Extraction of Natural Dyes from Forest Trees and their Application in Textiles

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Abstract—The environment - friendly natural dyes are recently enjoying resurgence in popularity because of the concern with the carcinogenic, mutagenic and sensitizing characteristics of many synthetic dyes. Showing concern towards the nature and to avoid pollution of our environment two forest plants Bixa orellana and Caesalpinia sappan were selected as natural dye sources. In this study, from the bark of C. sappan, seeds of B. orellana and combination of B. orellana and C. sappan, the dyes were extracted. The phytochemical compounds present in the extracts were analyzed as carotenoids in B. orellana and anthocyanin, phenolic acids, flavonoids in C. sappan using TLC. The extracts were then applied to cotton fabric using various natural and chemical mordants which were then tested for their color fastness against rubbing and washing to know the efficiency of the dye. This study will help in improving the natural dye resources and replace them with synthetic dyes, consequently leading to a safe environment

Keywords: Bixa orellana, Caesalpinia sappan, Colour fastness, extraction of natural dyes.

I. INTRODUCTION

Indians are considered as forerunners in the art of natural dyeing. The environment - friendly natural dyes are recently enjoying resurgence in popularity because of the concern with the carcinogenic, mutagenic and sensitizing characteristics of many synthetic dyes. The use of non-toxic and eco-friendly natural dyes on textiles has become a matter of significant importance because of the increased environmental awareness in order to avoid hazardous synthetic dyes. Natural dyes find use in the coloring of textiles, drugs, cosmetics, etc. Owing to their non- toxic effects, they are also used for coloring various food products. In addition to their dye-yielding characteristics, some of the plants which are sources of natural dyes also possess medicinal value [1]. Natural dyes produce very uncommon, soothing and soft shades as compared to synthetic dyes, and can be obtained from various parts of plants including roots, bark, leaves, flowers, seeds etc [2]. Although indigenous knowledge system has been practiced over the years in the past, the use of natural dyes has been diminishing over generations due to excessive demand, lack of documentation, precise technical knowledge on the extraction and dyeing techniques. Due to these reasons, it has not commercially succeeded like the synthetic dyes [1].

Almost all the synthetic colorants are being synthesized from petrochemical sources through hazardous chemical processes posses threat towards its eco-friendliness. Hence, worldwide, growing consciousness about organic value of eco-friendly products has generated renewed interest of consumers towards use of textiles (preferably natural fiber product) dyed with eco-friendly natural dyes [3].

Hence, showing our concern towards the nature and to avoid pollution of our environment we have selected this project. We have selected two forest plants Bixa orellana and Caesalpinia sappan as natural dye sources. Bixa orellana is the main source of the natural pigment annatto or bixin, an apo-carotenoid accumulated in large quantity in its seeds. The yellow to orange color is produced by the chemical compounds bixin (Figure1) and norbixin [4]. Another plant source, Caesalpinia sappan is whitish when freshly cut and turns red on exposure to air. The woody part contains brazilin (Figure2) and brasilein [5].

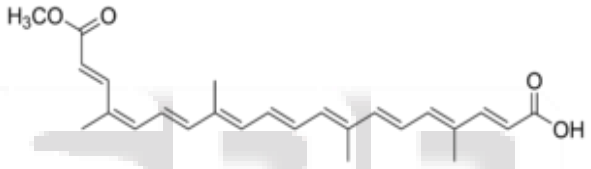

Fig. 1 : Chemical structure of bixin

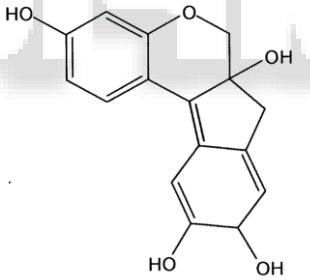

Fig. 2 : Chemical structure of brazilin

II. MATERIALS AND METHODS

A. *Collection of Plant Materials*

Bixa orellana seeds (Figure3) and Caesalpinia sappan bark (Figure4) were collected from Farming Trust of India, Kanjikodu, and Kerala.

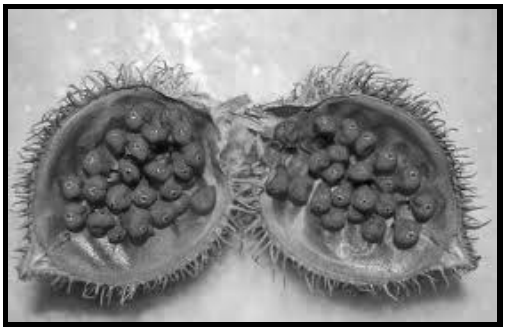

Fig. 3 : Seeds of Bixaorellana

Fig. 4 : Bark of Caesalpinia sappan

B. Selection of the best Solvent for extraction of dyes

The plant materials were ground using a mixer. The powdered materials were extracted using different solvents. The solvents include water, HCl, NaOH, acetone and ethanol. 1 g of each powder was extracted in 15 ml of above mentioned solvents in orbital shaker for 1 hr at 50 rpm speed. At the end of extraction, the extract was filtered using a Whatmann filter paper. The optical density of these extracts was taken using a UV-Visible spectrophotometer, wavelength ranging 200-495 nm [6]. Acetone and ethanol extracts showed higher values. Then, the samples were extracted using ethanol and acetone in a Soxhlet apparatus [7]. The fabric was desized to remove starch if any. For desizing, the cloth was allowed to boil in soap water for 30 min. Then, it was washed in warm water and allowed to air dry. The dye extracted using acetone did not fix properly in cotton fabric leading to a conclusion that ethanol works better.

C. Extraction of Dyes and Mordants

Fine powder of Bixa orellana seeds (24.26g) and Caesalpinia sappan bark (16.26g) were placed in thimbles. Then, extraction was carried out using ethanol as solvent in Soxhlet apparatus at 78°C for 3 days and the extracts were concentrated using a flash evaporator. The concentrated dye was stored at 4°C. Most natural dyes need mordant to fix in the fabric and to increase the fastness properties [1] (washing and rubbing fastness). Different natural mordants like pomegranate rind, orange peel and grape peel extracts were tried. The mordants were also extracted using Soxhlet extractor using ethanol as a solvent and then condensed using the flash evaporator.

D. Dying of fabric and selection of suitable mordants and mordanting methods

After extraction, the dyes were applied to the fabric (Figure 13) by three different mordanting techniques namely, pre-mordanting, simultaneous- mordanting and post-mordanting using specific quantity of mordants. A laboratory Winch machine was used for dyeing [8]. Pre-mordanting involved, initially the desized fabric's treatment with mordant, then with dye at optimal temperature and time. Simultaneous-mordanting included treating the desized fabric simultaneously with mordant and dye at the optimal time and temperature. Post-mordanting involved dyeing process initially and the addition of mordant at a later stage [9].

The dye extracted in ethanol was applied to the fabric using a variety of natural (orange peel extract, pomegranate rind extract, grape peel extract) and chemical mordants (ferrous sulphate, chrome, alum) and mordanting techniques. Through the subjective and objective analysis, the best mordant and mordanting technique was chosen as shown in Table3.

Material liquor ratio (MLR) 1:30

Without mordant:
Weight of the fabric * 10% dye

Pre mordanting:
Weight of the fabric*5% mordant
Weight of the fabric*10%dye

Simultaneous mordanting:
Weight of the fabric*5% mordant*10%dye

Post mordanting:
Weight of the fabric*10%dye
Weight of the fabric*5% mordant

Table 1 show the percentage dye absorption of the dye to the fabric [10] which was calculated using the formula:

$$\frac{\text{Optical density before dyeing} - \text{Optical density after dyeing}}{\text{Optical density Before dyeing}} \times 100$$

E. Colour Fastness

Colour fastness is a measure of the degree to which the fabric changes its colour when subjected to a particular treatment or environment. This property was assessed using grey scale. As per AATCC test method 8 and AATCC test method 61- standard procedures for rubbing and washing fastness tests were carried out using crock meter [11] and launder-o-meter [12] respectively.

F. Thin Layer Chromatography

Thin-layer chromatography (TLC) is a method for separating mixtures of compounds based on the polarity. The separation depends on the relative affinity of com o nds towards stationary and mobile hase he il ica el stationary hase was niformly coated onto the lass lates and the lates were dried in hot air o en at for hour. TLC chamber (development tank) was used for the development of TLC plate. The chamber maintains a uniform environment inside for proper separation of the solutes and development of spots. The mobile phase (solvent) was filled in the TLC chamber and saturated for 2 hrs by dipping a filter paper. This helps uniform rise in mobile phase over the length of stationary phase (silica gel plate). The mobile phase consists of a solvent or solvent mixture which are chemically inert to sample and stationary phase. Different mobile phases were used for the separation of Bixa orellana [13] (varying ratios) and Caesalpinia sappan [14] samples as given below:

Bixa orellana
Acetone: Propanol (5:5, 6:4, 7:3, 4:6)
Acetone: Ethanol (5:5, 6:4, 7:3, 4:6)
Acetone: Glacial acetic acid (5:5, 6:4, 7:3, 4:6)

Caesalpinia sappan
Chloroform: methanol (9.4:0.6, 4:6)
Hexane: ethylacetate (9.4:0.6, 4:6)

After saturation time, the silica plates with the samples spotted area at the lower end were placed in

separate development tanks. The setup is allowed for sufficient time to develop spots, and then the plates are removed and air-dried. The separated spots are visualized under UV light in order to detect the compounds resolved.

For *Bixa orellana* the solvent /mobile phase acetone: ethanol (7:3) and for *Caesalpinia sappan* hexane: ethyl acetate (4:6) were efficient to resolve the compounds.

III. RESULTS AND DISCUSSION

A. Extraction using Different Solvents

The grounded plant Bixa orellana and Caesalpinia sappan samples were extracted using solvent acetone, ethanol, sodium hydroxide, hydrochloric acid and water. The absorbance was noted to select the best choice of solvent for extraction.

Inference

Though acetone and ethanol showed higher value, the dye extracted from acetone does not subsequently fix to the fabric properly. Hence, ethanol was selected as the solvent for extraction of the natural dyes from the plant materials.

B. Absorbance of the Dye Extracted from seeds of Bixa orellana and bark of Caesalpinia sappan

The dyes extracted separately from the seeds of Bixa orellana and the bark of Caesalpinia sappan and a combination of both were examined using spectrophotometer, from 190-600nm and the maximum absorbance of the individual extract was determined. The maximum absorbance of dye extracted from Bixa orellana seeds was at 487 nm as in Figure5. The maximum absorbance of dye extracted from bark of Caesalpinia sappan was at 298 nm as in Figure6. The maximum absorbance of combination (1:1) of these two dye extracts was at 260 nm as in Figure7.

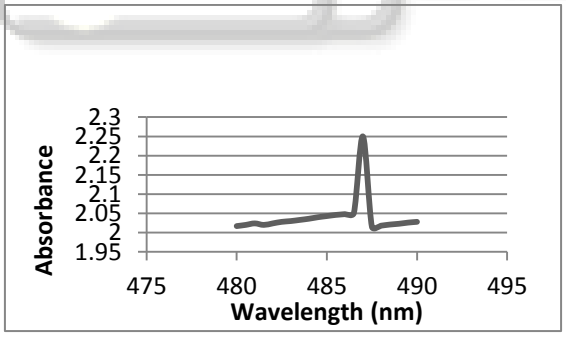

Fig.5 : Absorption maximum of dye extracted from the seeds of Bixa orellana

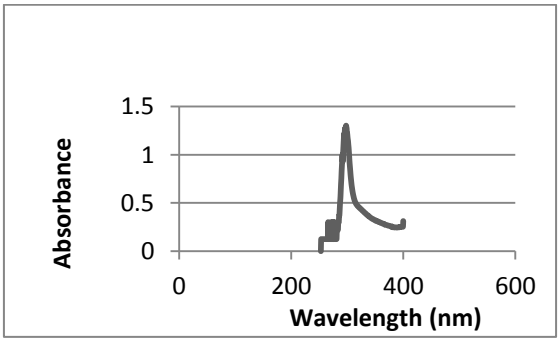

Fig.6: Absorption maximum of dye extracted from the bark of Caesalpinia sappan

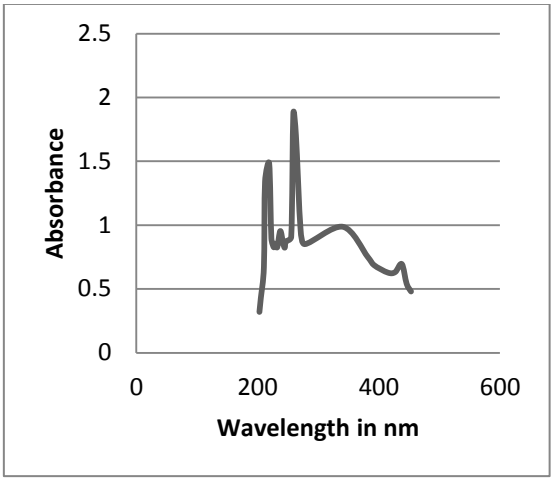

Fig.7 : Absorption maximum of Combination (1:1) of natural dyes from B. orellana and C. Sappan

C. Thin layer chromatography Separation

Upon the separation by TLC, Figure11 represents the extract of *B. orellana* that showed bright orange color indicating the presence of carotenoids. Similarly, Figure10 represents the extract of *C. sappan* that showed pink, brown and yellow color indicating the presence of anthocyanin, phenolic acids and flavonoids, respectively.

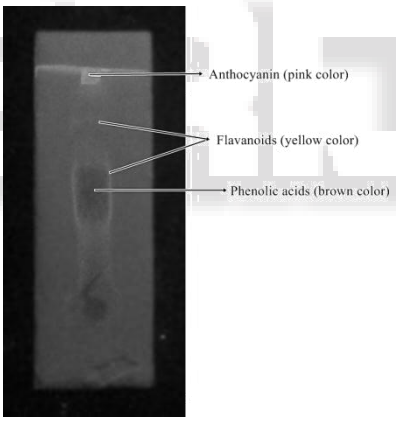

Fig.10: Thin layer chromatogram of extract from C.sappan bark

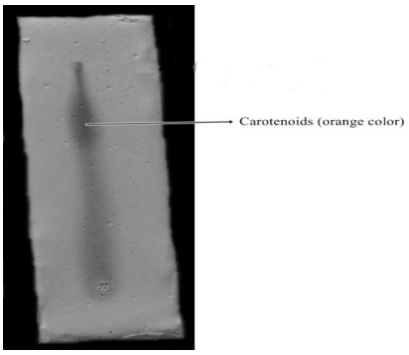

Fig. 11: TLC separation of extract from B.orellana seeds

Sample	OD of Before dyeing	OD of After dyeing	%dye absorption
Bixa orellana (w/o)	0.143	0.105	26.57
Bixa orellana (Nm)	0.492	0.176	64.22
Bixa orellana (Cm)	0.439	0.175	60.13

Caesalpinia sappan (w/o)	1.257	0.725	42.32
Caesalpinia sappan(Nm)	1.695	0.345	79.64
Caesalpinia sappan(Cm)	1.016	0.235	76.87
Combination (w/o)	0.519	0.079	84.77
Combination (Nm)	2.915	2.050	29.67
Combination (Cm)	0.613	0.25	59.21

Table.1: Percentage Absorption of the Dye applied on the cotton fabric

Sample	Washing	Rubbing
Bixa orellana(w/o)	3	3
Bixa Orellana (nm)	4/5	3
Bixa Orellana (cm)	3	2/3
Caesalpinia sappan (w/o)	1	4/5
Caesalpinia sappan (nm)	2	3/4
Caesalpinia sappan (cm)	1/2	2
Combination(w/o)	3/4	3
Combination(nm)	3/4	3
Combination(cm)	4	2/3

Table. 2: Change in Color Grade for Washing and Rubbing Fastness

Abbreviations:
Nm - Natural mordant
Cm - Chemical mordant
W/O - Without mordant
Grade scores:
1-Poor, 2-Fair, 3-Good, 4-Very good, 5-Excellent

D. Color fastness

Table 2 shows that, after performing the color fastness tests of the dyed fabrics, the *C.sappan* dyed fabric without the use of any mordants showed very good rubbing fastness were as the fabric dyed with extract from *B.orellana* dye using grape peel extracts as natural mordant showed very good to excellent washing fastness.

Source	Type of mordanting	Natural mordant	Type of mordanting	Chemical mordant
Bixa orellana	Pre-mordanting	Grape peel extract	Post-mordanting	Alum
Caesalpinia sappan	Pre-mordanting	Grape peel extract	Post-mordanting	Alum
Combination	Simultaneous mordanting	Pomegranate rind extract	Post-mordanting	Alum

Table.3: Choosing of best mordant and mordanting technique

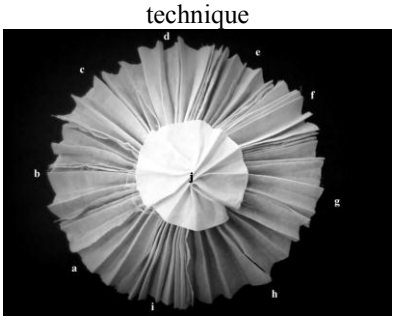

Fig.13: Naturally dyed cotton fabrics

a - *Bixa orellana* (without mordant)
b - *Bixa orellana* (natural mordant)
c - *Bixa orellana* (chemical mordant)
d - *Caesalpinia sappan* (without mordant)
e - *Caesalpinia sappan* (natural mordant)
f - *Caesalpinia sappan* (chemical mordant)
g - Combination of dyes (without mordant)
h - Combination of dyes (natural mordant)
i - Combination of dyes (chemical mordant)
j - White cloth (without dye)

ACKNOWLEDGEMENT

The authors sincerely thank the Management, Departments of Biotechnology, Fashion Technology, and TIFAC CORE of Kumaraguru College of Technology, Coimbatore, India for the support and infra-structure rendered for the successful conduct of the research. The authors also thank Mr.R.Manikam**,** Research Scholar, Karpagam College, for his unsolicited help and encouragement.

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