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IMPROVING THE EFFICIENCY OF *NESOGORDONIA PAPAEVERIFERA* (DANTA) AS A NATURAL DYE IN TEXTILE MAKING INDUSTRY

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ABSTRACT. An attempt was made to extract natural colourant from the bark of *N. papaeverifera.* The optimisation of the extraction procedure was evaluated using various solvents. The optimal colourant extraction was observed with acetone as solvent at constant temperature of 70 °C for 1 h. The main extracted colourant compound was isolated using column chromatography and characterised by ultraviolet-visible spectrophotometer, mass spectrometry, proton and ^{13}C nuclear magnetic resonance spectrometry and Fourier transform infrared spectroscopy. The dyeing and durability of the colourant extract were evaluated. This low-cost biomass was obtained from a timber industry and its extract used to dye cotton and nylon 6,6 fabrics with only small amounts (6% and 4%) of metallic mordants, namely, copper(II) sulfate and stannous chloride. Dyed cotton and nylon were analysed for their K/S, CIE L^*, a^*, b^*, c^*, h^* values and the colour fastness properties to light, crocking (rubbing) and washing. The UV-visible spectral result suggest the presence of such chromophores as C=O and –C=C–. The broad FTIR result at 3596 cm^{-1} is indicating a carbonyl group, the ^{13}C NMR spectrum showed absorption at 206 ppm while the proton NMR gave absorption at 4.37 to 5.24 ppm suggesting –OH protons. Colour shades of brown were obtained. Significant differences in colour depth were observed depending on the mordant type. Copper(II) sulfate was found to produce the most significant colour changes, the deepest brownish colour, and the best light fastness and wash fastness values. The colourant extract itself (without mordant) had a light fastness of 3 (fair) on cotton fabric and 3/4(good) on nylon 6,6 fabric. In general, colour fastness to light was good (grade 5 for cotton and grade 4/5 to 5 for nylon) colour fastness to washing was very good (grade 4 to 4/5 for cotton and grade 3/4 to 4 for nylon) and colour fastness to rubbing was very good (grade 3/4 to 5).

KEY WORDS: Natural colourant, Optimised extraction, Mordanting, Percentage of dye absorbed, Fastness

INTRODUCTION

Researchers have found out that some synthetic dyes are toxic and during the process of their manufacture, large amounts of chemical wastes are produced [1-3]. These processes discharge vast amounts of unsafe synthetic colourants into the environment as demands for coloured textiles increase because of their aesthetic, decorative and utilitarian applications. In recent times, however, the world has become aware of the environmental consequences of many of the synthetic colourants. This has led to an increase in the demand for natural colourants by consumers. The use of natural colourants [4] in both textile and other non-textile uses is a way of supporting sustainable development with respect to health and safety in the environment [5-8]. Nature has an abundance of dye-yielding plants and animal species. However, despite the availability of these natural resources, their identification and scientific study have not been fully harnessed and documented.

Although many plant and animal-based colourants are more environmentally sustainable and non-toxic than synthetic colourants, their poor fastness properties have limited their application as textile dyes [6, 9-11]. This problem has been taken care of by the use of metal salts as mordants in small amounts.

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Nesogordonia papaeverifera (*Family: Sterculiaceace*) is an evergreen decidous plant. It is found widely around the tropical forest of Africa around Nigeria, Sierra-Leone, Cameroon, Gabon and Ghana. The wood of the tree is used for interior and exterior applications such as boat components, general constructions, plywood and furniture making. The application of *N. Papaeverifera* dyeing extract for dyeing textile materials has been reported [12]. The study indicated that the durability of the colourant extract on the textile substrate was poor-to fair. The present study is focused on the optimisation of extraction procedures, mordanting and dyeing properties of the colourant on cotton and nylon 6,6 fabrics with a view to improving the fastness performance of the colourant. The effects of processing agents such as washing, light and rubbing were evaluated to determine the applicability of the colourant in the textile sector. Furthermore, the standardisation of the dyeing process was determined through the evaluation of the % exhaustion method.

EXPERIMENTAL

Plant materials

Among hundreds of dye-yielding plants in Nigeria, *Nesogordonia papaeverifera* was chosen for the study because its ready availability recommends it for use in the textile and other areas of human endeavours. The bark of *N. papaeverifera* was obtained from a saw mill in Sapele, Delta State, Nigeria. The plant was identified by a taxonomist at the University of Calabar, Nigeria (Voucher number = 206/Cal/HRB/1233).

Chemicals

Stannous chloride ($SnCl_2.2H_2O$), and copper(II) sulfate ($CuSO_4.5H_2O$), methanol, acetone, and potassium hydroxide were procured from Sigma Aldrich Chemicals and used for the extraction and dyeing processes.

Instruments

The main colourant component of the extract was characterised by using ^{1}H NMR, ^{13}C NMR (Agilent-NMR-Vnmrs 400), FTIR (FTIR-84000S), mass spectrometry (GC/MS-OP 2010) and UV-visible spectrophotometry. Soxhlet apparatus, round bottom flask, heating mantle, condenser and an electrical grinder were used for the extraction of the colourant while the Gyrowash was used for the mordanting and dyeing processes. The Crock meter (Taber@crockmeter, USA) was used to evaluate the rub fastness of the naturally dyed fabrics. In addition, the GretagMacbath, USA E700 instrument $D_{65}/10°$ was used for colour measurement.

Extraction of colourant

The already processed plant powder sample (10 g) was subjected to solvent extraction using 250 mL acetone as solvent in a Soxhlet extractor and heated at 20, 40, 60, 80 and 100 °C for 1 h respectively until the colour in the extract was negligible. Furthermore, extraction was repeated with other solvents such as water, methanol, a mixture of 60% potassium hydroxide and 60% methanol solution and a mixture of acetone and potassium hydroxide. Plots of extraction temperature and extraction time against absorbance were carried out. The extracts were combined, solvent-stripped and the colour components of the residue separated by chromatography on a silica column, eluting with chloroform/ethanol (70:30), monitoring by thin layer chromatography (TLC) [13].

Mordanting and dyeing

The dyeing and pre-mordanting experiments were carried out [2, 14]. The cotton fabric was first scoured by treating it with a solution in water containing 3% sodium hydroxide and 1.0 gL^{-1} of scouring agent at 100 °C for 60 min and liquor : materials ratio 30:1. On the other hand, nylon 6,6 fabric was scoured by treatment in a solution containing 1.0 gL^{-1} of non-ionic detergent in distilled water at 60 °C for 20 min with liquor ratio 40:1. The cotton fabric (0.6 g) was immersed into 20 mL of 6% aqueous $CuSO_4$ and 4% aqueous stannous chloride solutions, respectively at 1:30 material : liquor ratio at 80 °C for 40 min. The mordanted fabrics were then air dried for 20 min. The colourant solution was prepared with 5% of colourant in 1:50 material : liquor ratio and mordanted cotton fabric was immersed into 100 mL of colourant solution. Dyeing was carried out at 85 °C for 1 h. The dyed cotton samples were washed with cold water and then with soap solution, followed by thorough washing with water. The samples were dried at room temperature [15].

Mordanted nylon 6,6 fabric was dyed with 5% of colourant solution whose pH was adjusted to 5 with formic acid to promote metal fixation in the nylon substrate. Dyeing started at 40 °C and the temperature was raised to 100 °C and maintained as such for 1 h. The dyed nylon samples were then washed with water followed by soap solution and again several times with water until the washing water became clear. The dyed wet samples were then dried in air. Direct dyeing of the substrates was also performed as a control.

Percentage of dye absorbed

Ultraviolet-visible absorption measurements were done to determine percentage of colourant absorbed using Perking Elmer Lambda 35 UV-vis spectrophotometer at 220-800 nm wavelength. The absorbance of 5% colourant solution was measured before and after dyeing the cotton and nylon 6,6 fabrics, taking an average of three readings. The amount of colourant absorbed was calculated by using equation 1 [16].

$$\%A = \frac{A_0 - A_1}{A_0 \times 100} \quad (1)$$

where %A is percentage of dye absorbed, A_o and A_1 are the absorbances before and after dyeing.

Colour measurement

The colour measurement values of CIE L^* a^* b^* c^* h^* and values of reflectance of dyed samples were measured on a Gretag Macbeth (USA) E700 instrument ($D_{65}/10^o$) and the colour yield expressed as K/S of dyed samples were calculated using the Kubelka-Munk equation:

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

where R is the observed reflectance, K is the absorption coefficient and S is the light scattering coefficient.

Colour fastness test

The test for effect of washing was determined by the ISO 105 - CO3 standard test method [17], during which the test specimen was stitched together with two pieces of undyed fabrics (cotton and nylon) and then washed with a standard soap solution at 60 °C for 30 min. The

combinations of the specimen and the tested fabrics were washed with water and dried. The extent of staining on the adjacent fabrics was assessed by comparing with the greyscale (1-5), where 1 and 5 represent poor and excellent, respectively.

To test for light fastness, the dyed samples were exposed to sunlight continuously along with a set of light-sensitive blue wool standards for 72 h, in accordance with ISO 105 – BOI: 2014 method [18]. The standards are usually designed to fade after given periods of time. The changes in colour of the coloured fabrics were evaluated by comparing with the blue scale for light fastness (1–8). Colour fastness to rubbing was tested according to ISO 105 – X12 method [19]. The test was determined using a Crock meter, during which the dyed fabric was subjected to rubbing treatment with a sample of standard undyed cotton. Two sets of tests were carried out, one with wetted cotton fabric and the other with dry cotton fabric. The extent of colour transfer was assessed based on the greyscale rating (1–5) where 1 and 5 represent poor and excellent, respectively.

RESULTS

Optimization of solvent extraction

Several experiments were performed for the extraction of colourant from *Nesogordonia papaeverifera* stem bark using water, acetone, mixture of potassium hydroxide solution and methanol solution. The results of the experiments are shown in Figures 1 to 4. It was observed that the yield of the natural dye using acetone alone as solvent was higher compared to those obtained using other solvents or water mixed with other additives.

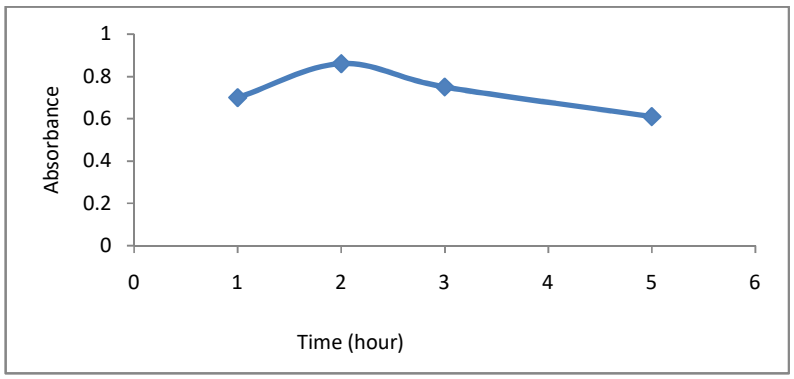

Figure 1. Effects of dyeing time on absorbance of dye extract under conditions of 70 °C, 1 h and acetone (300 mL).

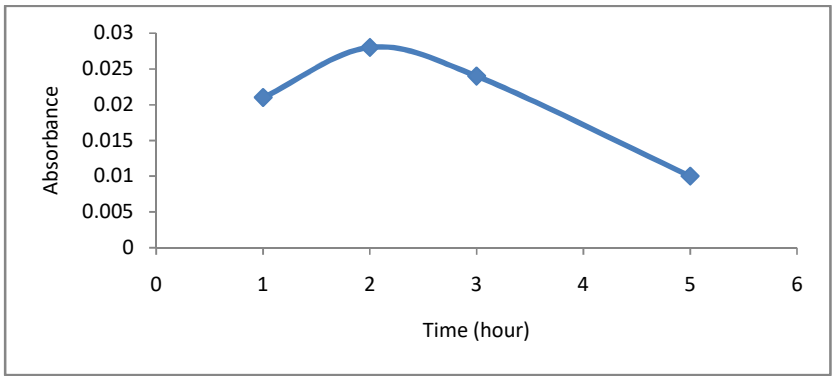

Figure 2. Effects of dyeing time on absorbance of dye extract under condition of 70 °C, 1 h and 80% methanol.

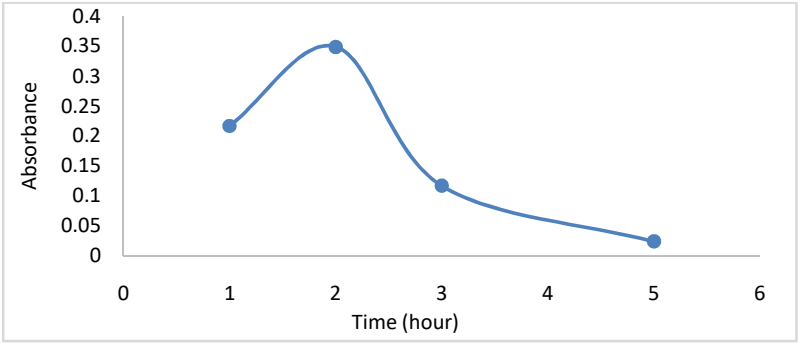

Figure 3. Effects of dyeing time on the absorbance of dye extracts under conditions of 70 °C, 1 h and water.

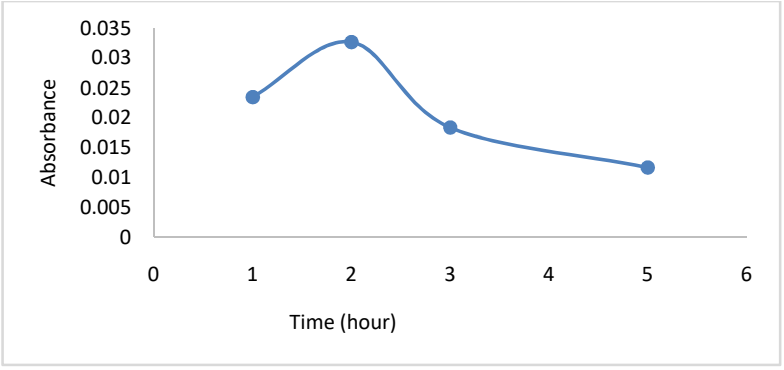

Figure 4. Effects of dyeing time on the absorbance of dye extracts under conditions of 50% KOH and 50% methanol and 1 h.

From Figure 1, 2, 3 and 4 it could be seen that the optimal extracting efficiency of the colourant from the stem bark of *N. papaeverifera* could be obtained using acetone as solvent within 1 hour at 70 °C.

Characterization of colourant extracts

The FT-IR spectrum of the extracted colourant showed a broad peak appearing at 3596 cm^{-1} region in the IR spectrum, which is suggestive of the O-H stretching vibrations of the –COOH group. The stretching vibration of C-H is illustrated by the peak at 2963 cm^{-1}. This indicates that the colourant structure has hydrogen atoms attached to sp^2 carbon atoms [19]. The absorption observed at 1649 cm^{-1} is assignable to the carbonyl (C=O) group of carboxylic acid. The bending vibrations of C-H are illustrated by the peaks at 2528, 2324, 2130, 1413 and 1280 cm^{-1}. The absorption band at 1059 cm^{-1} suggests the presence of C-O stretch. The ^{1}H NMR spectrum showed a singlet at δ 1.65-1.67 ppm suggests the presence of CH_3 protons while the singlets at δ 3.99 and 4.61 ppm indicate $CH_3$and CH_2 protons, respectively. The two broad peaks at δ 4.37-5.24 ppm suggest O-H protons [20]. The singlet at δ 5.94 ppm suggests the presence of CH_2 protons close to an electronegative atom or vinylic hydrogen atoms. The ^{13}C NMR spectrum gave an absorption at δ 17.9 ppm suggests methyl carbons whereas the absorptions at δ 29.1, 29.3 and 29.5 ppm were all assigned to vinylic (CH) (sp^2) carbons. The signals observed at δ 54.2 and 57.2 ppm suggest alkoxy groups since the δ value were shifted to higher absorptions. The absorption peak observed at δ 206.0 ppm is suggestive of the sp carbons of the carbonyl (C=O) group. This established the presence of the –COOH group observed in the broad IR band at 3596 cm^{-1}. The UV-visible data, indicates that there were absorptions at 304 and 748 nm which suggest the presence of chromophores such as C=O and (-C=C-), respectively. In addition, the GC/MS results of the colourant extract, indicates that the molecular ion peak was observed at m/z 430.

Table 1 shows the result of the shade light and rubbing fastness properties of the colourant on dyed and mordanted cotton and nylon fabrics. It was observed that the unmordanted cotton fabric gave fair light fastness of grade 3, dry rubbing fastness of grade 3/4 and wet rubbing fastness of grade 3. However, the introduction of 4% $SnCl_2$ and 6% $CuSO_4$ as mordants increased the results to grade 5 and grade 4/5. The results obtained for the wet rubbing fastness was observed to be grade 3.

Table 1. Shade, light and rubbing fastness properties of dyed and mordanted cotton and nylon 6,6 fabrics.

Fabric	Mordant	Shade	Light Fastness	Rubbing fastness	
				Dry	Wet
Cotton	Direct	Brown	3	3/4	3
	$SnCl_2$	Brown	5	5	3
	$CuSO_4$	Brown	5	4/5	3
Nylon 6,6	Direct	Brown	3/4	3/4	3/4
	$SnCl_2$	Brown	4/5	5	4/5
	$CuSO_4$	Brown	5	5	3/4

The wash fastness result of the dyed cotton and nylon fabrics are listed in Table 2. The alteration in colour for the unmordanted fabrics were found to be grade 2/3 for cotton and grade 2 for nylon while both had ratings of grade 3/4 for staining of adjacent fabrics. However, with the introduction of mordants, the alteration in colour increased to grade 3/4 and 4 for cotton and nylon fabrics, respectively.

Table 2. Wash fastness properties of dyed and mordanted cotton and nylon 6,6 fabrics.

Fabric	Mordant	Shade	Change in colour	Staining of adjacent fabric
Cotton	Direct	Brown	2/3	3/4
	$SnCl_2$	Brown	4	4/5
	$CuSO_4$	Brown	4/5	5
Nylon 6,6	Direct	Brown	2	3/4
	$SnCl_2$	Brown	3/4	4/5
	$CuSO_4$	Brown	4	4/5

The percentage exhaustion values of 5% concentration of colourant with dyed and mordanted nylon and cotton are listed in Table 3. The results show that with direct dyeing the percentage exhaustion of colourant on cotton and nylon were 56 and 46, respectively. When copper(II) sulfate was used as mordant, the percentage exhaustion increased to 76 for cotton fabrics and 60 for nylon fabrics. For stannous chloride as mordant, the percentage exhaustion was 68 for cotton and 58 for nylon.

Table 3. Percentage of dye absorbed of 5% concentration of natural colourant with dyed and mordanted cotton and nylon 6,6 fabrics.

Fabric	Mordant	% of dye absorbed (% exhaustion)
Cotton	Direct	56
	$CuSO_4$	76
	$SnCl_2$	68
Nylon 6,6	Direct	46
	$CuSO_4$	60
	$SnCl_2$	58

The CIE L^*, a^*, $b^*$$c^*$ $h^*$and K/S values of the dyed textile samples are given in Table 4. It is seen from the results that mordant usage affected both the lightness-brownness of L^* value and nuance (a^* and b^* values) of the colour. In terms of mordant dyeing, the L^* values decreases and the yield K/S increases. The results obtained, indicate that the colour gets more brownish. The most important change in colour occurs in a^* and b^* values. It is seen from Figure 5, and most importantly in the presence of $CuSO_4$ that the brown colour turns from light brown to deeper brown shade.

Table 4. Colorimetric data L^*, a^*, b^*, c^*, h^* and K/S values of the dyed samples.

Materials	Mordant type	L^*	a^*	b^*	c^*	h^*	K/S
Cotton		80.5	4.9	10.7	11.7	58.5	2.2
	$CuSO_4$	72.1	5.6	12.5	13.8	65.2	4.6
	$SnCl_2$	64.3	3.2	14.6	14.9	80.9	4.0
Nylon		70.4	7.3	14.7	15.4	61.4	1.5
	$CuSO_4$	65.5	4.6	16.5	18.2	73.5	4.2
	$SnCl_2$	61.6	1.8	11.8	12.5	81.6	3.3

DISCUSSION

This paper gives the optimization of extraction of natural colourant from *Nesogordonia papaeverifera* using various solvents and application of the colourant on cotton and nylon fabrics. The optimization of extraction procedure forms the basis of the increase in the concentration of colourant molecules, thus enables improvement on the fastness properties of the colourant on the fabrics. The characterization of the major extracted colourant compound showed the presence of some groups such as O-H, C-H and C=O, which are part of the colourant structure, and these could interact with the textile fibre during dyeing.

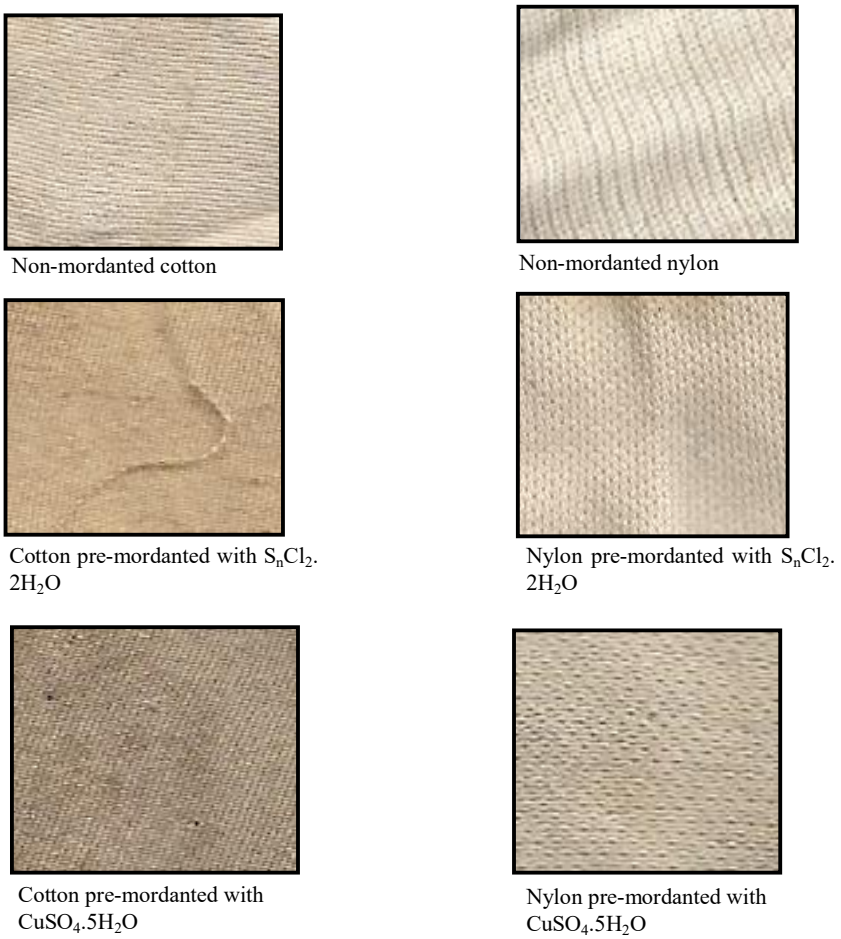

Figure 5. Images of dyed fabrics.

In terms of the overall fastness performance of the colourant on the two dyed textile materials (cotton and nylon), good to excellent results were obtained. This indicates a better fastness performance of the colourant on the fabrics compared to those obtained in the previous study with the same colourant [12]. This is attributable to the increase in the colourant concentration in the dyed cotton and nylon fabrics due to the optimization of extraction procedures. The concentrations of the Cu(II) ions and Sn(II) ions in the spent bath solutions were not estimated to ascertain any ecological problems they could cause, hence, metal mordants fixation in the cotton and nylon fabrics (take up) was ensured by the addition of an organic acid (glacial acetic acid) to increase the pH, thus making the spent bath less harmful to the environment. Mordants form a stronger bridge between the fibre and colourant, thus they inhibit photofading of the colourant via catalytic processes, and in this way, light fastness is improved. The colour hues produced on nylon and cotton fabrics by mordanting separately with the two salts showed significant improvements compared with those of non-mordanted samples. As a result, it can be concluded that pre-mordanting is a very useful process for obtaining deeper and more durable colours in cotton and nylon with the natural colourant from *N. papaeverifera* stem bark.

Regarding the percentage exhaustion of the colourant on the cotton and nylon fabrics, it has been reported [20] that most natural colourants show little intrinsic affinity for fibres obtained from cellulose such as cotton without the use of mordants. However, the percentage exhaustion of the colourant on cotton fabric was observed to be more than 50% even without the use of mordants. This, therefore, indicates that the colourant under study has sufficient natural affinity for cotton fibre. The shades of brown exhibited by the colourant on the dyed cotton fabrics and nylon fabrics corroborated this observation. Moreover, the use of mordants further increased the percentage exhaustion of the colourant to more than 70% on cotton and 60% on nylon. The effects of the Cu(II) and Sn(II) salts as mordants show that Cu(II) as mordant exhibited better technical performance compared to that of Sn(II), hence, the mordant type has a direct correlation with the colourant% exhaustion as well as the type of fibres.

Figure 5 shows the scanned images of dyed cotton and nylon fabrics using the two dyeing methods. The results indicate that adding the sequence of the salt mordants to the dyeing bath could affect the colour appearance of the dyed cotton and nylon fabrics. The alteration in colour depth and appearance was ascribed to the methods of dyeing, during which there was interaction of mordant with natural dyes and cotton and nylon fabrics respectively. Overall, the copper mordanted method was preferred for the dyeing of both cotton and nylon fabrics with the extracted natural dye due to the production of deeper brown shade.

Finally, with regards to the colorimetric changes, it was found that there was increase in the K/S values of the colourants on the dyed cotton and nylon fabrics after mordanting. The K/S values of the pre-mordanted samples are satisfactory and higher than those of the non-mordanted ones. This can be explained by the fact that mordants form a strong bridge between the fibre and the colourant. Furthermore, it was observed that the K/S values of the $CuSO_4$ mordanted samples were higher than those of the $SnCl_2$-mordanted samples. This indicates that mordant types exhibit variable colourant affinity and thus can enhance exhaustion of natural colourants on textile fibres.

CONCLUSION

Nigeria is endowed by nature with many colourant yielding plants yet to be exploited and developed. Natural colourant was extracted from *N. papaeverifera* and optimal extraction was achieved at 70 °C with five extractions in the presence of acetone under 1 h each. *N. Papaeverifera* colourant extract was used with and without mordants for the dyeing of cotton and nylon 6,6 fabrics, with very good fastness properties. The major colouring component obtained by column chromatography was characterised using UV-vis spectrophotometry, FTIR spectroscopy, ^{1}H, and ^{13}C NMR and mass spectrometry to identify its chemical groups. The UV-visible spectral results suggest the presence of chromophores such as C=O and -C=C-, respectively. Although light fastness results showed that $CuSO_4$ and $SnCl_2$ improved the photostability of the colourant on nylon 6,6 and cotton, it is significant that the non-mordanted fabrics also exhibited fair-to-good results, thus it may be concluded that *N. papaeverifera* bark could be used as a low-cost and easily accessible source of natural colourant for textile dyeing.

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