



EXTRACTION AND APPLICATION OF DYES DERIVED FROM *Gmelina arborea* LEAVES AND THEIR DYEING PROPERTIES ON COTTON FABRIC

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ABSTRACT

Extraction and application of natural dyes for dyeing polyamide fibre was studied using *Gmelina arborea* leaves. The amount of natural dyes that was exhausted and fixed on the fibre was found to depend on the application conditions. It was observed from the experiment that the longer the dyeing time the higher the dye exhaustion up to 90 minutes, after which a decrease in colour strength followed. The dye absorption by cotton fibre decreases with the increase of dye concentration in the dye bath. The colour strength was increased by increasing electrolyte concentrations which was best at 8 % with the exhaustions of 72 %, 74 % and 68 % for water, ethanol and methanol extracts respectively. The rate and degree of dye fixation on the nylon increased significantly by raising dyeing temperature up to boiling but the dye obtained for all the extract was not uniform. The best results in terms of uniformity were obtained at 60 °C for ethanol extracts and 70 °C for water and methanol extracts respectively. The influence of dyeing methods with mordants was determined. The light and wash fastness of dye-treated samples were measured and compared with untreated samples. The mordanted samples have excellent fastness properties to washing all within the rating of 5 and 4-5 for colour change and staining respectively. It can be concluded that *Gmelina arborea* leaves have good potentiality for dyeing of polyamide fibres.

Keywords: Cotton, fastness properties, *Gmelina arborea*, leaves, mordant, natural dye

INTRODUCTION

A renewed international interest has arisen in natural dyes due to increased awareness of environmental and health hazards associated with the synthesis, processing and use of synthetic dyes in a new emerging technology called 'green technology' [1]. Natural dyes comprise of colourants that are obtained from animal or vegetable matter including roots, bark, leaves, flowers, and fruit without any chemical processing and have a wide range of shades [2]. During the last decade, the use of natural dyes has gained momentum due to increased demand by the food, pharmaceutical, cosmetic as well as

the textile colouration industries [3]. Moreover, in many cases, the fastness properties obtained by natural dyes are inferior to those obtained by synthetic dyes [4]. Since the advent of widely available and cheaper synthetic dyes in 1856 with moderate to excellent colour fastness properties, the use of natural dyes with poor to moderate wash and light fastness has declined to a great extent [5]. However, recently there has been revival of the growing interest on the application of natural dyes on natural fibres due to worldwide environmental consciousness [6]. The widely and commonly used synthetic dyes impart strong colours but causes carcinogenicity and inhibition of benthic photosynthesis [7].

Populace in most developing countries preferred natural dyes than those in the developed countries, because they are non-allergic, non-carcinogenic and have lower toxicity and better bio degradability than the synthetic dyes [8]. Natural dyes can offer not only rich and varied source of dye stuff, but also the possibility of an income through sustainable harvest and sale of these plants [9].

G. arborea Roxb called 'gmelina or white teak' in English, 'gumbar' in Bengali, 'gumhar' in India and 'melaina' among the Hausa people of Nigeria, belongs to the family *Lamiaceae*. It is a large deciduous tree that attains moderates to large height up to 40m with trunk straight to 4.0 m girth [10]. It grows on different localities and prefers moist fertile valleys with 750 – 4500 mm rainfall and does not flourish on ill – drained soil and remains stunted on dry sandy soil. Drought also reduces it to shrub form [11, 12]. *G. arborea* is a native of Philipines and Malasiya and distributed in India, Burma, Myammar, Thailand, Laos, Cambodia, Vietnam, and Ceylon, southern provinces of China were it is found at altitudes from sea level to 1,500 metres. It was also introduced to Brazil, Honduras, Gambia, Malawi, Ivory Coast, Sierra Leone and Nigeria as a valuable timber [13-16]. This tree is commonly planted as a garden and an avenue tree; growing in villages along agricultural land and on village community lands and wastelands [17].

G. arborea wood is pale yellow to cream colour when fresh, turning yellowish brown on exposures to light. It is lustrous when fresh, usually straight to irregular or rarely wavy grained and medium course textured [18]. Flowering takes place between February to April when the tree is more or less leafless whereas fruiting starts from May onwards up to June. The fruit is about 2.5 cm long, smooth, dark green, turning yellow when ripe and has a fruity smell [19]. Different parts of *G. arborea* tree have been used for different purposes. The fruits and leaves are used as feed stuff in India, while the leaf, flower and root have been reported to contain several chemical constituents having medicinal value [20]. *G. arborea* is also used as a shade-trees. The timber products are useful for making building materials, pulp and paper, dyes, stains, inks, tattoos and mordants, carpentry and related applications, farming, hunting and fishing apparatus, fuel and lighting, farming, forestry, household, ship building, plywood, domestic and personal items [21]. The timber products is also used in matchwood industry.

It is also used for making handles of tennis rackets, frames and reinforcements of carom boards and packing cases and crates [17, 22]. *G. arboea* wood is used as raw material for cellulose [23], firewood [24], polewood [25], particle board [26], veneer and some other structural uses [27].

The objective of this study was to extract natural dyes from the leaves of *G. arborea* and the extracted dye will be applied on cotton fabric and the fastness properties of the dyed material with and without mordant will be analysed.

MATERIALS AND METHODS

Sample collection and preparation

Gmelina arborea leaves were gathered from the wild in Lafia Local Government Area of Nasarawa State, Nigeria and were authenticated at the Department of Forestry and Wildlife, Nasarawa State University, Keffi, Nigeria, with a Voucher No FWF/O117 which was deposited at the Departmental Herbarium. The samples were washed thoroughly with water and dried at room temperature. The air dried sample was ground into fine particles by the use of mortar and pestle and transferred into airtight containers with proper labelling and kept away from moisture for further analyses.

Extraction of sample

The dried powdered leaves sample of *Gmelina arborea* were extracted using cold extraction method according to the method described by Usman et al., [28] with slight modification. Briefly, 100 g sample was suspended in 500 ml of each extracting solvents (water, ethanol and methanol) for 48 hours. The mixtures were stirred using glass rod after every 6 hours. Finally, the dye extracts were decanted and filtered with Whatman No. 1 filter paper three times to ensure clear dye solution. Each batch of the filtrates was re-concentrated using rotary evaporator at 40 °C, weighed and store in the refrigerator at 4 °C.

Dyeing procedures

Dyeing of the unmordanted cotton fabric samples was done as per standard parameters recommended for all fabric, reported in Clariant manual [29]. The pH of dye bath was maintained at 3 by adding acetic acid. The material- liquor ratio was maintained at 1:50 during dyeing operation. Dyeing started at 30 °C, the temperature was slowly increased to 90 °C within 10 min and heating was maintained at this temperature for 90 minutes with occasional stirring. During dyeing and at exactly 45 minutes, boiled distilled water was added to the dye baths in order to maintain the level content. After dyeing, the fabrics were squeezed over dye baths, washed with distilled water (two times), rinsed and then dried at room temperature.

Dyeing of the mordanted cotton fabric samples

The wetted out cotton fabric samples were put into dye baths with material-to-liquor ratio of 1:50. Sodium carbonate (0.5 g) was added followed by 0.5 ml of acetic acid at ten minutes intervals. Finally, 2 g of different metal salts (mordants), which were alum, nickel chloride, ferrous sulphate, stannous chloride, copper sulphate or zinc nitrate were added. Dyeing started at 60 °C for 30 minutes and continued at boiled for 1 hour. After dyeing, the dyed sample was washed, rinsed and dried under the sun [30]. The results are shown in Table 5.

Wash fastness treatment

The printed fabrics (wrappers) were subjected to International Standard Organization Test 4 (ISO 4) wash fastness treatment method as follows:

International Standard Organization Test 4 (ISO 4) Treatment

The ISO 4 treatment involves the preparation of soap and detergent solution using 5 g/L soap/detergent and 2 g/L soda ash (Na_2CO_3) in a liquor ratio of 1:50 for 30 minutes at a temperature of 95 °C.

Assessment of Change in Colour

The specimen to be tested was stitched with an undyed cloth measuring 5 cm x 4 cm as the composite sample. The composite test specimen was agitated in a washing wheel machine containing 5g/ soap solution with other additives under specified condition of liquor ratio, temperature and time. The test was conducted for 45 min at a temperature of 50 °C as specified under the ISO 4. The composite specimen was then removed, rinsed and dried. The change in colour of the tested specimen and the staining of the adjacent undyed cloth were assessed using the 1 – 5 scale for specifying fastness as indicated in Table 5.

Assessment of staining

The staining was assessed using the grey scale for assessing stain (BS 1006:978). This scale consist of nine pairs of pieces of cards numbered 5,4-5,4,3-4,3,2-3,2,1-2,1 where 5 is a pair of white and the other consist of a grey, giving series of contrast increasing in geometric progression. No. 1 showing the highest contrast 36.2 CIELAB units of colour difference

RESULTS AND DISCUSSION

The effect of dyeing parameters, which were dye concentration, electrolyte concentration, dyeing time, dyeing temperature, colour fastness and change in hue of the extracted dye when mordents were added was ascertained. From the Table 1, it can be observed that dye absorption by cotton fibre decreases with the increase of dye concentration in the dye bath. This is due to the presence of more dye ions

that hindered the absorption of dye by the fibre. It is to be mentioned that with the increase of dye concentration, the absolute absorption quantity diminishes [31].

The size of dye particles in solution usually depends on temperature, concentration of electrolyte and concentration of dyes. The size of dye particles always increased with an increase in dye concentration [32]. At equilibrium a more or less pronounced selective absorption of the dye by the fibre is observed up to full exhaustion of the dye bath. Forces of interaction of the dye and the fibrous material induce this selective absorption. The exothermic reactions taking place at dyeing confirm the occurrence of the interaction [33]. From the experiment, it was observed that bright and uniform shades were produced when cotton fibre was dyed with 4 % dye. Above or below these percentages of dye, dull and uneven shades were obtained. The effective concentration of 4 % dye for water, ethanol and methanol extracts correspond to 70, 71 and 74 % dye exhaustion from the dye baths respectively. The behaviour of dye absorption in dyeing of cotton fibres depends upon the characteristics of dye towards the fibres [34].

The effect of electrolyte concentration on colour strength is shown in Table 2. The colour strength is increased by increasing electrolyte concentrations which was best at 8% with the exhaustions of 72 %, 74% and 68% for water, ethanol and methanol extracts respectively. A further increase in electrolyte concentration has a negative effect because of increased dye aggregation [35]. Acetic acid tends to neutralize or reduce the negative electric charge (zeta potential) of cotton fabric, thus facilitating the approach of the dye anions to the fabric within the range of formation of hydrogen and other bonds between the dye molecules and fabric [36]. Electrolytes are used as exhausting agent in case of dyeing cellulosic materials with anionic dyes owing to the similarity of charges on both fibres and dyes. Therefore, as a result of the presence of more than one kind of anions in the same dye bath, a competition for the positively charged sites in the fibre will occurred since one anion is capable of replacing another [37].

Table 1: Effect of dye concentration on dyeing of cotton fabric with *Gmelina arborea* extracted dye

Water extract		Ethanol extract		Methanol extract	
Dye concentration	Dye exhaustion (%)	Dye concentration	Dye exhaustion (%)	Dye concentration	Dye exhaustion (%)
1	85	1	90	1	94
2	80	2	86	2	88
3	76	3	80	3	82
4	70	4	74	4	71
5	64	5	70	5	70
6	63	6	70	6	69

Table 2: Effect of dye electrolyte concentration on dyeing of cotton fabric with *Gmelina arborea* extracted dye

Water extract		Ethanol extract		Methanol extract	
Dye electrolyte	Dye exhaustion (%)	Dye electrolyte	Dye exhaustion (%)	Dye electrolyte	Dye exhaustion (%)
0	52	0	46	0	32
2	56	2	58	2	56
4	68	4	64	4	60
6	70	6	72	6	64
8	72	8	74	8	68
10	72	10	75	10	69

Table 3: Effect of dye time (minutes) on dyeing of cotton fabric with *Gmelina arborea* extracted dye

Water extract		Ethanol extract		Methanol extract	
Dye time	Dye exhaustion (%)	Dye time	Dye exhaustion (%)	Dye time	Dye exhaustion (%)
15	62	15	54	15	72
30	76	30	68	30	86
45	77	45	74	45	88
60	78	60	75	60	89
90	80	90	77	90	90
105	77	105	76	105	87

Table 4: Effect of dye temperature (°C) on dyeing of cotton fabric with *Gmelina arborea* extracted dye

Water extract		Ethanol extract		Methanol extract	
Dyeing temperature	Dye exhaustion (%)	Dyeing temperature	Dye exhaustion (%)	Dyeing temperature	Dye exhaustion (%)
40	38	40	54	40	46
50	47	50	55	50	48
60	55	60	64	60	55
70	63	70	80	70	65
80	83	80	97	80	82
90	83	90	97	90	82
105	83	105	97	105	82

Table 5: Effect of mordant on the fastness properties of the extracted *Gmelina arborea* leaf on cotton fabric

Types of Mordant	Water extract			Ethanol extract			Methanol extract		
	Colour Developed	Change in Colour	Staining	Colour Developed	Change in Colour	Staining	Colour Developed	Change in Colour	Staining
<u>Zn(NO₃)₂</u>	Antique <u>belge</u>	5	5	Taupe	5	5	Beige	5	5
Alum	Yellow	5	4-5	Palm	5	5	Palm	5	5
CuSO ₄	Cork	5	5	Teak	5	5	Brass	4	4
FeSO ₄	Camel	5	5	Straw	5	5	Cocoa	5	4-5
<u>TnCl</u>	Palm	5	5	Palm	4	5	Wheat	5	5
NiCl ₂	Wheat	5	5	Wheat	5	5	Cork	5	5
Control (non <u>mordanted</u> sample), Colour - Heather <u>belge</u> , Change in colour - 3, Staining - 3-4.									

The effect of dyeing time on colour strength is shown in Table 3. The dyeing of cotton fibre samples were carried out at 15, 30, 45, 60, 90 and 105 minutes under fixed dyeing conditions of 60 % extract, at pH=3 and at boiling. It was observed from the experiment that the longer the dyeing time the higher the dye exhaustion, until the dye exhaustion attains equilibrium and there is no significant increase after further increases in dyeing time. The maximum dyeing time for all the extracts was 90 minutes as observed in Table 3. The decrease in colour strength at 120 minutes of dyeing may be attributed to desorption of the dye molecules as a consequence of over dyeing [36].

The effect of temperature on dye exhaustion is shown in Table 4. As evident, the effect of temperature on dye exhaustion stays almost the same between 80-105°C. However, the maximum dye exhaustion for water, ethanol and methanol extracts was obtained at 83°C, 97°C and 82°C, but the dye obtained for all the extract is not uniform. The best results in terms of uniformity were obtained at 70°C for water and methanol extracts while for ethanol is 60 °C. It is obvious from Table 4 that the rate of dyeing increases gradually by raising the temperature. Maximum dye fixation is achieved at boiling and the rate of dyeing depends to great extent on the nature of both dye molecules and the fibre itself [37].

Comparing the fastness properties of the control (unmordanted) with the mordanted samples, the mordanted samples performed better than the unmordanted samples as shown in Table 5. The mordanted samples have excellent fastness properties to washing which were all within the rating of 5 and 4-5 for colour change and staining respectively. This improved fastness properties may be attributed to the complex reaction between the dye and the fibres. However, for the change in the hues as a result of mordant addition, the colour of the extracted dyes changed considerably. From table 5, it is evident that the colour of the control sample was straw coloured but when mordant was added the change gave beautiful and bright colours as seen on the table.

CONCLUSION

The present work shows that, leaves of *Gmelina arborea* can be used as dye for colouring cotton fabrics. The plants are grown throughout Nigeria and it is easily available plant. Different shades of colour can be obtained using different mordants. The washing fastness of all dyeing with mordants were quite good and also dye extract has shown change in hue when the mordants were added. The dye has good scope in the commercial dyeing of cotton.

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