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THE USE OF THE BARK OF *Byroscarpus coccineus* Schum AND Thunn AS NATURAL DYE SOURCE FOR NATURAL AND SYNTHETIC FIBRES

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ABSTRACT
Natural colourant was extracted from the bark of *Byrsocarpus Coccineus* Schum. Thunn. Analysis of the designed experiment showed that extraction could reach an optimal production of natural colourants when extraction was carried out at 100 ^{o}C for 4 hrs in the presence of 15 gl^{-1} of sodium hydroxide solution under reflux. The extracted natural colourant was applied to the dyeing of polyethylene terephthalate (PET), nylon 6, and cotton fabrics using different methods, with or without a mordant. It was found that mordants had a significant effect on the colour of dyed textile fabrics (PET, nylon 6, and cotton). The textile fabrics dyed with the pre-mordant technique using Copper Sulphate, stannous chloride and Potassium dichromate showed higher fastness performance compared to the other mordants. An interesting aspect of the results is that a light fastness of grade 3 (fair) was observed on all the three textile substrates used for the study without using a mordant. The *B. Coccineus* bark colourant extract gave a bright pink colour on the textile substrates. Thus, the results of this study show that the colourant extract possesses potential affinity for both natural and synthetic fibres and could be exploited in the textile industry.

Keywords: *Byroscarpus coccineus*, natural dyes, characterization, dyeing methods, fastness properties.

INTRODUCTION

The use of natural dyes dates back to prehistoric times for many purposes especially in Europe, Africa and Asia. In fact, the utilization of natural dyes was part of Europe's cultural identity. For instance, naturally dyed textiles were once symbols of status (Casas-catalan and Domenech-carbo, 2005). They were not only expensive but were reserved for the Wealthiest class of people. One of the limitations of natural dyes is that they are presently produced at a rate not comparable to those of synthetic dyes to meet future demands (Ana *et al.,* 2007). Thus the need for the production of natural plant dyes must involve large agricultural land through direct farming (Haji *et al.,* 2013; Yi and Chao, 2008). Recently, there has been renewed attention paid to natural dyes by researchers and manufacturers alike due to their obvious safety advantages over synthetic dyes. Thus, consumers of textile fibres (such as cotton, silk, wool and synthetic fibres such as polyester, polyamide and polyacrylonitrile apparels) are now more than ever, interested in using textiles dyed with natural dyes. In addition, some natural pigments of annatto, cutch, pomegranate fruit rind and golden duct were reported to have been used as colorants for the preparation of water-based ink-jet inks for digital textile printing. These inks have been found to exhibit the same colour

consistency and fastness results especially after fixation, comparable to those of synthetic dyes. They have also been found to be mostly suitable for the digital printing of cotton fabrics (Savvidis *et al.,* 2014).

In another method of printing, natural dye extracts of the leaves and bark of eucalyptus plant has been reportedly used to carry out screen printing of Lyocell fabrics and the printed fabrics interestingly showed good fastness towards light, washing and rubbing (Ellams *et al.,* 2014).

It is possible to dye synthetic fibres such as nylon, polyester and acrylic fibres with selected natural dyes. However, very few natural dyes have high affinity for synthetic fibres and deep and bright shades have been achieved. For instance, the anthra-quinone based dyes have been found to show very good fastness properties (Hobson and Wales, 1998). Also, non-polar natural dyes such as lawsone, and juglone exhibited similar fastness behaviours as disperse dyes on polyester. (Ebrahimi and Ghashti, 2015).

The natural dye obtained from Annatto is an acid dye, but usually exhibits different behaviours on different textile fibres. For instance, it behaves as an acid dye on nylon fabric and as a disperse dye on polyester. In each of these cases, metal salts were used to improve the absorption of the colourant on the textile substrates (Haji *et al.,* 2013).

Some other drawbacks of dyeing with natural dyes, are low dye uptake and poor light fastness (Otutu, 2008; Wang *et al.,* 2012). It has been reported that the use of biomordants could be a possible approach to improving the dye uptake and colour fastness (Savvidis *et al.,* 2014) of natural coloured textiles. According to Dalby (1993), however, the use of biomortants alone have not been efficient enough compared to the traditional metal mordants such as alum, Cu, Cr and Fe to bring out the much needed dye uptake and colour fastness of naturally dyed textiles (Ali, 1993; Glover and Pierce, 1993). There is therefore, the need to develop a more effective and efficient method of extracting natural dyes in order to achieve deeper shades with the concomitant higher dye molecules concentration in the dyed textile materials in terms of optimisation and standardization processes.

According to Literatures, the plant *B. coccineus* belongs to the family Connarceae and it has been reported to have various uses including beautification or ornamental value (Burkill, 1985), treatment of earache, jaundice and veneral disease (Akindele *et al.,* 2014), skin and mouth problems, gonorrhea, urinary tract infections, German measles, anaemia, primary and secondary sterility in both male and females, cancers, diarrhea, dysentery (Dalziel and Hutchinson, 1995), kidney problems, diuretics, antiplasmodial activity (Akpan *et al.,* 2012), mollucidal, uterotonic, anti-inflammatory and anti-microbial activities (Okonji and Iwu, 1998; Amos *et al.,* 2002; Akindele and Adeyemi, 2007; Ahmadu *et al.,* 2006).

Although known for a long time for its medicinal properties, no study has been conducted so far to investigate the use of the plant for the purpose of producing natural dyes. Hence this study was undertaken to investigate the possible use of *B. coccineus* as a potential source of natural dues. Emphasis was also placed on the optimization of extraction processes as well as the use of various dyeing methods. The use of *B. coccineus* as a natural dye source would be beneficial not only because of its medicinal values but also the fact that it can easily be propagated vegetatively by stem cuttings.

EXPERIMENTAL

Materials

Polyester (100% terylene), nylon 6, and cotton fabrics were purchased from a local market (Nigeria). The stem bark of *B. coccineus* was collected from Obiaruku in Delta State (Nigeria). Sodium hydroxide (NaOH), Acetone (CH_3COCH_3), Copper (II) sulphate pentahydrate ($CuSO_4.5H_2O$), potassium dichromate ($K_2Cr_2O_7$), potassium aluminium, sulphate ($KAI(SO_4)_2.12H_2O$), potassium sodium tartrate ($KNaC_2H_4O_6$) acetic acid (CH_3COOH) were all

of analytical grade and obtained from Merck (Germany).

Extraction of natural dye

The bark of the *B. coccineus* Schum Thonn was cleaned with enough water and dried in an oven. The dried bark of the plant was ground to form powder by using an electrical grinder with a steel blade. The average particle size was measured to be 52.35 μm. Extraction was carried out under the following conditions: the concentration of sodium hydroxide was (5 – 30 g/l) at 100 ^{o}C and time of 4 hours respectively was used.

Acetone (300 ml) was also used as an extraction solvent to extract natural dyes from the stem bark of *Byroscarpus coccineus* (10 g) each at 10^{0}C, 20^{0}C, 30^{0}C, 40^{0}C, and 50^{0}C, respectively and a plot of absorbance against extraction temperature was carried out (Bechtold *et al.*, 2006).

Absorbance measurement of dye extract

The λmax of the maximum absorbance of an extract was measured by an ultraviolet (UV) 3300 spectrophotometer (Hitachi, Japan) and was set at 400 nm. The absorbance of the colourant extract at 400 nm was correlated with the natural colourant concentration in the extract. Thus, the absorbance at 400 nm was determined to detect any change in concentration of the natural colourants in the extracts.

Dyeing

The natural dye extracted under the optimal conditions was dried in a vacuum oven at 100^{0}C to give the crude solid dye. The crude product was purified by soxhlet extraction with acetone. A 10 ml of this solution was used for the dyeing of the various fabrics.

Direct dyeing of cotton

The raw cotton fabric (100%) with the following characteristics: thickness under 1 kPa pressure, 0.27 mm; yarn number, wrap 14 tex, weft 16.5 tex; fabric counts, wrap 31 threads/cm, weft threads/cm; weight, 115 g/m^{2}, was placed in boiling water (1*l*) to which soap flakes and sodium carbonate (1.50 g) were subsequently added. The mixture was then heated for 1 h. The cotton fabric was then removed, washed with hot water and cold water, squeezed to remove excess liquor and air-dried. The fabric was then treated with 1 M hydrochloric acid at room temperature for 30 mins, removed, washed with deionized water until the rinsed water was neutral and dried at room temperature.

The cotton fabric was added to a bath containing 10 ml dye solution at a liquor ratio of 30:1 at 90 ^{0}C. Sodium sulphate was added to the bath to sufficiently increase exhaustion. Dyeing temperature was increased to 100 ^{o}C and maintained for 60 mins. The dyed fabric was then removed from the bath, rinsed with cold water soaped off. A second rinsing with cold water was carried out followed by drying in a vacuum oven.

Direct dyeing of nylon 6

Nylon 6 fabric was scoured in a solution of 1.0 gl^{-1} non-ionic detergent (Liss apol N) for 1 hour at 60 ^{0}C for 30 mins, washed with and air-dried. Dyeing was carried out in a shaking water-bath under the following conditions: temperature 90 ^{0}C; liquor ratio 30:1 time (min) 60; concentration, 10 ml and pH of 4 adjusted with acetic acid. The dyed nylon 6 fabric was soaped off using the non-ionic detergent, rinsed with water and air-dried.

Direct dyeing of Polyester

The polyester fabric was first treated in a soap solution for 1 hour at 100 ^{0}C, rinsed with water and air-dried to remove impurities. The polyester was then dyed in a dye bath containing 10 ml of dye solution and 1.0 gl^{-1} of anionic dispersing agent with liquor ratio of 30:1. The pH of dye liquor was adjusted to 5 using dilute acetic acid. The polyester fabric was introduced into the dye bath at 50 ^{0}C, and the temperature was raised at 2 ^{0}C/min until the maximum temperature of 130 ^{0}C was reached and then kept constant for 60 mins in a high-temperature high-pressure laboratory dyeing machine. The dye bath temperature was then cooled to 80 ^{0}C, and the samples were removed from the dye

bath and rinsed thoroughly with water, after which the dyed fabric was soaped in a mixture of 2.0 gl^{-1} of non-ionic detergent and 2.0 gl^{-1} of sodium carbonate using a liquor ratio of 30:1 at 70 ^{0}C for 30 mins, rinsed with water and air-dried.

Pre-mordant dyeing

The three types of fabrics: polyester, nylon 6 and cotton were treated in a bath containing 0.6 g alum (dissolved in warm (55 ^{0}C) water) and 0.2 g potassium sodium tartrate in 100 ml water, and another bath containing 0.2 g $CuSO_4$, $SnCl_2$ (0.2 g) ($K_2Cr_2O_7$) (0.2 g) respectively. The different fabric samples were each wetted with water and added to the mordant solutions respectively. This was performed at 70 ^{0}C for 1 h in a liquor ratio of 30:1. After the pre-treated samples were rinsed with cold water and dried in a vacuum oven, they were respectively added to a bath containing 10 ml of dye solution with liquor ratio of 30:1 at 80 ^{0}C 60 min for cotton fabric, 90 ^{0}C for nylon 6, and 130 ^{0}C for polyester fabrics at the appropriate pH values (adjusted using acetic acid).

Post mordant dyeing

The three fabrics: Polyester, nylon 6 and cotton were differently added to a dye bath containing 10 ml dye solution at a liquor ratio of 30:1 at 130 ^{0}C for polyester; 90 ^{0}C for nylon 66, and 80 ^{0}C for cotton fabric respectively at the appropriate pH values (adjusted using acetic acid). Dyeing was carried out for 60 mins. The dyed fabrics were gently rinsed with cold water and then added to a bath containing 0.6 g Alum and 0.2 g Potassium sodium tartrate respectively.

One-bath dyeing

Polyester, nylon 6 and cotton fabrics, each was added separately to a bath containing 10 ml dye solution and 0.6 g Alum and 0.2 g potassium sodium tartrate at liquor ratio of 30:1 at 80 ^{0}C for cotton fabric, 90 ^{0}C for nylon 6 fabric and 130 ^{0}C for polyester fabric at 60 mins.

All the dyed samples were subjected to sequential treatments; rinsing with cold water, washing in a bath that contains non-ionic detergent (2.0 gl^{-1}). Liquor ratio 30:1 at an ambient temperature for 20 mins. The samples were also rinsed with cold water and dried in a vacuum oven.

Fastness Standard Tests

The Wash fastness of the dyed fabrics was tested according to AATCC test method 61 – 1996 (AATCC, 1996). Light fastness was measured using Atlas Xenotest Alpha for light fastness (ISO, 1994). The rubbing fastness of the dyed fabrics was tested according to ISO 105-X12 test method (ISO, 2001). The changes in shades under artificial light were evaluated according to the standard blue wool fabrics (SDC) methods.

Characterization of dye extract

The Fourier Transform-infrared (FTIR) spectrum was measured on a Shimadzu 8400S 2010 series. The UV-visible spectrum was recorded on a Hitachi UV-3300 spectro-photometer (Japan). The GC MS of the dye extract was recorded using Shimadzu-QP 2010 series.

RESULTS

Figure 1 below shows the effect of $NaOH_{(aq)}$ concentration on the absorbance of the dye extract under conditions of 80 ^{o}C and 4 h. It was observed that the optimum absorbance was found to be 4.137 when 20.0 g/L of NaOH was used. Similarly, the effect of varying the temperature of extraction of the natural dye using acetone as solvent in a soxhlet apparatus is shown in the Figure 2 below. The result showed that at 20 and 30 ^{o}C, the maximum absorbance was constant (1.456). Thus, comparing the extraction results in figure 1 and 2, it was observed that optimum extraction of dye from the bark of B. coccineus could be better achieved using acetone at a temperature of 20 – 30 ^{o}C in 4 h.

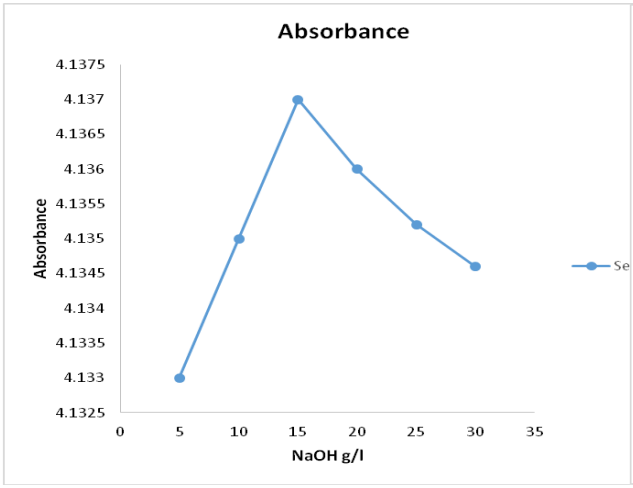

Figure. 1: Effect of NaOH concentration on absorbance of extract under the conditions of 80 ^{0}C and 4 h

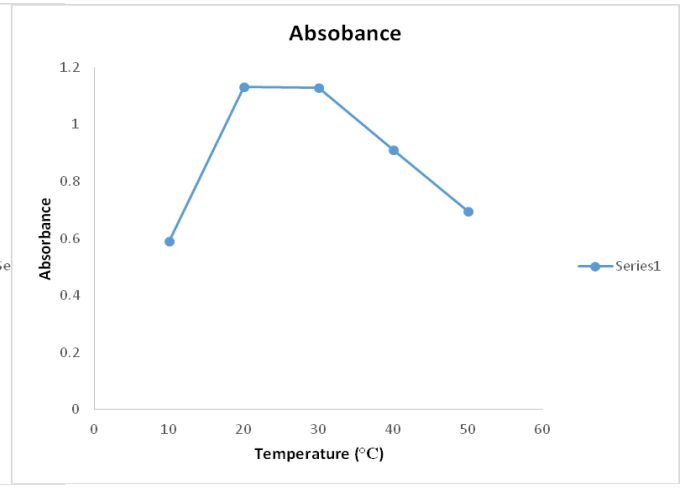

Figure 2: The Plot of Absorbance against extraction temperature (^{0}C) with acetone as solvent

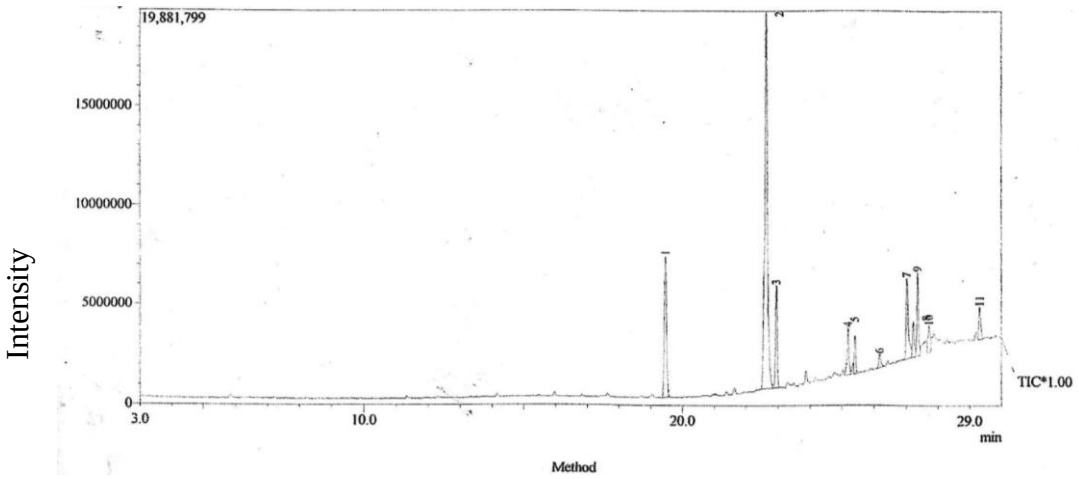

Figure. 3: GC/MS Chromatograph of dye extract

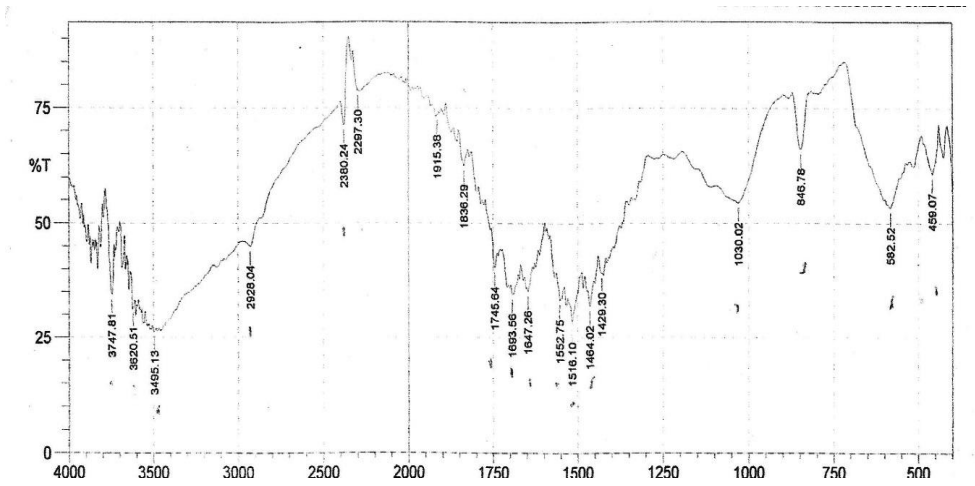

Figure. 4: FTIR spectrum of dye extracts

The results of the GCMS analysis (Figure 3) gave different M/z ratios corresponding to the different fragments in the extracted dye compound as follows:
m/z: 87(14.4%), 97(38.78%), 298(10.41%), 129(4.83%), 326(3.91%), 147(1.40%), 129(8.22%), 166(3.65%), 87(8.51%), 149(2.65%), 382(3.22%) (M^+).

Figure 4 shows the FTIR spectrum of the *Byroscarpus coccineus* stem bark dye extract. The sharp band at 3747 cm^{-1} corresponded to the N-H stretching vibrations in the dye structure. The broad bands at 3495-3620 cm^{-1} region were assigned to the –OH stretching vibrations. In the IR spectrum of the dye extract, vibrations occur as sharp medium band at 2928 cm^{-1} which is due to the C-H stretching vibration; the absorbance maximum at 1745 cm^{-1} was assigned to C=O functional group; the bands at 1647 – 1693 cm^{-1} region suggests the presence of aryl ketone stretching vibrations. The IR spectrum of the dye extract also showed absorption band at 1552 cm^{-1} indicating the presence of amide N-H. The band at 1516 cm^{-1} is indicating the in-plane bending vibration of C-H group. The C-O stretching vibration is shown at 1030 cm^{-1} confirming the presence of –OH group. The strong band at 582 – 846 cm^{-1} region is assigned to aryl or alkyl halides (Paula, 1995).

The absorption maximum of the dye extract was observed at 419 nm, which is in the visible region. This is in line with the pink colour observed on the dyed textile fabrics.

Tables 1, 2 and 3 show the colour fastness properties of the dyed nylon 6, cotton and polyester fabrics respectively. As shown in Tables 1, 2 and 3, the colour change of the wash fastness test results was generally found to be 4 and 4-5 in the three textile fabrics (nylon 6, cotton and polyester). The colour fastness to rubbing values of the dyed polyester fabrics were generally higher compared with those of nylon 6 and cotton fabrics. The light fastness values for the fabrics were generally quite similar (4 and 3-4) and 3 (fair) when dyed without mordant.

Table 1: Colour Fastness of dyed nylon 6 fabrics

Dyeing Method	Mordant	Colour fastness to washing	Colour fastness to rubbing		Light fastness
			Dry	Wet	
Direct	No mordant	4	4-5	4	3
One-bath	KAI $(SO_4)_2.12H_2O$	4	4-5	4-5	3-4
Post-mordant	KAI $(SO_4)_2.12H_2O$	4	4	4-5	3-4
Pre-mordant	KAI $(SO_4)_2.12H_2O$(0.3g)	4	5	4-5	3-4
Pre-mordant	KAI $(SO_4)_2.12H_2O$(0.6g)	4-5	5	4-5	4
Pre-mordant	$CuSO_4.5H_2O$	5	5	4-5	4-5
Pre-mordant	$SnCl_2.H_2O$	4-5	5	4-5	3-4
Pre-mordant	$K_2Cr_2O_7$	5	5	4-5	4-5

Table 2: Colour Fastness of dyed cotton fabric

Dyeing Method	Mordant	Colour fastness to washing	Colour fastness to rubbing		Light fastness
			Dry	Wet	
Direct	No mordant	4	4-5	4	3
One-bath	KAI $(SO_4)_2.12H_2O$	4	4-5	4	3
Post-mordant	KAI $(SO_4)_2.12H_2O$	4	4-5	4	3
Pre-mordant	KAI $(SO_4)_2.12H_2O$(0.3g)	4	4-5	4	3
Pre-mordant	KAI $(SO_4)_2.12H_2O$(0.6g)	4	4-5	4	3

Pre-mordant	$CuSO_4.5H_2O$	4	4-5	4-5	3-4
Pre-mordant	$SnCl_2.H_2O$	4	4-5	4	3
Pre-mordant	$K_2Cr_2O_7$	4-5	4-5	4-5	3-4

Table 3: Colour Fastness of dyed Polyester

Dyeing Method	**Mordant**	**Colour fastness to washing**	**Colour fastness to rubbing**		**Light fastness**
			Dry	**Wet**	
Direct	No mordant	4	4-5	4	3
One-bath	KAI $(SO_4)_2.12H_2O$	4	4-5	4-5	3-4
Post-mordant	KAI $(SO_4)_2.12H_2O$	4	4	4-5	3-4
Pre-mordant	KAI $(SO_4)_2.12H_2O$(0.3g)	4	4	4-5	3-4
Pre-mordant	KAI $(SO_4)_2.12H_2O$(0.6g)	4-5	5	5	4
Pre-mordant	$CuSO_4.5H_2O$	4-5	5	4-5	4-5
Pre-mordant	$SnCl_2.H_2O$	4-5	5	4-5	4
Pre-mordant	$K_2Cr_2O_7$	4-5	5	4-5	4-5

It can be seen from figure 5 that there is an increased colour shade of the natural dye on the dyed fabrics in all the dyeing methods including without mordants. This may be attributed to the fact that the natural dye extract from *B. coccineus* has more affinity for the nylon 6 fibre by comparison with the other two fibres (polyester and cotton).

It was observed that the light fastness values for the dyed cotton fabrics were lower compared with those of nylon 6 and polyester fabrics. This may be due to the lack of groups that could interact with the mordants and the fabrics.

DISCUSSION

The application of colours, whether natural or synthetic is one of the keys to commercial success of textile products. Many synthetic dyes do not conform to safety laws, and now more than ever, emphasis is being focused on having environmentally safe products while continuing to provide an effective product. The demand for natural dyes is increasing as more areas of natural dye application is being discovered by researchers and manufactures.

Most natural dyes do not give good, strong colours on cellulose fibres and modern fibres without the aid of either a mordant or a fixative. And since the use of biomordants alone have been found not to be efficient enough, it is believed that a plant dye in addition to a metal mordant extracted under optimised conditions will further increase the dye uptake and colour fastness properties (Velmurugan *et al.*, 2013).

Copper (II) sulphate and potassium dichromate produced the most significant colour changes and best light fastness values on the three textile fibres used for the study. In terms of the colour and fastness properties, the metal mordants concentration in the spent bath were considerably reduced by the addition of acetic acid to the mordant bath in order to reduce the ecological effect by encouraging more metal fixation on the fabrics (Dalby, 1993). Although good light and wash fastness ratings have been obtained without using mordants, the introduction of the mordants further broadened the colour gamut and stronger colour hues. This was clearly observed in figure 5showing the colour images for the dyed fabrics.

Premordanting with chrome and copper have brought about an increase in the fastness properties and change in shade of the naturally dyed fabrics. Potassium dichromate and copper (II) sulphate are known for their ability to form a coordinated complex and therefore are easily chelated with the dye.

Figure. 5: Images of some dyed fabrics

The coordination numbers of chromium and copper metals are 6 and 4 respectively. This suggests that some coordination sites are not occupied on interaction with the fibre. Thus, the functional groups such as the amino group in nylon 6 and the carboxylate groups in the polyester fabrics can occupy these vacant sites. The metals can therefore form a tertiary complex at one site with the fibre and on another site with the dye. This is believed to be the reason for the increase in the light fastness performance and colour change in shade of the natural dye extract on nylon 6 and polyester fabrics. The low light fastness of the fabrics dyed with Alum metals could be attributable to the fact that Alum metals usually form weak coordination complexes with the dye. There are slight differences in washing, rubbing and light fastness properties in the majority of cases (Savvidis *et al.,* 2004). However, with respect to washing and rubbing, the fastness properties are slightly lower in the case of post-mordanting by comparison with pre-mordanting with alum in the three fabrics.

The light fastness ratings of the *B. coccineus* dye extract on cotton fabrics were lower than those found for polyester and nylon 6 fabrics mordanted with copper (II) sulphate and potassium dichromate. This may be due to the fact that the cotton fabrics do not contain groups that could possibly interact with the metal mordant to form a strong bond.

CONCLUSION

The natural dye extracted from the bark of *Byroscarpus coccineus* at 100 ^{0}C for 4h with extractions in the presence of 15g1^{-1} of sodium hydroxide solution is described. The dye extract was applied to polyester, nylon 6 and cotton

fabrics using various methods. The results obtained in this study suggest that the natural dye extracted from bark of *B. coccineus* possesses intrinsic affinity for both natural and synthetic fibres even without mordants. The observed potential affinity of the dye extract for the textile substrates used for the study may be due to the presence of tannins, a biomordant. The results of the study show that the dye extracts imparted bright pink colour on the textile substrates. However, the addition of mordants improved the fastness performance of the dye extract on the fabrics. Further studies are required to isolate identify and characterize the active colouring components and determine their chemical structures.

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