the temperature raised at 70 °C, the colour strength gradually increased with increasing the dye bath temperature, so the colour strength of silk and nylon fabric dyed with *A. catechu* dyes obtained at ($f_k = 17.66$ & 7.728), respectively. However, when the temperature was increased more than 80 °C, the colour strength (f_k) gradually decreased. The colour strength (f_k) of silk fabrics was higher than nylon fabrics.

It can be explained that the betel nut extracted dye is the formation of arecoline, which can be converted into arecaidine by the influence of aqueous condition. Arecaidine formed a covalent bond with the amino group (–NH–) of silk and nylon (Scheme 2). Silk fibre consists of higher number of amino groups present in dye molecules, which is influenced by more dye penetration, on the other hand nylon fibre closely packed with crystalline structure so dye penetration rate is low.

Figure 9 illustrates the higher colour strength of betel nut extracted natural dye on nylon and silk fabric at various pH levels. An acidic condition (pH 2 and 4) has clearly shown the colour strength (f_k) of *A. catechu* dyed silk fabric ($f_k = 8.9$ and 8.0) and nylon fabric ($f_k = 3.1$ and 2.9), respectively. Nylon and silk fibres have a variety of amino acids containing NH_2–(X)–COOH both basic and acidic groups. So oppositely charged ionic species attracted electrostatically or coulombically the anions of natural dyes and the positive charge with the silk and nylon fibres. The neutralisation of the carboxylate anions of nylon and silk, increased rapidly with the increasing acidity of the fibre, particularly below pH 5. This condition enhanced the greater substantivity of the dyes to fibre.

Table 3 shows that mordant usage has a good effect on the colour strength of the natural *A. catechu* dyes. The reason for this mordant is influenced by the dye bonding onto the fibre surface. The mordant usage affected the lightness–-darkness (L^*) value and redness and yellowness (a^* and b^* values) of the dyed fabric. In case of mordant usage, the L^* value decreased that means depth of shade becomes darker and the colour strength (f_k) increased. Due to the presence of ferrous sulphate, colour of dyed fabric turned from reddish to greenish. In general, due to the usage of mordants, coordinate covalent bonds are formed (Scheme 2) between dye molecule and fibre and this complex formation is the possible reason for colour change. Alum are considered as neutral mordants so the depth of colour change affected so much, on the other hand iron and copper enhanced colour darker and greener. Ferrous sulphate has shown higher colour strength (f_k) than other mordants.

It can be explained that ferrous sulphate acts as transition metal ions formed a large number of complexes with the dye molecules, because it can be formed a ternary complex ion in one site with the fibre and other site with the dye. Furthermore, a weak co-ordination complex ion formed by alum and copper mordant can block the dye and reduce its interaction with the fibre. The build-up properties of silk and nylon fabric dyed with *A. catechu* dye are compared in Figure 10. The visually-weighted function of *K*/*S* has given

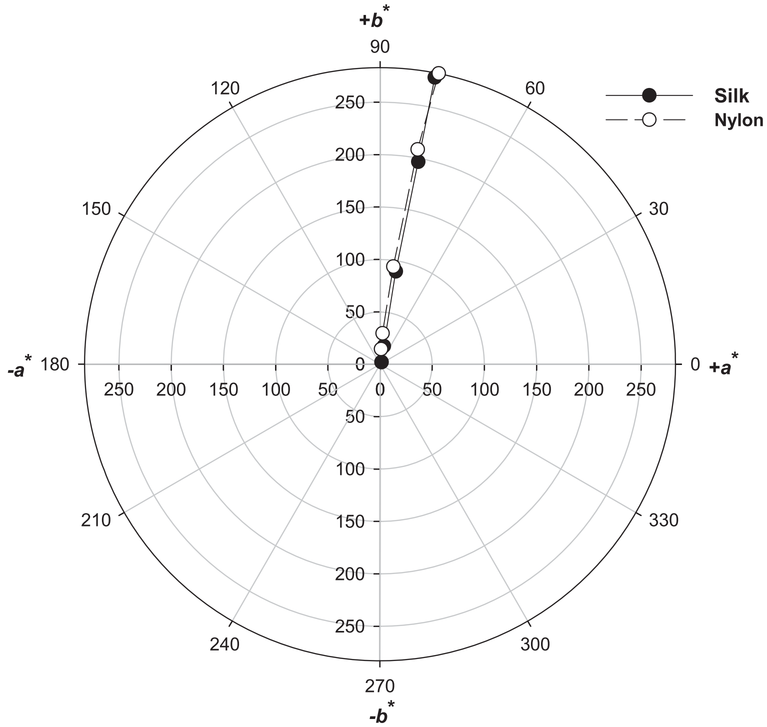


Figure 11. Colour properties (CIELAB *a** and *b**) of the dyed silk and nylon fabrics (dyeing concentration: 0.5, 1.0, 2.0, 4.0 and 6.0% owf, Mordant: $FeSO_4$ (10%), Temp: 70 °C, pH: 4.0 and Time: 50 min).

the colour strength (f_k) values. The upward trend of f_k values is shown in figure that higher concentration of dye increased the higher colour strength.

This result could describe that silk fabric has number of (O = C–NH–) amide group so more coordination complexes ion are formed with the dye than nylon fabric. The saturation point was reached at 4.0% owf for both of the fabrics. Figure 10 illustrates that betel nut extracted dyed silk produced a higher colour strength (f_k 46.41) than nylon (f_k 18.38) at 4.0% owf under the influence of mordant and acidic condition. Therefore, it can be demonstrated that betel nut extracted dye have shown good colour strength with satisfactory uniformity.

The CIELAB *a** and *b** coordinates of the dyed, silk and nylon fabrics are shown in Figure 11. As the dye concentration is increased, the CIELAB *a** and *b** coordinates approach the achromatic point with a zero chromaticity. In particular, the colour coordinates of *A. catechu* dyes have shown similar trend for nylon and silk fabric, dyed fabric turned yellowish to reddish colour in hue angles and chroma appeared lighter to darker when the dye concentration increased.

The colour fastness to washing, perspiration, rubbing, sublimation and light of the dyed silk and nylon fabrics was evaluated. The wash fastness ratings in Table 4 have shown that *A. catechu* dyed silk and nylon provides almost identical fastness ratings of 'good' to 'excellent' (within numerical grades 4–5).

It was expected that the *A. catechu* dyed silk and nylon fabrics comprise an excellent rubbing fastness because of the strong coordination complex bonds formed with the natural dye and fibre. However in case of wet rubbing, the significant colour change has been observed. It can be demonstrated that in deeper shade, dye molecules were more saturated and tend to move out from the interior of the fibre due to rubbing. In addition, in case of light fastness, silk and nylon dyed samples have shown almost identical ratings.

Colour fastness to perspiration (Table 5) of natural betel nut extracted dyed silk and nylon fabrics has shown ratings within the range of 4–5 in case of colour staining on multifibre fabric. The colour fastness to perspiration of *A. catechu* dyed fabric showed lower colour change (numerical rating was 3). This result illustrates that in case of acidic and alkaline perspiration dye performed a halochromic effect, so dye molecules have been changed on the surface of the dyed fabrics.

Conclusions

In this experiment, natural *A. catechu* dyes were extracted and their chemical structure has been confirmed by using FTIR, ^{1}H NMR spectroscopy. The betel nut extracted dye developed yellow to red colours when changing the pH (2.0–11.0). Dyeing on silk and nylon fabric with *A. catechu* dyes in different pH, also appeared the change of shade yellow to red colours. Suitable dyeing parameters of natural *A. catechu* dyes on silk and nylon have been studied. By using the mordants, coordinate covalent bonds are formed between dye molecules and fibres which enhanced the colour strength of the dyed fabrics. Dye build-up and fastness properties of *A. catechu* dyes on silk and nylon have been evaluated and compared. The dyeing on silk fibre performed better build-up properties than nylon, probably due to less hydrophobic effect. The fastness properties of silk and nylon

Table 4. Fastness values of dyed samples with *Areca catechu* (dyeing concentration: 4% owf, Mordant: $FeSO_4$ (10%), pH: 4.0, temp: 70 °C and time: 50 min).

Dyed (4 % owf *Areca catechu*) fabric sample	Washing fastness (ISO 105-C06 A2S:2010)							Rubbing fastness (ISO 105-X12:2016)		Light fastness (ISO 105-B02:2013)
		Colour staining								
	Colour change	Acetate	Cotton	Nylon	Polyester	Acrylic	Wool	D	W	
Silk	4–5	4–5	4–5	4–5	4–5	4–5	4–5	4–5	3–4	4–5
Nylon	4	4–5	4–5	4–5	4–5	4–5	4–5	3–4	3	4–5

Table 5. Perspiration fastness values of dyed samples with *Areca catechu* (dyeing concentration: 4% owf, mordant: $FeSO_4$ (10%), pH: 4.0, temp: 70 °C and time: 50 min).

Dyed (4 % owf *Areca catechu*) fabric sample	Perspiration fastness (ISO 105-E04:2013)						
		Colour staining					
	Colour change	Acetate	Cotton	Nylon	Polyester	Acrylic	Wool
Silk	3	4–5	4–5	4–5	4–5	4–5	4–5
Nylon	3	4–5	4–5	4–5	4–5	4–5	4–5

have demonstrated almost identical results, however the perspiration fastness results attributed to the halochromic effect of the dye. This research endeavour illustrates the potential application of *A. catechu* which is non-toxic, eco-friendly and it has multifunctional properties especially for dyeing the silk and nylon fabrics.

Disclosure statement

No potential conflict of interest was reported by the authors.

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