Extraction of Dyestuff from *Justicia carnea hooker* and Its Application in the Dyeing of Wool, Leather and Cotton

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**Abstract:** Dye was extracted from whole plant of *Justicia carnea hooker*, using soxhlet extraction method. The dyestuff extracted gave a percentage yield of 92.8% and was successfully applied on leather, cotton, and wool. The extracted dye showed solubility in distilled water, HCl, H2SO4, ethanol and NaHCO3 respectively. Dye extract also shows a pH 9, TLC revealed blue, green and yellow colours with RF values of 0.83, 0.86 and 0.89, λmax was 480 and gave a green shade. Sulphanilic diazonium chloride was used as the coupler and it produced a brown shade in each of the substrates applied on and fastness test gave good result. pH for the coupled dye was 8.2 while TLC revealed yellow and orange colours with RF values of 0.98 and 0.95 respectively. λmax obtained were 400nm and 480nm. Acid fastness, alkaline fastness, sunlight fastness and wash fastness of coupled dye were determined and compared to a standard Gray Scale rating. Thin layer chromatography carried out showed that dyestuff can be extracted from *Justicia carnea hooker*.

**Keywords:** Dye, Fastness, Extraction, Solubility.

**1. Introduction**

About 1,100 plant species can be used for dyeing [1]. Therefore it is an important aim in research to screen and select species which fit for modern sustainable cultivation techniques as well as
for dyeing on a large scale. At the Thüringer Landesanstalt für Landwirtschaft (Germany), 108 dye plant species are assessed on the basis of their suitability for modern cultivation systems, on yields, and on dyeing quality. Of these species, 19 species were considered as useful for cultivation and dyestuff production. Madder (Rubia tinctorum), Weld (Reseda luteola), Canadian Golden Rod (Solidago canadensis), Dyer’s Chamomile (Anthemis tinctoria), and Dyer’s Knotweed (Polygonum tinctorium) are considered to play a decisive role in future dye plant cultivation and processing [2]. Nevertheless, other species may also become important for dyeing, as the recent example of Rhubarb (Rheum rhabarbarum) shows: it is perfectly suitable for the dyeing and tanning of leather and is already used in practice by one company [3].

A dye is a colored substance that has an affinity for the substrate to which it is being applied. It is generally applied in an aqueous solution, and requires a mordant to improve the fastness of dye on the fibre [4]. Both dyes and pigments appear to be colored because they absorb some wavelengths of light more than others. In contrast with a dye, a pigment generally is insoluble, and has no affinity for the substrate. Some dyes can be precipitated with an inert salt to produce a lake pigment, and based on the salt used; they could be Aluminum Lake, Calcium Lake, or barium lake pigments [5]. Research has shown that the natural dyes are quite safe and environment friendly [6].

The use of plants for dyeing is environmental friendly. Synthetic dyes have been widely used all around the world, and the demand for natural dye product is on the increase. Hence natural dye will succeed once more. The aim of this work is to extract the dye and apply it on cotton, leather and wool

2. Methodology

2.1. Sample Collection

The stem of plant justicia carnea hooker was brought in from Bamenda (Cameroun) in 2013 and planted at Graceland, Zaria, Kaduna state, in Nigeria, from where the plant was collected and prepared for the analysis.

2.2. Sample Preparation

The sample was collected and dried at room temperature (room drying) after which it was grinded into fine particles using mortar and pistil, and then sieved using 0.01 mm sieve.

2.3. Extraction of the Dye of Justicia carnea hooker Using Soxhlet Method

The extraction of dye from justicia carnea hooker was carried out using soxhlet extraction method. The sample was weighed (50g) and transferred into a timbling chamber of the apparatus, using methanol and water as solvents. The extraction was carried out based on solid-liquid extraction,
a type of continuous extraction. The sample was placed in the porous thimble and inserted into the inner tube. The extractor was then fitted to the bolt head flask containing the solvent and the reflux condenser. The solvent boiled gently as the vapour passed through the tube while it condenses and the condensed solvent falls into the thimble and slowly filled the body of the soxhlet. When the solvent reaches the siphoning point, it siphoned over the flask and thus removes that portion of substances which it has extracted. The process repeated automatically until complete extraction was achieved. The extracted compound was then isolated from it solution by using a calibrated evaporating dish.

2.4. Preparation of Diazonium Salt

2.5g of sulphanilic acid powder was weighed and dissolved in 50ml of distilled water then 0.75g sodium carbonate was weighed and added gradually into the solution and stirred while heating at a very low temperature for 10 minutes then it was labeled sample A then 1.8g sodium nitrite (NaNO\textsubscript{2}) was dissolved in 10ml distilled water and was labeled sample B. Then sample A was placed in an ice water bath and sample B was added gradually to the cold solution of sample A and was stirred constantly in a fume cupboard until sample B was completely added into sample A and the mixture was allowed to stand for 30 minutes in an ice water bath then 10ml of concentrated hydrochloric acid was measured and diluted into 10ml distilled water and the solution was gradually added to sample A, and a starch iodide paper was used to confirm the end point of the reaction.

2.5. Coupling Reaction of the Dye

1g of the dye extract was weighed and dissolved in 10ml distilled water in a beaker then 20% sodium hydroxide(20ml) was measured and added to the solution and cooled in ice water bath for 30 minutes. The sulphanilic diazonium chloride was gradually added to the solution of the extract and stirred continuously in a fume cupboard until it was completely added and the mixture was filtered into a flat bottom flask using a filter paper and the filtrate was poured into an evaporating dish and concentrated by placing in an oven for 24 hours at 60\textdegree c.

2.6. Characterization

The dye extract was analyzed for its solubility, functional group present and the absorbance.

2.7. Solubility Test

The coupled dye was dissolved in different media respectively, to test for its solubility. The coupled dye (0.2g) was weighed and placed in eight (8) different test tubes labeled A, B, C, D, E, F, G, & H, respectively, and 4ml each of the following solvents; distilled water (H\textsubscript{2}O), ethyl ether, concentrated & dilute hydrochloric acid (HCl), concentrated and dilute sulphuric acid(H\textsubscript{2}SO\textsubscript{4}), ethanol
(C₂H₅OH), 10% sodium bicarbonate (NaHCO₃) were added respectively and shake for 2 minutes and warmed for 1 minute in hot water bath at 60°C.

2.8. pH Determination of the Dye

The dye extract (1g) was dissolved in distilled water (5ml) and pH meter was calibrated using distilled water and inserted into the dye extract solution. Also, the coupled dye (1g) was dissolved in distilled water (5ml) and pH meter was calibrated using distilled water and inserted into the coupled dye solution.

2.9. Thin Layer Chromatography of the Dye Extract

The dye extract(1g) was dissolved in methanol (2ml) in a test tube and capillary tube was used to stain the dye extract solution on chromatography plate (2cm x 10cm) and allowed to dry and was placed in a mixture of methanol (3.5ml) & n-hexane (1.5ml) in chromatography tank.

3. Ultraviolet Spectroscopic Analysis

The ultraviolet spectroscopic analysis of the dye was carried out using a JENWAY 6305 spectrometer from the Nigerian Institute of Leather and Science Technology (NILEST), Zaria, to obtain the wavelength of maximum absorption. The Spectrum of the dye is shown in Figure 7.

3.1. Dyeing of Chrome Tanned Leather with the Coupled Dye Extract

The chrome tanned leather (5g) was weighed and the coupled dye (0.25g) was weighed and distilled water (4ml) was measured and boiled to 60°C and the chrome tanned leather was soaked in cold water for 30 minutes then the dye was dissolved in warm water at 60°C. The wet chrome tanned leather was placed in the dye solution in a conical flask and was agitated using a shaker for 30 minutes then 1ml of formic acid was added into the mixture and agitated using a shaker for another 30 minutes and the dye solution was decanted and the dyed leather was rinsed using cold water until there was no visible colour change on addition of water and the dyed leather was placed on a white tile and allowed to air dry at room temperature and the dyed leather was fastened for 24 hours on toggle frame using toggle clips.

3.1.1. Dyeing of Cotton with the Coupled Dyed

The cotton fibre (2g) was soaked in warm water (60°C) for 2 hours then 1ml of formic acid was added into a beaker containing 10ml of water (60°C) and the dye (2g) was dissolved in the solution and the wet cotton was added into the solution and was stirred regularly and kept on simmer for 1 hour and the cotton was removed and allowed to dry at room temperature.
3.2. Assessment of Dyeing Property

3.2.1. Fastness of light (sunlight)

This test was carried out according to the official method of analysis by the society of Leather Technologists and Chemists; A piece of dyed leather of size 3 x 3 cm was half covered with a black polythene sheet such half of the length, 1.5 x 1.5 cm was half covered and the remaining half was left uncovered exposed to daylight for 5 consecutive days between the hours of 9:00am – 3:00pm (6 hours); after which the change in color was compared to the uncovered and unexposed leather respectively; using the standard grey scale.

3.2.2. Alkaline fastness

The leather 3cm X 3cm was dipped in sodium carbonate solution for 30 minutes, and dried at room temperature without rinsing. The change in color was determined using the standard grey scale.

3.2.3. Acid fastness

A piece of dyed leather of size 3cm x 3cm, was immersed in 0.1M or 56% glacial acetic acid solution for 30 minutes, and then allowed to dry at room temperature without rinsing. The color change was assessed using the standard grey scale.

3.2.4. Wash fastness

The dyed leather 3cm x 3cm was dipped into a solution made by adding 5g (0.5%) of detergent powder into 200cm³ of deionized water. This was placed in a water bath, and the temperature allowed to rise to 40°C and was vigorously stirred for 30 minutes at 5 minutes interval. After 30 minutes, the leather was placed in 100cm³ of distilled water at room temperature and allowed to stand undisturbed for 1 minute and it was removed and gently squeezed by the hand. This washing and rinsing was carried out 5 times and the leather was dried in the oven at 60°C. The change in the relevant surface of the dyed leather was assessed by comparing it with unwashed leather, using the standard grey scale.

4. Results and Discussion

As presented in Table 1, it is possible to achieve a very soluble solution in both alkaline and acidic medium except in ethyl ether which was partially soluble. The pH determination of the dye extract and coupled dye is presented in table 2. The results prove that the dye is basic. Physical test of the dye on acid, alkaline, light (sunlight) and wash fastness of the dyed cotton and leather in tables 4,5,6,7 respectively, showed good fastness property.

The percentage yield of the dye was calculated as follows:
Percentage yield = \frac{\text{weight of sample} - \text{weight of extract}}{\text{weight of sample}} \\
= \frac{50 - 36 \times 100\%}{50} \\
= \frac{46.4 \times 100\%}{50} \\
= 0.928 \times 100\% \\
= 92.89\%

Table 1: The solubility test results

<table>
<thead>
<tr>
<th>EXPERIMENT</th>
<th>OBSERVATION</th>
<th>INFERENCE</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>Dye + distilled H$_2$O</td>
<td>Soluble with a light brown colour</td>
</tr>
<tr>
<td>B</td>
<td>Dye + ethyl ether</td>
<td>Two layer solution is formed</td>
</tr>
<tr>
<td>C</td>
<td>Dye + Conc. HCl</td>
<td>Soluble with dark orange colour</td>
</tr>
<tr>
<td>D</td>
<td>Dye + 1M HCl</td>
<td>Dark brown solution is formed with effervescence</td>
</tr>
<tr>
<td>E</td>
<td>Dye + Conc. H$_2$SO$_4$</td>
<td>Dark brown solution is formed with effervescence</td>
</tr>
<tr>
<td>F</td>
<td>Dye + 2M H$_2$SO$_4$</td>
<td>Brown solution is formed with no precipitate</td>
</tr>
<tr>
<td>G</td>
<td>Dye + ethanol</td>
<td>Light orange solution was formed with precipitate</td>
</tr>
<tr>
<td>H</td>
<td>Dye + 10% NaHCO$_3$</td>
<td>Light brown solution is formed</td>
</tr>
</tbody>
</table>

Table 2. Characterization of the extracted dye

<table>
<thead>
<tr>
<th>SAMPLE</th>
<th>pH</th>
<th>TLC</th>
<th>RF VALUE</th>
<th>$\lambda_{\text{max}}$(nm)</th>
<th>COLOUR</th>
</tr>
</thead>
<tbody>
<tr>
<td>DYE EXTRACT</td>
<td>9.0</td>
<td>Blue, Green</td>
<td>0.83, 0.86, 0.89</td>
<td>480</td>
<td>Green</td>
</tr>
<tr>
<td>COUPLED DYE</td>
<td>8.2</td>
<td>Yellow, Orange</td>
<td>0.98, 0.95</td>
<td>400 &amp; 480</td>
<td>Brown</td>
</tr>
</tbody>
</table>

Table 3. The result of dyeing from coupled dye

<table>
<thead>
<tr>
<th>EXPERIMENT</th>
<th>RESULT</th>
<th>AMOUNT OF DYE USED</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dye with chrome tanned leather</td>
<td>Brown shade was obtained with partial fixation</td>
<td>5% of the dye</td>
</tr>
<tr>
<td>Dye with cotton</td>
<td>Pale brown shade was obtained with partial fixation</td>
<td>2% of the dye</td>
</tr>
</tbody>
</table>
Table 4. The result of acid fastness with coupled dye

<table>
<thead>
<tr>
<th>DYED SAMPLE</th>
<th>FASTNESS RATING</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chrome tanned leather</td>
<td>4</td>
</tr>
<tr>
<td>Cotton</td>
<td>3</td>
</tr>
</tbody>
</table>

Table 5. The result of alkaline fastness with coupled dye

<table>
<thead>
<tr>
<th>DYED SAMPLE</th>
<th>FASTNESS RATING</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chrome tanned leather</td>
<td>4</td>
</tr>
<tr>
<td>Cotton</td>
<td>4</td>
</tr>
</tbody>
</table>

Table 6. The result of sunlight fastness with coupled dye

<table>
<thead>
<tr>
<th>DYED SAMPLE</th>
<th>FASTNESS RATING</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chrome tanned leather</td>
<td>3</td>
</tr>
<tr>
<td>Cotton</td>
<td>3</td>
</tr>
</tbody>
</table>

Table 7. The result of wash fastness with coupled dye

<table>
<thead>
<tr>
<th>DYED SAMPLE</th>
<th>FASTNESS RATING</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chrome tanned leather</td>
<td>3</td>
</tr>
<tr>
<td>Cotton</td>
<td>2</td>
</tr>
</tbody>
</table>

Table 8. Key (interpretation of results fastness colour change)

<table>
<thead>
<tr>
<th>GRAY SCALE RATING</th>
<th>ASSESSMENT</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>Excellent</td>
</tr>
<tr>
<td>4</td>
<td>Very good</td>
</tr>
<tr>
<td>3</td>
<td>Good</td>
</tr>
<tr>
<td>2</td>
<td>Poor</td>
</tr>
<tr>
<td>1</td>
<td>Very poor</td>
</tr>
</tbody>
</table>

THIN LAYER CHROMATOGRAPHY OF DYE EXTRACT

Identification of component in chromatogram

\[ R_f = \frac{\text{distance moved by the compound from origin}}{\text{solvent movement from the origin}} \]

Dye Extract

For Blue

\[ RF = \frac{5.8}{7.0} = 0.83 \]
For Green
RF=$\frac{6.0}{7.0}$ = 0.86

For Yellow
RF=$\frac{6.2}{7.0}$ = 0.89

Coupled Dye
For Yellow
RF=$\frac{5.4}{5.5}$ = 0.98

For Orange
RF=$\frac{5.2}{5.5}$ = 0.95

**Fig 1.** Chrome tanned leather before dyeing

**Fig 2.** Chrome tanned leather after dyeing

**Fig 3.** Toggling of chrome tanned leather after dyeing

**Fig 4.** Cotton before dyeing

**Fig 5.** Cotton after dyeing

**Fig 6.** Thin layer chromatography of dye
Fig. 7. UV/Vis spectroscopy of coupled dye

5. Conclusion

The result obtained indicates that the dye extracted gave a good yield and was successfully applied on leather, cotton, and wool. However, there was decrease in glossy appearance on the dyed cotton and no colour change was observed with fastness property. The TLC showed that the dye extract contained three components (blue, green and yellow) which prove that Justicia carnea hooker contains dyestuff. Also, the pH determination shows that the dye is basic.

References


