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Utilization of *Terminalia chebula* Retz. fruits pericarp as a source of natural dye for textile applications

Praveen Onial[1]*, Rameshwar Dayal[1], M S M Rawat[2] and Rakesh Kumar[1]

[1]Chemistry Division, Forest Research Institute, Dehradun, Uttarakhand, India
[2]Department of Chemistry, Hemwati Nandan Bahuguna Garhwal University, Srinagar, Garhwal, Uttarakhand, India

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Terminalia chebula Retz. (Hindi-Harra), Family-Combretaceae, Trade name-Myrobalan, fruits pericarp powder was taken for the utilization as a dye. The dried fruits constitute one of the most important vegetable tanning materials and have been used in India for a long time. This paper describes the isolation of dye from its fruit pericarp (36.24 %) which can impart fast shades on silk, wool and cotton using alum, copper sulphate, ferrous sulphate, potassium dichromate and stannous chloride as mordants with varying degree of colorfastness to washing (4-5), light (4-5), crocking (4-5) and perspiration (4-5). The fruit pericarp may thus find its use for the isolation of dye and is an excellent value addition of the available raw material.

Keywords: *Terminalia chebula* Retz, Fruit pericarp, Colour fastness, Dyes, Mordants.
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Introduction

The colour gives a pleasant look to the substrate as well as express emotions and ideas. Natural materials were the only source of dyes until 1856. At that time Sir Henry Perkin, quite by accident, discovered a mauve coloured coal tar product, which he named "mauvine". This discovery proved to be the beginning of the artificial dye industry. Gradually, natural dyestuffs were discarded and finally with the synthetic indigo blue substitution in production in 1900, natural dyes became a part of the past. The advantages of using natural colorants are manifold as they are eco-friendly, safe for body contacts, unsophisticated and harmonized with nature[1]. Environmental awareness as well as presence of toxicity in the synthetic dyes has revived the interest in the renewable, biodegradable and eco-friendly natural dyes[2,3]. Therefore the demand for natural dyes is increasing constantly and new sources of natural dyes are required to be explored for sustainable supply of natural dyes[4]. Also suitable combinations of natural dyes and mordant can produce some unique shades.

Terminalia chebula Retz. (Family Combretaceae) a tree 15-24 m in height and 1.5-2.4 m in girth, is found in the sub-Himalayan tracts from the Ravi eastwards to West Bengal and Assam, ascending up to an altitude of 1,500 m in the Himalayas. The mature fruits are collected during January-April by shaking the trees, and are dried in thin layers, preferably in shade, and graded for marketing.

The raw Myrobalans are graded under different trade names, selection being based upon their solidness, colour and being free from insects attack[5]. The dried flesh surrounding the seed is rich in tannin (av 30-32 %) whose content considerably varies with the different grades of myrobalans from different areas.

The hydrolysable tannins, chebulagic acid ($C_{41}H_{30}O_{27}.H_2O$; m p >240 ^oC), chebulinic acid ($C_{41}H_{32}O_{27}$) and corilagin are the major tanning constituents present in myrobalans; these belongs to ellagitannin class. They are accompanied by varying proportions of the following products on their complete and incomplete hydrolysis such as-chebulic acid ($C_{14}H_{12}O_{11}$), 3:6-digalloylglycose ($C_{20}H_{20}O_{14}$), ellagic acid, gallic acid, and β-D-glucogallin. The presence of terchebin, 1,3,6-trigalloylglucose and 1,2,3,4,6-pentagalloylglucose has been reported[6]. Other constituents include phenolics such as chebulinic acid, ellagic acid and anthraquinones. Some of the other minor constituents are polyphenols such as corilagin, galloyl glucose, punicalagin, terflavin A and maslinic acid[7]. Besides, fructose, amino acids, succinic acid, β-sitosterol, resin and

*Correspondent author:
E-mail: onialp@icfre.org

purgative principle of anthraquinone are reported to be present[8,9]. Phytochemicals such as flavonol, glycosides, triterpenoids, and coumarin conjugated with gallic acids called chebulin as well as other phenolic compounds were also isolated[7,10-12]. Twelve fatty acids were isolated from *T. chebula* of which palmitic acid, linoleic acid and oleic acid were the main constituents[13]. Triterpenoid glycosides such as chebulosides I and II, arjunin, arjunglucoside, 2α-hydroxyursolic acid and 2α-hydroxymicromiric acid have also been reported[14]. The leaves were found to contain polyphenols such as punicalin, punicalagin, terflavins B, C and D[6,15,16]. Phloroglucimol and pyrogallol, along with phenolic acids such as ferulic, p-coumaric, caffeic and vanillic acids are also reported[17]. Oil extracted from kernels yielded palmitic, stearic, oleic, linoleic, behenic and arachidic acids[17].

During maturation of the fruits, the amount of tannin decreases whereas the acidity increases. The literature survey revealed that no study has been done on the natural dye from fruits and other parts of the plant. Therefore, systematic study on the dye from fruits of *T. chebula* was carried out. The study on natural dye from the fruits will be a value addition besides its use in many other Ayurvedic preparations, tanning and cosmetics.

Materials & Methods

The fruits of *T. chebula* were collected in March 2008 from the campus of Forest Research Institute (FRI), Dehradun and dried in shade. The specimen was identified by Dr. Sas Biswas, Ex Head of the Systematic Botany Division, FRI and a voucher specimen (No. 156864-65 7/3/2008) was deposited in the Herbarium at FRI, Dehradun. The fruits pericarp was separated and powdered (approx 50 meshes), Plate 1a. The mordant viz, alum, copper sulphate, ferrous sulphate, potassium dichromate and stannous chloride used in this study were of LR grade. Pure handloom silk, wool and cotton were used in dyeing experiments. The Optical density (OD) was measured by Chemito 2500 UV–VIS spectrophotometer. IR spectra were recorded on a FT-IR (Shimadzu). Washing and light fastness were measured by using laundrometer of Metrex Scientific Instrument (P) Ltd. and Digital Light Fastness Tester of Labin Scientific Instruments and Calibration Pvt. Ltd., respectively while perspiration fastness and rubbing fastness were determined by using perspirometer and crockmeter, respectively of Globe–Tex Industries. The CIEL*a*b* values were determined on a reference bench Top Spectrophotometer of Gretag MC Bath (Colour Eye) 7000, USA.

Process optimization for dye extraction

Prior to extracting the dye, plant material to solvent ratio and time of extraction were optimized. (2, 4, 6, 8, 10 and 12) g of dried and powdered plant material were placed in 6 beakers and each of them were extracted with 100 mL of water for 60 min at 95-98 ^{o}C, filtered and made to 100 mL. One mL of aliquot was diluted to 100 mL and the optimum amount of fruit pericarp powder to water ratio for dye extraction was determined by measuring the OD. Similarly, the optimum time for extraction was determined by measuring the OD.

Extraction of dye

Based on the optimization, solid dye from the fruit pericarp was extracted by heating them with water (1:10) at 95-98 ^{o}C for 60 min. The dye solution was filtered and evaporated to dryness to obtain the solid dye (36.24 %), Plate 1b.

Physico-chemical evaluation of the dye

To maintain the quality, extracted dye was evaluated for its physico-chemical traits, viz. moisture content, melting point, color with $FeCl_3$, solubility, pH, total ash, water-soluble ash, acid-insoluble ash and spectral properties IR & UV.

Determination of optimum dye concentration

Dye solutions of different concentration (0.2 to 1.2 %) were prepared and the OD of each solution was measured after diluting 1 mL of solution to 100 mL with water. Thereafter 6 silk samples of 1.2 g each were dyed in each of the respective dye solution for 60 min at 80-85 ^{o}C. The OD of each dye solution was also measured after dyeing. The residual dye solution was made up to 100 mL and then diluting 1 mL of this solution to 100 mL. Similar concentrations (0.2 to 1.2 %) were used for measuring the OD before and after dyeing wool and cotton samples.

Determination of optimum mordant concentration

Mordant solutions of (0.2 to 1.0 %) were prepared and the OD of each solution was measured after diluting 1 mL of solution to 100 mL with water. Mordant concentration of 0.5 g / 100 mL was found to be optimum by visual observations of dyed samples prepared under different conditions. It was also experimented that a maximum of 1.2 g of silk could be dyed in 0.8 % of dye solution. The criteria for

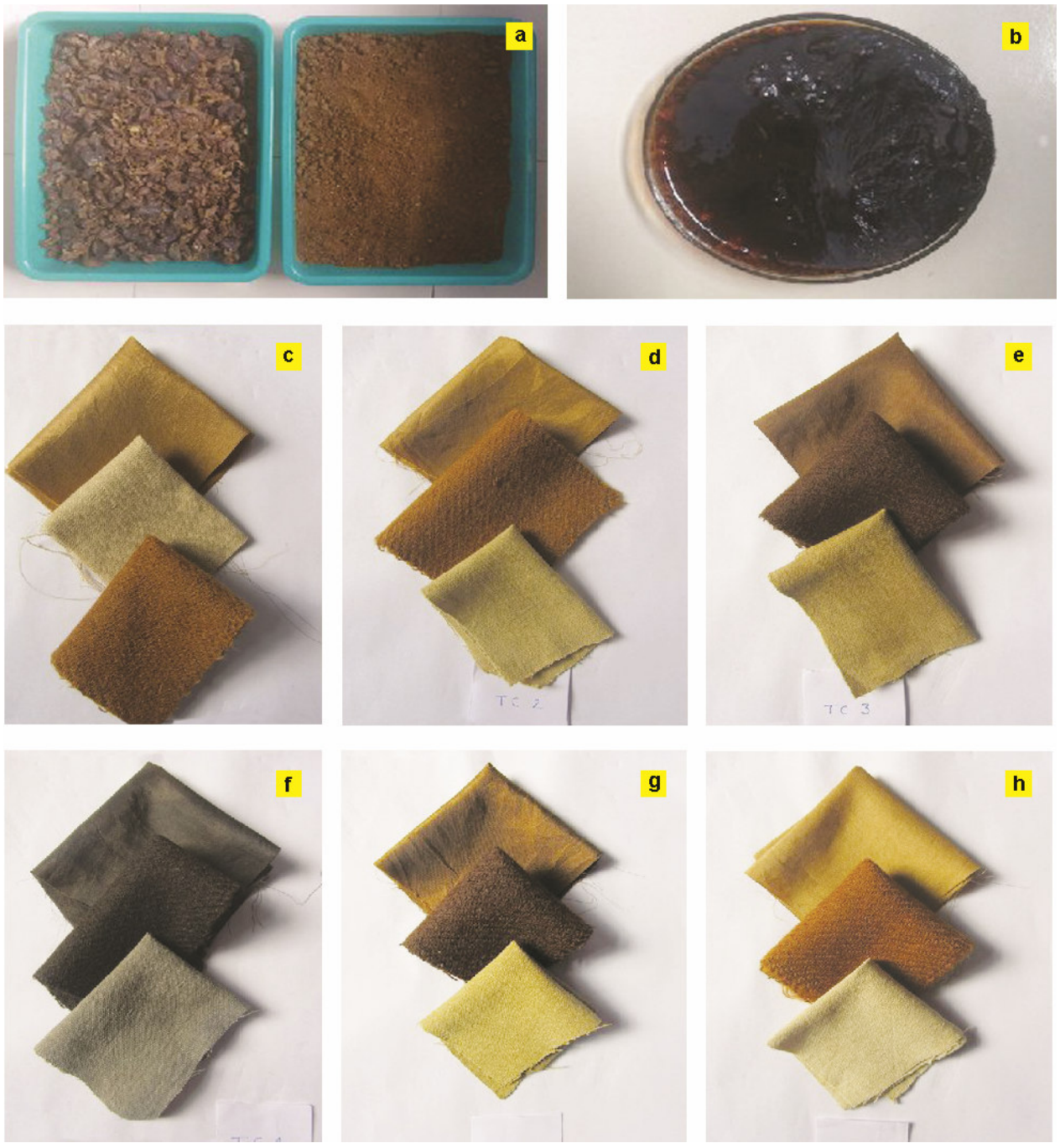

Plate 1—Dyed Silk, wool and cotton from *Terminalia chebula* Retz fruit pericarp dye: a. Fruit pericarp (powdered); b. extracted dye from fruit pericarp; c-h. samples of dyed silk, wool and cotton – (c) without mordant, (d) mordant-alum, (e) mordant copper sulphate, (f) mordant ferrous sulphate, (g) mordant potassium-dichromate, (h) mordant-stannous chloride.

evaluation were the depth of colour, evenness of dye and brightness of shade. The mordanted textile samples were named as TC 1-without mordant, TC 2-Alum, TC 3-Copper sulphate, TC 4-Ferrous sulphate, TC 5-Potassium dichromate and TC 6-Stannous chloride respectively.

Scouring of fabrics

The fabric was washed for 30 min in a bath containing 0.5 g/L sodium carbonate and 2 g/L of a non-ionic detergent at 40-45 ^{o}C to remove any impurities. Fabric was then thoroughly washed with water and air-dried at ambient temperature.

Subsequently it was soaked in tap water for 5 min prior to dyeing.

Dyeing

Dyeing of silk, wool and cotton samples were carried out by post mordanting method, which produced improved shades in terms of hue and darkness. The dye bath was set at 8 g/L of the dye by keeping the material to liquor ratio of 1:10. The dye bath was heated up to a temperature of 80-85 ^{o}C and maintaining the solution level for 45 min for dyeing the silk and wool samples and a temperature of 95-98 ^{o}C was maintained for 60 min in dyeing of cotton. The dye bath was cooled to room temperature and the dyed fabrics were directly transferred to mordant bath for mordanting.

Mordanting of dyed samples

The mordant solution (0.5 %) prepared in distilled water was used for the mordanting. The dyed samples were directly immersed in the mordant bath by keeping the material to liquor ratio of 1:10. The bath was heated to a temperature of 80-85 ^{o}C for 30 min for fixing the dye on silk and wool samples. In case of cotton, the mordanting was continued for 60 min at a temperature of 95-98 ^{o}C. The samples were taken out on cooling, washed with water and dried in shade (Plate 1c-h).

Fractionation of dye

In order to find out the dyeing fraction, the isolated dye was fractionated into methanol soluble and insoluble fraction. The dye was refluxed with methanol for one hr on a water bath. The methanol soluble portion was removed and the process was repeated two more times for the complete recovery. All methanol soluble fractions were combined together and solvent was removed by evaporation on water bath and the yields were calculated.

Determination of colorfastness

Washing fastness test was carried out according to Colorfastness to washing method: IS 687: 1979 temp 40 ^{o}C, liquid vol 150 mL, time 30 min, steel balls 10, sodium oleate 5g/L and light fastness using MBTL Colorfastness to light method: IS: 2454: 1985. Rubbing fastness using Colorfastness to rubbing method, IS: 766: 1988 and perspiration fastness was done using Colorfastness to Perspiration (silk) method: IS 971: 1983, respectively[18]. The samples were assessed against the standard grey scales for change of colour and staining while for light fastness comparison up to blue scale 4 was done.

Results and Discussion

Optimization of plant material to solvent ratio and time for extraction

Based on the mean highest OD values, the optimum plant material to solvent (water) ratio (M:L) was determined as 1:10 (OD_{max} 2.144) and extraction time was found to be 60 min (OD_{max} 2.162) at 95-98 ^{o}C for extraction of dyes from the fruit pericarp.

Physico-chemical evaluation

The average dye yield was recorded to be 36.24 % on moisture free basis. The extracted dye appeared as dark brown powder, m p 280 ^{o}C and produced dark blue-black color with alcoholic ferric chloride. It showed UV absorption λ_{max} (MeOH) at 212 nm and IR spectrum showed bands at ν_{max} (KBR) 3703.45, 3419.9, 2926.11, 2160.33, 1705.13, 1618.33 /cm. The average moisture content was 8.6 % and solubility of the dye in cold water, hot water and methanol was found to be 75.4, 85 and 66.7 %, respectively and pH of 1% solution was recorded to be 3.5-4. The total ash, water soluble ash and acid insoluble ash content were recorded as 6.97, 70.43 and 1.18 %, respectively.

Optimization of dye concentration

Data pertaining to optimization of dyeing conditions indicated 0.8 % as the optimum concentration of dye for dyeing 1 g of silk, based on highest absorption (11.95 %). Optimum dyeing concentration for silk and wool fabric was found to be 0.8 % with M:L ratio of 1:40, extraction time, 45 min at 80-85 ^{o}C. In case of cotton fabrics, optimum dye concentration and M:L ratio were same as that for silk and wool whereas dyeing time and temperature were found to be 60 min at 95-98 ^{o}C.

Optimization of mordant concentration

Mordanting of dyed fabrics was carried out at 0.2, 04, 0.6, 0.8 and 1.0 % with M:L ratio of 1:40. Mordant concentration of 0.5 g/100 mL and 30 min mordanting time for silk and wool and 45 min for cotton were found to be optimum by visual observations of dyed samples mordanted under different mordant concentration and time. The criteria for evaluation were the depth of color, evenness of dye and brightness of shade.

Fractionation of the dye

Fractionation of dye with methanol yielded methanol soluble (66.7 %) and methanol insoluble (33 %).

Colorfastness to washing

The wash fastness values of silk, wool and cotton samples dyed using above mordants is depicted in Table 1. The data indicate that wool and silk samples have very good wash fastness properties (4-5) while the fastness of cotton samples were (4), which indicate that the dye is very good in case of wool, silk and cotton.

Colorfastness to light

The values of light fastness (Table 2) indicate that the silk, wool and cotton samples dyed using the fruit pericarp dye with alum, copper sulphate, potassium dichromate, iron salts and tin salts as mordants have very good light fastness (4).

Colorfastness to rubbing

The results of the colorfastness to crocking have been given in Table 3. The values indicate that silk, wool and cotton samples dyed with alum, copper sulphate, potassium dichromate, iron salts and tin salts as mordants have very good (4-5) fastness under dry as well as under wet conditions for all mordants.

Colorfastness to perspiration

The dyed samples were also tested for their colorfastness to perspiration (Table 4). The color change and staining in acidic and alkaline medium varied between 4-5 of the silk, wool and cotton samples dyed using alum, copper sulphate, potassium dichromate, iron salts and tin salts as the mordant.

Determination of colour coordinates for dyed samples

Variation in `L' values (Table 5) indicate that the maximum dark color was obtained using ferrous sulphate in silk (33.96), potassium dichromate in wool (30.41) and using ferrous sulphate in cotton (45.57) fabrics. The silk samples without any mordant has the

Table 1—Colorfastness to washing

S. No.	Test Fabric	Change in Color of Test Fabric on Grey Scale			Staining on Adjacent Fabric 1			Staining on Adjacent Fabric 2		
		Silk	Wool	Cotton	Silk (Silk)	Wool (Wool)	Cotton (Cotton)	Silk (Cotton)	Wool (Cotton)	Cotton (Wool)
1.	TC1	4-5	4-5	4	4-5	4-5	4-5	4-5	4-5	4-5
2.	TC2	4-5	4-5	4	4-5	4-5	4-5	4-5	4-5	4-5
3.	TC3	4-5	4	4	4-5	4-5	4-5	4-5	4-5	4-5
4.	TC4	4	4	4	4-5	4-5	4-5	4-5	4-5	4
5.	TC5	4	4	4	4-5	4-5	4-5	4-5	4-5	4-5
6.	TC6	4-5	4	4	4-5	4-5	4-5	4-5	4-5	4-5

Silk: - Adjacent Fabric 1: Silk and Adjacent Fabric 2: Cotton; Wool: - Adjacent Fabric 1: Wool and Adjacent Fabric 2: Cotton; Cotton: - Adjacent Fabric 1: Cotton and Adjacent Fabric 2: Wool.

Table 2—Colorfastness to light

S. No	Test Fabric	Change in Color of Test Fabric on Grey Scale			Change in Color of Test Fabric on Blue Wool Standard		
		Silk	Wool	Cotton	Silk	Wool	Cotton
1.	TC1	4-5	4-5	4-5	4	4	4
2.	TC2	4-5	4-5	4-5	4	4	4
3.	TC3	4-5	4-5	4-5	4	4	4
4.	TC4	4-5	4	4-5	4	4	4
5.	TC5	4-5	4	4-5	4	4	4
6.	TC6	4-5	4-5	4-5	4	4	4

Table 3—Colorfastness to rubbing

S. No.	Test Fabric	Staining on rubbing cloth Dry rubbing			Staining on rubbing cloth Wet rubbing		
		Silk	Wool	Cotton	Silk	Wool	Cotton
1.	TC 1	4-5	4-5	4-5	4-5	4-5	4-5
2.	TC 2	4-5	4-5	4-5	4-5	4-5	4-5
3.	TC 3	4-5	4-5	4-5	4-5	4	4-5
4.	TC 4	4-5	4-5	4-5	4	4-5	4-5
5.	TC 5	4-5	4-5	4	4	4-5	4
6.	TC 6	4-5	4-5	4	4-5	4-5	4

Table 4—Colorfastness to perspiration

S. No.	Test Fabric (silk)	Acidic			Alkaline		
		Change in Color of Test Fabric on Grey Scale	Staining on Adjacent Fabric 1	Staining on Adjacent Fabric 2	Change in Color of Test Fabric on Grey Scale	Staining on Adjacent Fabric 1	Staining on Adjacent Fabric 2
silk							
1.	TC 1	4-5	4-5	4-5	4	4-5	4-5
2.	TC 2	4-5	4-5	4-5	4-5	4-5	4-5
3.	TC 3	4	4-5	4-5	4-5	4-5	4-5
4.	TC 4	4	4-5	4-5	4	4-5	4-5
5.	TC 5	4	4-5	4-5	4	4-5	4-5
6.	TC 6	4-5	4-5	4-5	4	4-5	4-5
wool							
1.	TC 1	4-5	4-5	4-5	4	4-5	4-5
2.	TC 2	4-5	4-5	4-5	4-5	4-5	4-5
3.	TC 3	4-5	4-5	4-5	4	4-5	4-5
4.	TC 4	4-5	4-5	4-5	4	4-5	4-5
5.	TC 5	4	4-5	4-5	4-5	4-5	4-5
6.	TC 6	4	4-5	4-5	4-5	4-5	4-5
cotton							
1.	TC 1	4	4-5	4-5	4	4-5	4-5
2.	TC 2	4	4-5	4-5	4	4-5	4-5
3.	TC 3	4	4-5	4-5	4	4-5	4-5
4.	TC 4	4	4-5	4-5	4	4-5	4-5
5.	TC 5	3-4	4	4	3-4	4	4-5
6.	TC 6	4	4-5	4-5	4	4-5	4-5

Adjacent Fabric 1: Silk and Adjacent Fabric 2: Cotton in case of silk,
Adjacent Fabric 1: Wool and Adjacent Fabric 2: Cotton in case of wool,
and Adjacent Fabric 1: Cotton and Adjacent Fabric 2: Wool in case of cotton.

Table 5—CIEL*a*b* and K/S Values of dyed fabrics

Sl. No.	Sample	L*	a*	b*	C*	h^0	K/S
Silk							
1.	TC1	60.23	7.49	29.64	30.57	75.82	9.68
2.	TC2	55.88	6.92	28.66	29.48	76.43	9.09
3.	TC3	48.59	7.56	24.82	24.92	73.03	10.65
4.	TC4	33.96	1.21	7.54	7.64	80.91	12.53
5.	TC5	51.35	7.49	30.28	31.19	76.10	11.07
6.	TC6	57.84	7.18	28.15	29.06	75.69	8.32
Wool							
1.	TC1	51.73	9.78	32.57	34.01	73.29	21.84
2.	TC2	45.88	8.03	30.73	31.76	75.35	21.43
3.	TC3	30.61	6.09	19.55	20.47	72.70	27.91
4.	TC4	31.04	0.77	8.41	8.44	84.73	20.38
5.	TC5	30.41	8.15	22.42	23.85	70.03	29.68
6.	TC6	49.69	12.23	39.07	40.94	72.61	25.61
Cotton							
1.	TC1	70.05	2.60	14.68	14.91	79.96	2.21
2.	TC2	65.45	4.16	21.43	21.83	79.03	3.50
3.	TC3	61.57	3.70	20.11	20.45	79.56	4.13
4.	TC4	45.67	0.91	3.71	3.82	76.14	4.77
5.	TC5	66.31	1.20	17.98	18.02	86.17	3.11
6.	TC6	74.60	3.19	17.83	18.11	79.85	1.96

lightest colour (L=60.23). The L* values for other silk samples mordanted with alum, copper sulphate, potassium dichromate and stannous chloride is 55.88, 48.59, 51.35 and 57.84 respectively. Maximum redness was observed by using copper sulphate, stannous chloride and alum as mordant in silk (7.56), wool (12.23) and cotton (4.16), respectively. Minimum redness was found in silk (1.21), wool (0.77) and cotton (0.91) with ferrous sulphate. The maximum yellowness was obtained by using potassium dichromate as a mordant in silk (30.28), wool (39.07) and cotton (21.43), respectively where as the minimum yellowness in silk (7.54), wool (8.41) and cotton (3.71) was obtained using ferrous sulphate as a mordant. The K/S values for silk shows that sample mordanted with stannous chloride has the lowest K/S value (8.32) while ferrous sulphate has the highest K/S value (12.53), where as silk mordanted with alum, copper sulphate and potassium dichromate have the K/S values 9.09, 10.56 and 11.07, respectively. Wool mordanted with alum, copper sulphate, ferrous sulphate, potassium dichromate and stannous chloride has the K/S values of 21.43, 27.91, 20.38, 29.68 and 25.61 which indicate that wool mordanted with potassium dichromate has the darkest shade. Cotton has lesser K/S values in comparison with silk and wool, indicating their lighter shades. Cotton dyed and mordanted with ferrous sulphate has maximum K/S value (4.77) whereas when mordanted with stannous chloride has minimum K/S value (1.96).

Conclusion

T. chebula fruits are available widely but not fully utilized and hence extraction of dye may lead to greater utilization. A process has been developed for the isolation of dye from fruit pericarp. Our study indicates that the pericarp can be used as a source of vegetable dye and the dye yield is more from the air dried than the fresh fruits pericarp. During dyeing only $1/4^{th}$ of dye is taken up by the dyed fabric and $3/4^{th}$ remains, which can be reused for dyeing new fabric of similar weight three more times. Natural dye pilot-plant has been installed and commissioned for the first time in FRI, Dehradun for exploring the possibility of forest biomass utilization for value added products. Trials were carried out in pilot-plant at 20-40 kg batch scale. A number of shades were produced from the isolated dye and their blends were prepared. The dye yield and the colorfastness to washing, light, crocking, and perspiration are very good (4-5) which is among the best from the plant source. Extracted dye has great potential as a source of sustained livelihood for local & tribal people, cottage industry and may also improve forest productivity. The dye may find use in dyeing of silk, wool and cotton which will help to augment the export of natural dyed garments to the countries where the use of azo dyes has been banned. Natural dyeing technology has employment generation potential and justifies the utilization of *T. chebula* fruits as a source of dye without harming the tree. *T. chebula* fruits pericarp powder is one of the main ingredients of the edible Ayurvedic formulation 'Trifla'. Fabrics dyed with *T. chebula* dye are safe to wear.

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