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Aqueous and Solvent Extraction of Natural Colorants from Tagetes Erecta L., Lawsonia Inermis, Rosa L for Coloration of Cellulosic Substrates

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Abstract: Natural dyes obtained from renewable resources of nature such as plant and animal, although natural dyes from minerals of the earth are also known. Extraction of natural dyes from plant sources and improvement of physico-chemical properties of corresponding dyed fabrics through introducing different mordant were reported in this research. Three south Asian plants (Tagetes erecta L., Lawsonia inermis, Rosa L.,) were selected as natural sources for the extraction of natural colorants. Aqueous and Solvent extraction method using water, acetone and ethanol was employed for liquid dyes. With the help of mordents extracted colorants has been installed into the cellulosic fabric. The samples were characterized with spectrophotometer. Physical test were performed under corresponding ISO standards. Results shows excellent coloring ability of extracted colorants thus can be a potential substitute of synthetic dye as a sustainable approach to clean production.

Keywords: Natural Dyes, *Tageteserecta L., Lawsoniainermis, Rosa L.* Physico-Chemical Properties

1. Introduction

At present, there is an excessive use of synthetic dyes, estimated at around 10,000,000 tons per annum [1], the production and application of which release vast amounts of waste and unfixed colorants, causing serious health hazards and disturbing the eco-balance of nature. Currently, ecological considerations are becoming important factors in the selection of consumer goods all over the world. Since the mid-1980s, more interest has been shown in the use of natural dyes and a limited number of commercial dyes, and small businesses have started to look at the possibility of using natural dyes for coloration [2]. Today, in the world of growing environmental consciousness, natural colourants have attracted the attention of everyone. Natural dyes used in food are screened for safety but the information is not known for most of the natural dyes used in craft dyeing and with potentially wider use. There is a tendency to assume that consumable natural products are safer and better than synthetic product because they came naturally. The safety of natural dyes needs to be proved if they are used more widely and in commercial process [3]. Indigo, a blue dye was extracted from the leaves of leguminous plant [4] and turmeric (Curcuma longa), a yellow dye, was extracted from the ground root of Indian saffron plants. Turmeric was the only yellow dye that did not require a mordant to fix it on cotton or silk but is sensitive to light, soap and alkali which reduce its value considerably. Logwood [5] is the most important in black and blue dyeing. It was used on cotton, silk and wood with various mordents for a wide range of colours, but its fastness to light was generally rather poor. Henna (Lawsoniainermis) leaves is an ancient dye, evidence being the Egyptian mummies found in the tombs who had their nails dyed with henna. In the present times, it is used in many countries for dyeing hair, eyebrows and fingernails. During religion festivals and marriages etc. the use of henna for dyeing the palms and fingernails is an auspicious ritual in most state. For use a colouring material, henna powder is pasted with water and applied to the part to be dyed. For dyeing hair, it is applied as a pack, it acts as a substantive dye for keratin and imparts an orange red colour. It is harmless and causes no irrigation of skin [5] This plant is used to gardening and dyeing the part of bodies hair and also used in Ayurvedic medicine. Mordant is an anchoring agent that anchor dye components with fibers. In most of the lawsones 2- hydroxy-1-4-naphthoquinone is the prominent colorant which contains a –OH group in the position 2 of ringed structure of naphthoquinone replacing a –H atom with it.

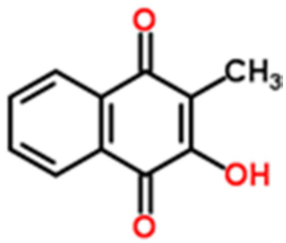

Figure 1. *(2- hydroxy-1-4-naphthoquinone).*

Marigold in its petal contains Xanthophylls, oxygen containing carotenoid, for the expression of its beautiful color that to be extracted. The flower heads exhibit a high yield of Xanthophylls content [6]. The central chain of Xanthophylls consists of 18 carbon atoms bonded alternatively with single and double covalent bonds with four methyl groups attached always in the same position. This single and double bonds act as conjugated system for color construction.

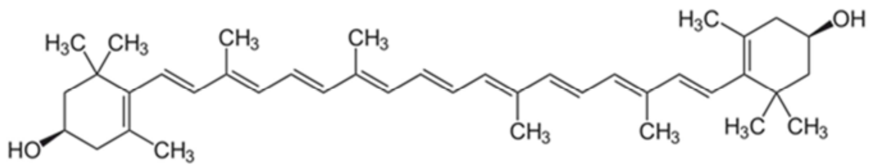

Figure 2. *Xanthophyll. ($C_{40}H_{56}O_2$).*

Rose flowers exhibit colors due to the presence of anthocyanins, the highly colored flavonoids. In fact anthocyanins are the glucosides of anthocyanidins and they may have different sugars bonded to their ringed structure. It has been found pH dependent [7].

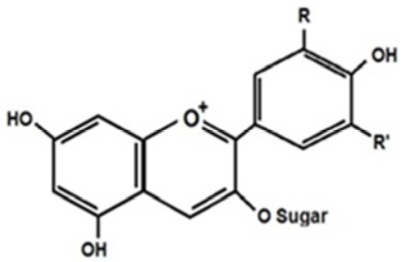

Figure 3. *Anthocyanin.*

Mordants may be the salts of transitional metals (Fe, Cu) alum, chrome etc. [8, 9]or organic acids (oxalic acid, tannic acid etc.) or even natural origins (tamarind seed coat, date palm etc.). The present work was conducted using alum, tannic acid, ferrous sulphate and copper sulphate as mordants.

Chemistry involved in natural dyeing

Most of the natural dyes have no substantivityon cellulose or other textile fibres without the use of a mordant. The majority of natural dyes need a mordanting chemical (preferably metal salt or suitably coordinating complex forming agents) to create an affinity between the fibre and dye or the pigment molecules of natural colourant. These metallic salts as mordantorm metal complexes with the fibres and the dyes. After mordanting, the metal salts anchoring to the fibres, attracts the dye/organic pigment molecules to be anchored to the fibres and finally creates the bridging link between the dye molecules and the fibre by forming coordinating complexes. Aluminiumsulphate or other metallic mordants anchored to any fibre, chemically combine with certain mordantable functional groups present in the natural dyes and bound by coordinated/covelent bonds or hydrogen bonds and other interactional forces as shown below:

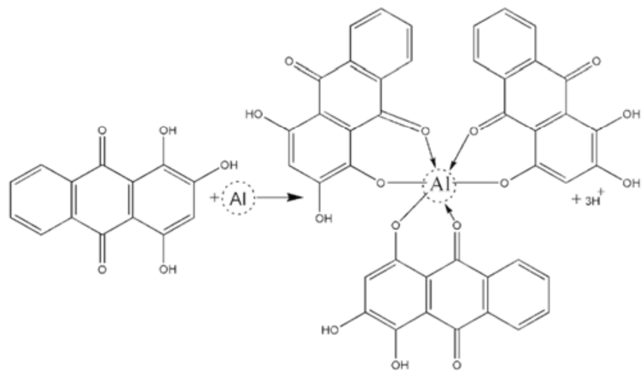

Figure 4. *Mechanism of fixation of natural dyes through mordants.*

Thus, for proper fixation of natural dyes on any textile fibre, mordanting is essential in most of the cases. The said mordanting can be accomplished either before dyeing (pre-mordanting), or during dyeing (simultaneous mordanting) or after dyeing (post-mordanting).

2. Experimental

2.1. Materials

For the present study, such plants a plentiful quantity of that grows in south Asian climate in a commercial scale and a noticeable quantity of which is found as wastage after meeting the internal demands, are selected. About millions people of south asia are directly or indirectly involved in floriculture business [10]. But total of the unsold quantity is, however, left as wastage due to lacking of modern storage facilities. [11]. Marigold and rose flowers that suffer this a lot are selected in this consideration. This two with other selected plants with their binomial name have been listed in table-01.

Table 1. *Binomial names of selected plants.*

Name	Binomial name
Marigold	Tageteserecta. L
Henna	Lawsoniainermis,
Rose	Rosa L

A plain single jersey knitted fabric that was scoured and bleached and bio-polished, was taken as substrate for dyeing. Yarn type: 100% cotton, Yarn count: 32s, GSM: 160, Sample weight = 5 gm.

Chemicals

Ethanol (C_2H_5OH b.p-78.3°C) and acetone (CH_3COCH_3 b.p-56°C) were used as extracting solvents and copper sulphate ($CuSO_4$), was employed as mordent.

2.2. Methods

Extraction of colorants, separation of color components, making substrate suitable for dyeing, treatment involved anchoring of dye-fiber, application of dye extracts on substrates and testing of color retention capability of dyed fabrics were carried in following methods.

2.2.1. Dyes Extractions

The flower heads of marigold and rose and leaves of henna were cut as precise as possible keeping an average size of 1 cm. 200 gm of each of these four plants were weighed and blended by a blending machine separately. Then the blended quantity of each plant was divided into three equal parts which were allowed to dissolve in water, acetone and ethanol (amount must cover the entire sample). On the other hand, 100 gm grains of tea leaves was divided into three equal parts which also were allowed to dissolve in water, acetone and ethanol respectively. All the extraction samples were, however, then heated in water bath for 1 hour at the temperature depending on the boiling point of solvents used.

Table 2. *Extraction Temperature.*

Solvents used	Boiling Temp (°C)	Extraction Temperature (°C)
Water	100	80
Ethanol	78.3	60
Acetone	56	45

At the end of 1 hour all the extraction samples were kept for 24 hours for further extraction of colorants at room temperature. After 24 hours all were strained when almost all the color components had been extracted. Then the strained solutions were double filtered to obtain dye extracts. The solvents (ethanol and acetone) were removed by heating at their boiling point (b.p.) (ethanol b.p-78.3°C, acetone b.p - 56°C). Amount of ethanol and acetone could be recovered by soxhlet apparatus [10, 12]

2.2.2. Mordanting

Mordant that is in fact a chemical link, fixes the dye to a substrate. Natural dyes are not so much color fast with fabrics. This is why all of the twenty samples were treated with 5% (owf) $CuSO_4$ before dying. Treatment of mordanting was carried out for 60 minutes at 100°C temperature with M: L ratio 1: 20.

2.2.3. Dyeing with Extracted Dyes

Mordanted samples ($CuSO_4$) was dyed with every of 3 natural dye solutions (Extracted from Marigold, Henna, Rose,). In the recipe formulation the ratio of material to liquor was kept 1:20, dyeing running time was 60 minutes at 100°C temperature (1.5°C /min). It is needed to be mentioned that no salt or soda was used. Rinsing sequence was cold wash- cold wash-hot wash –cold wash after dyeing. At last 20 dyed samples were collected.

2.3. Test Methods

2.3.1. Evaluation of Color Fastness to Wash Test

For ISO_3, the sample was washed with 5g/l of soap and 2g/l of soda ash in a solution of liquor ratio 50:1, at a temperature of 60°C for 30mins, followed by rinsing and drying. The change in color of the tested specimen and the staining of the adjacent undyed cloths were assessed with the appropriate grey scales.

2.3.2. Evaluation of Color Fastness to Light Fastness Test

The specimen and the blue standard were exposed behind a glass and inserted into the light fastness testing machine. Exposure was carried out for 48hrs. Exposure was terminated after the contrast between the exposed and the unexposed portion of the specimen is equal to the grades on the grey scale, for assessing change in colour. Change in colour was assessed by comparing the tested fabric and original fabric under a white light with the blue standard as reference.

2.3.3. Evaluation of Color Fastness to Rubbing Test

As per ISO 105 E04 method, specimen size at least 50 x 140 mm, Vertical Load - 9 +/- 0.2 N, Finger Diameter – 16 mm Position warp parallel to long dimension for one specimen and weft parallel to long dimension for other specimen or diagonally. Rubbing distance: 104 +/- 3mm. Evaluation is done by Grey scale in a dyed color matching cabinet and rate from 1 to 5.

2.3.4. Evaluation of Color Fastness to Perspiration Test

Sample size will be 10 CM * 4 CM then Wet-0ut the composite test sample in mentioned alkaline or acidic solution at room temperature. The Material ration will be 1:50 and leave for 30 minutes. Then Pour off excess solution and place the composite sample between two glass plate or acrylic plate under a pressure of 4.5 KG and place in an oven for 4 hr. at 37±2 degree centigrade temperature. Then Remove the specimen and hang to dry in warn air not exceeding 60°C. Evaluation is done by Grey scale in a dyed color matching cabinet and rate from 1-5.

3. Results and Discussions

3.1. Assessing Color Fastness

3.1.1. Color Fastness to Wash

Color fastness to wash test (ISO 3) values of samples are presenting in table-3. And comparative analysis of fastness with different mordant on dry and wet condition shown in figure 5.

Table 3. *Color fastness to wash test (ISO 3) values of three samples.*

Samples	Wash test rating	
	Staining	Color change
$CuSO_4$+Marigold	3	4
$CuSO_4$+Henna	3/4	3/4
$CuSO_4$+ Rose	3/4	3/4

Rating: 5 = excellent 4 = very good 3 = good 2 = moderate 1 = poor.

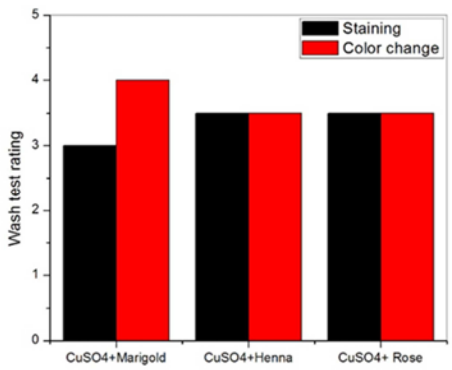

Figure 5. *Comparative analysis of Color Fastness to Wash (Staining, Color Change).*

3.1.2. Color Fastness to Light

Table 4 shows the Color fastness to light test values of samples. Figure 6 indicates the light fastness grade (Blue scale)

Table 4. *Color fastness to light test values of samples.*

Samples	Color fastness to light
$CuSO_4$+Marigold	6
$CuSO_4$+Henna	4
$CuSO_4$+ Rose	5

Ratings: 8=Outstanding (No fading), 7=Excellent (Very slight fading), 6=Very good (Slight fading), 5=Good (Moderate Fading), 4=Moderate (Appreciable fading), 3=Fair (Significant fading), 2=Poor (Extensive fading), 1=Very poor (Very extensive fading)

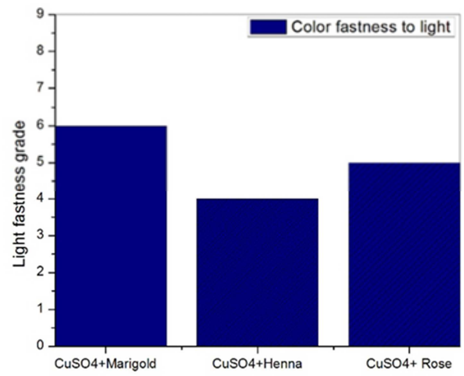

Figure 6. *Comparative analysis ofColor fastness to light.*

3.1.3. Color Fastness to Rubbing

Color fastness to rubbing test in dry and wet condition per ISO 105 E04, ISO- 105-AO3 method displaying on table 5 and figure 7 displays the influence of different mordant on different dyes respectively.

Table 5. *Color fastness to rubbing test (ISO 105 E04, ISO- 105-AO3).*

Samples	Dry Rubbing	Wet Rubbing
$CuSO_4$+Marigold	3	2
$CuSO_4$+Henna	3	2/3
$CuSO_4$+ Rose	3/4	3

Ratings: 5 = excellent 4 = very good 3 = good 2 = moderate 1 = poor.

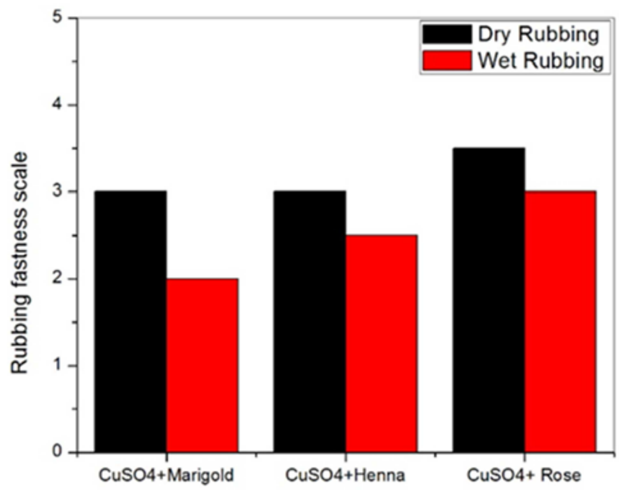

Figure 7. *Comparative analysis of Color fastness to rubbing (wet, dry).*

3.1.4. Color Fastness to Perspiration

Test result on color fastness to perspiration test in acid and alkali condition for three sample shown in table 6 where figure 8 shows the analysis in a row.

Table 6. *Color fastness to perspiration test in acid condition.*

Samples	Perspiration in acidic condition	Perspiration in alkali condition
$CuSO_4$+Marigold	3	2/3
$CuSO_4$+Henna	3	2
$CuSO_4$+ Rose	3	3

Ratings: 5 = excellent 4 = very good 3 = good 2 = moderate 1 = poor.

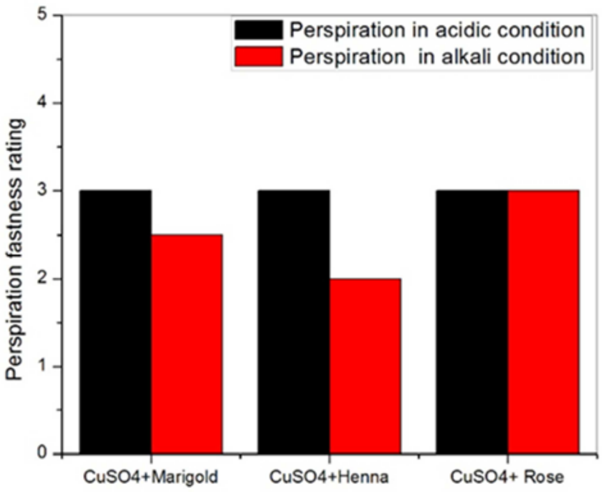

Figure 8. *Comparative analysis of Color fastness to perspiration (acidic and Alkali medium).*

3.2. Discussions

Natural dyes extracted from the selected plants using ethanol and acetone had a well depth of color. The prominence of ethanol and acetone extracted dye solutions over water extracted dyes solutions was observed. It was reported that solvent extraction of natural dyes can lower cost of dyeing while improving their color strength [13-15]. Extracted dyes behaved differently when in contact with different agents, for instance, the dyes which were fast to washing in water might not be fast to light. More over the use of mordants influenced color fastness properties in a wide range. The total test results according to ISO standard has been displayed with mordants used in the figures above. The resistance of a dye or pigment to a chemical or photochemical attack is an inherent property of dye chromophores. Using mordants in the present work,this property was intensified. From the testing reports the present study proves that dyed fabrics had exhibited slight loss in depth while using $CuSO_4$ as mordants for all the plant extracts resulting in fair to good color fastness to washing whereas alum and then tannic acid mordanted dyed fabrics had contributed to poor to fair washing fastness in grey scale rating for changing. Good washing fastness in case of $CuSO_4$ was due to making dye- fibre complex by Cu^{2+} ions and this complex compound had the effect of insolubilizing the dye molecules in the faber making it color fast. On the contrary poor washing fastness for alum and tannic acid might be assessed due to more than one factors such as detaching the dye molecules from fiber components due to the wear of week dye-fiber bonds, ionization of color components as they contain –0H groups etc. The performance of $CuSO_4$ as mordants to make the fabrics color fast to washing has been represented by the following diagrammatic evaluation. But the effect of mordants was quiet different in case of rubbing fastness where alum and tannic acid imparted fair to good grey scale rating for staining but $CuSO_4$ did significant to appreciable loss of depth of color that have been expressed. Most of the natural dyes behave like weak acids. Flavonoids color components from onion skins, tea leaves, Xanthophylls from marigolds, Anthocyanins from roses all are $_{p}$H dependent and are sensitive to alkaline and then acidic solutions. The dyes having –OH groups ionize in alkaline solution [2, 16]. Hence color fastness to perspiration was not so pleasant i,e; there was significant loss in depth of color in alkaline solution whereas in acidic solution the loss was appreciable. The figure 7 exhibits the effects of $CuSO_4$ in both of acidic and alkaline perspiration tests. ISO 105 B02 test method for light fastness provided very pleasant results that $CuSO_4$ mordanted colored samples had shown good to very good light fastness in which degree of fading was moderate to slight respectively in Blue Wool rating. Good light fastness was due to the formation of co-ordination complex between colorants and fibers with Cu^{2+} ions which protect the chromophore from photolytic degradation and the photons sorbed by chromophoric groups dissipate their energy by resonanting within six members ring thus formed and hence protecting the dye.

4. Conclusion

There is a potentiality of the utmost utilization of unsold quantity of marigolds and roses and unused quantity of henna for yielding natural color which will serve environment and earth protecting from pollution and ecological imbalances in other way. It is reported that precise knowledge, compatible documentations and suitable assessment of dye-yielding plants are necessity. Good washing and light fastness using $CuSO_4$ mordents may be a pleasant source of thinking about the commercialization of natural dyes for such textile goods which is in its end uses needed to be color fast while washing and exposing to light specially. To cope with the existing traditional practices and so called disadvantages of natural dyeing, a much more researches and scientific practices are required to go in this area.

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