COLLEGE OF Textiles NC STATE UNIVERSITY

JTATM
Journal of Textile and Apparel, Technology and Management

NC STATE UNIVERSITY

Volume 6, Issue 1, Spring2009

Utilization of Temple waste flower -*Tagetus erecta* for Dyeing of Cotton, Wool and Silk on Industrial scale

Padma S. Vankar*[a], Rakhi Shanker[a], Samudrika Wijayapala[b],
[a]Facility for Ecological and Analytical Testing
Indian Institute of Technology, Kanpur-208 016, India
[b]Department of Textile and Clothing Technology,
University of Moratuwa, Moratuwa, Sri Lanka

ABSTRACT

Huge amounts of Tagetus erecta (marigold) flowers are offered in temples in India, creating a very large waste. These waste flowers were collected and used for industrial dyeing. Tagetus belongs to the family Asteraceae. It produces natural dye from its flowers (petals) consisting mainly of carotenoid-lutein and flavonoid- patuletin, these colorants have been isolated and identified. The crude extract has been used for dyeing textiles. In the present study innovative dyeing with Tagetus has been shown to give good dyeing results. Pretreatment with 1-2 % metal mordant and 5 % of plant extract (owf) was found to be satisfactory and showed very good fastness properties to dyed fabrics of cotton, wool and silk. Innovative solvent extraction of dry flowers of Tagetus erecta with ethanol was found to have high extraction efficiency allowing selective extraction of flavonoids and carotenoids. This is an easy extraction process, where ethanol solvent was removed and recovered. Crude extract was diluted with deionsed water and used for dyeing. CIELab and K/S of the dyed fabrics were also evaluated. The superiority of solvent extraction over conventional extraction has been established through this study.

Keywords: Tagetes erecta, Ethanolic extraction, Cotton, Wool, Silk, Natural dyeing, Industrial scale

1. Introduction

Tagetus erecta (Marigold) belongs to the family Asteraceae. It is a small shrub and bears yellowish orange flowers in abundance during the flowering season which lasts for more than 6-8 months. It occurs in humid climate in different parts of India and even in Sri Lanka. *Tagetus* pigments mainly consists of carotenoids and flavonoids, these have been used mainly as natural food colorants and feed [1]. *Tagetes erecta* L. flower pigments have been extracted and used as a natural food additive to color egg yolks orange and poultry skinyellow [2]. Five cultivars were examined by Bosma *et al.* for lutein production on commercial scale. 'E-1236' (a variety of *Tagetes)* was found to produce the largest quantity of lutein, 22.0 kg ha^{-1}, and 'Orange Lady' (another hybrid variety) produced 21.3 kg ha^{-1}.

The flowers are offered to the deities in temples and are thus available in huge quantities as temple waste. The total waste generated in Kanpur city alone has been estimated to be 20 tonnes per day. Most of these flowers are either dumped by the side of river Ganga or allowed to naturally decay and used as compost. We have made use of this waste material and utilized its colorant for dyeing purpose. Figure-1 shows the picture of the Tagetus flower.

Aqueous extract from Tagetus has shades of light yellowish green hue color. A

Figure-1 Tagetus flower

Traditional Knowledge of its use: Ethanolic extract of the yellow to orangish red flowers of Tagetus, have been known as rich source of lutein. This pigment has acquired greater significance because of its antioxidant property and for its use in eye health protection. Although Tagetus flower extract has been mainly used in veterinary feeds and as a natural food colorant. The chemical processing and stability of the pigment and its applications in these areas has been well established, but it has not been explored for fabric dyeing on commercial scale because of some drawbacks. The aqueous extract of the flower does not produce attractive hue color. Color fastnesses of these dyed fabrics do not fit into the acceptable norms of natural dyeing.

Since proper extraction of the colorant seemed to be the major drawback, we planned our study with *Tagetus* by developing an efficient extraction process using ethanol. The innovative extraction method gave better dye yield and deeper hue color to the extract. The ethanol was

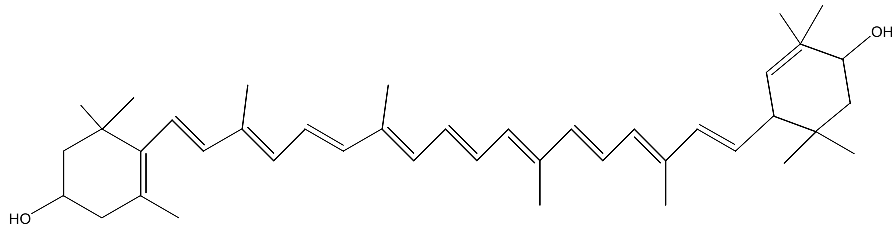
1.2.

laboratory experiment [3] showed that use of metal salts (mordants) can produce different colors on fabrics dyed with its flower extract. This seems to demonstrate the potentiality of colorant for fabric dyeing. As the flower has good pigment content we chose Tagetus erecta flowers for the study of natural dyeing.

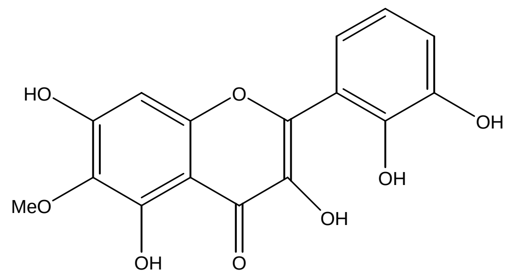
removed under reduced pressure on a rotatory evaporator, aqueous extract was prepared and used for natural dyeing of the three natural materials.

Although some attempts on natural dyeing with *tagetus* has been made by natural dyers. Jute dyeing has been attempted by Pan et al. [4,5] and another group the dye for silk dyeing [6]. Excellent colors were produced on sheepskin leather [7] with this dye when it was used in conjunction with three natural tannins and three mordants. Agarwal et al. [8] studied fabric printing and its effect with seven mordants through simultaneous mordanting on cotton fabric.

1.1. Chemical structure of the yellow colorant: Lutein was isolated according to the method developed by Li [9]. Dry flowers were ground to 10-40 mesh size and dissolved in 8 % ethanolic solution of KOH and washed with the same solution 4-6 times at 65-70 ºC under stirring. Addition of water followed by neutralization and filtration yielded an extract, this extract was further extracted with ethyl acetate 6-10 times, finally crude lutein was obtained after distillation of ethyl acetate and recrystalization from THF and deionized water yielded the final product of lutein crystal with a purity >90%. The isolated molecule was identified by NMR, Mass spectra and the structure was as shown in figure-2 a.



2. 2a -Lutein



2b - Patuletin

Figures 2- Structures of Main colorants: 2a) Lutein 2b) Patuletin

1.2. Flavonoids in *Tagetus*: Literature procedure of extraction of flavonoids from *Tagetes patula* L. by Guinot et al.[10] showed preferential isolation of Patultrin and patuletin as main flavonoids and investigated for their dyeing potential on wool using only alum mordant.. The investigators had used a tedious method of macerating the flowers and had used solvents ethanol: water in varied composition 2:8, 3:7 and 5:5 v/v for extraction.

However we carried out column chromatography of crude extract on silica gel using the elution systems 20% EtOAc/hexane- 50% EtOAc/hexane and further on with 40% EtOAc from *Tagetus erecta* extract to isolate one major flavonoid - patuletin. It was identified by NMR and Mass spectra and the structure was as shown in figure-2 b. The spectral data were matched with literature data.

2. Experimental

2.1. Materials

The cotton fabric of 105 g/m^2 (warp-30 , weft-20) was scoured with a solution containing 5 g/l of sodium carbonate and 3 g/l of non-ionic detergent (Labolene) under the boiling condition for 4 h, after this it was thoroughly rinsed and air dried at room temperature.

The munga silk of GSM-45 fabric was scoured with solution containing 0.5 g/L sodium carbonate and 2 g/L non-ionic detergent (Labolene) solution at 40-45° C for 30 min, keeping the material to liquor ratio at 1:50. The scoured material was thoroughly washed with tap water and dried at room temperature. The scoured material was soaked in clean water for 30 min prior to dyeing or mordanting.

The pure new wool(J sagar) 60 gm was scoured with solution containing 2 g/L non-ionic detergent (Labolene) solution at 30-35° C for 30 min, keeping the material to liquor ratio at 1:50. The scoured material was thoroughly washed with tap water and dried at room temperature. The scoured material was soaked in clean water for 30 min prior to dyeing or mordanting.

2.2. Methods

Since huge amount of flowers were easily, cheaply and readily available we planned our natural dyeing study with

Tagetus by developing an efficient extraction process for better dye yield. Aqueous extract was used for dyeing although the initial extraction of the colorant was done with ethanol from the dried flowers of *Tagetus erecta* as described in 2.2.1. The ethanol was removed and aqueous extract was prepared and used for dyeing. Pretreatment of the fabric was done with Tannic acid (only in the case of cotton). Cotton fabrics have been reported to be pretreated with tannic acid [11] that provide carboxylic acid (-COOH) groups to the fabric for better dye adherence. The premordants used were alum, ferrous sulphate, stannic chloride, stannous chloride, copper sulphate and potassium dichromate. All other chemicals used were laboratory grade reagents.

2.2.1. Extraction of the dye

Innovative extraction: 100 gm of dry flowers from *Tagetus* were crushed and dispersed in ethanol (500 ml) and heated to (50^0C) in a round bottom flask kept over water bath for quick extraction for 1.5 hours. All the colors were extracted from flowers by the end of 1.5 hours(our unpublished work). After extraction the extract was filtered through ordinary filter paper, the filtrate was collected, and the solvent was evaporated on rotatory evaporator and recovered to dryness. 100 ml of distilled water was added to this extract. The absorbance was recorded for determination of concentration of the aqueous extract. UV-Visible spectrum of the extract showed higher color content in this case. This was further used for dyeing the fabrics as shown in figure-3. Dye yield from the ethanolic extract has been shown in table-1.

Conventional extraction: The dried flowers of *Tagetus* (100gms) were crushed and dissolved in distilled water(500ml) and allowed to boil in a beaker kept over water bath for extraction for 3 hour. All the color was extracted from flowers of *tagetus* by the end of 3 hours. The solution was filtered, evaporated to half volume (250 ml). The extract was used for dyeing for the comparison purpose. UV-Visible spectrum of the extract showed poor color content in this case. Dye yield from the aqueous extract has been shown in table-1. CIELab and K/S of the dyed fabrics were evaluated for the differently extracted dye solutions. However both in terms of dye content and hue color, it was not satisfactory as shown in table-2 and figure-3. Thus this extract was not found suitable for dyeing purpose.

2.2.2. Treatment of fabric before dyeing

After scouring of cotton fabric it was treated with 2 % (owf) solution of tannic acid in water. The fabric was dipped in tannic acid solution for at least 4-5 hours. It was squeezed and dried. Pre-mordanting was used for this study, fabric which was already treated with tannic acid was dipped, in mordant (2% for alum and 1% for stannous chloride, stannic chloride, ferrous sulphate, copper sulphate, and potassium dichromate separately) solution and was kept on water bath at 40°C for one hour. It was squeezed and dried. Silk and wool were directly premordanted with metal salts, no tannic acid treatment was required in the latter cases.

2.2.3. Dyeing

Dyeing was carried out in following manner: A stepwise dyeing of pretreatment of cotton only with tannic acid (2%, owf) and then mordanting(in the ratio of 1 or 2 % mordant, owf) was used for dyeing with aqueous extract of *tagetus* (5%, owf) at its original pH. The dyeing process was carried out for 2 hour at temperature 30-40^0C [12]. The dyed fabrics were dipped in saturated brine solution (15 min) which acts as dye-fix and then rinsed thoroughly in tap water and the dyed fabrics were allowed to dry in open air. The colorimetric data obtained from dyed fabrics and yarn which had been pretreated with tannic acid/metal mordants in the case of cotton and only metal mordants in the cases of silk and wool reveal that pretreatment markedly improved the wash fastness, in terms of change of shade of the dyed fabrics with respect to controlled samples. It also increased the color strength and flattened the shade of the dyeings. In each experiment controlled dyed samples were also prepared.

2.3. Analysis of isolated pigments from extracted dye

The extracted dye was dissolved in a suitable solvent system (water) and scanned through on Heλious α UV lamda UV-Visible spectrophotometer. Fourier transform – infra-red spectrum was recorded on Vertex 70 model of Bruker. Nuclear Magnetic Resonance: Isolated fractions from column chromatography were analyzed on Jeol JNM LA 400MHz model. Mass spectrometry was carried out on Gas chromatograph mass spectrometer Finnigan, Fission Model. The GC/MS was equipped with a capillary column DB-5 (30 m × 0.25 mm × 0.25 µm film thickness). The GC column was operated in a temperature programmed mode with an initial temperature of 60°C held for 5 min, then the temperature was raised up to 280°C at a rate of 10°C min^{-1}, and held at this temperature for 5 min. The injector and interface temperatures were 250 and 280°C, respectively.

2.4. Fastness Testing of dyed samples

The dyed samples were tested according to Indian standard methods [13]. Color fastness for light, **IS-2454-85**; color fastness to rubbing, **IS-766-88**; color fastness to washing; and **IS-687-79**; color fastness to perspiration, **IS-971-83.** Xenoster was used to test the light fastness of the dyed fabric. Wash wheel of Thermolab model was used test the washing fastness of the dyed fabric. Perspirometer of Sashmira model was used for the testing of perspiration fastness of the dyed fabric. Crock meter of Ravindra engg. Model was used for testing the rubbing fastness of the dyed fabric. Color Matching system used for the measurement of reflectance of dyed fabrics was on a Premier Colorscan.

2.5. Measurements and analysis: Color measurements

The relative color strength of dyed fabrics expressed as K/S was measured by the light reflectance technique using the Kubelka–Munk[14,15] equation. The reflectance of dyed fabrics was measured on a Premier Colorscan.

2.6. Dye exhaustion

The dye exhaustion percentage (%*E*) was calculated according to the following equation (1).

$$\%E = [A0 - A_r / A0] \times 100 \quad (1),$$

where A_0 and A_r are, respectively, the absorbance of the dye bath before and after dyeing at λ_{max} 265 and 392 nm of the dye used. The absorbance was measured on a Heλios UV-Vis spectrophotometer at λ_{max} 265 and 392 nm of the dye used.

2.7. Statistical analyses

K/S values of each dyeing condition were evaluated by analysis of variance and each treatment was replicated three times. The standard error of the difference SED(±) was calculated for each mordant.

3. Results and Discussions

Through our innovative extraction process we have exemplified that the dye contents obtained by ethanolic extraction was higher than that obtained by aqueous extraction as shown in table-1. The ethanol after each extraction was recovered and has been reused for 3 consecutive times. For further use the ethanol had to be dried and distilled.

Table-1 Dye content from *Tagetus* flowers by different solvents

Part of the Plant from 50 gm	Dye content by Aqueous method	Dye content by Ethanolic method	Color of the Aq. extract	Color of the Eth. extract
***Tagetus* flower**	a. 2.40 gm b. 2.37 gm c. 2.38 gm	a. 3.55 gm b. 3.45 gm c. 3.48 gm	Yellow	Dark yellow

Through the different extraction processes we have also shown that the process is ecofriendly, since the ethanol is completely recovered in rotatory evaporator and reused. Dyeing shows deeper shades: Aqueous dyeing of cotton fabric with ethanolic extracted dye showed better color strength, CIELab and K/S values as compared to dyeing done by convention method of aqueous extraction and dyeing as shown in table-2 and figure-3. We have shown repeatability of the process and the consistency in the color content and is therefore recommended for Industrial application.

Table-2 Comparison of L, a, b and K/S values of cotton dyed by differently extracted dye

Serial No.		Common Name of the Plant	L	a*	b*	K/S
1.a	*Tagetus* (EtOH)	Genda	67.17	10.78	39.44	141.73
1.b.	*Tagetus* (Aq)	Genda	52.69	4.62	27.55	77.59

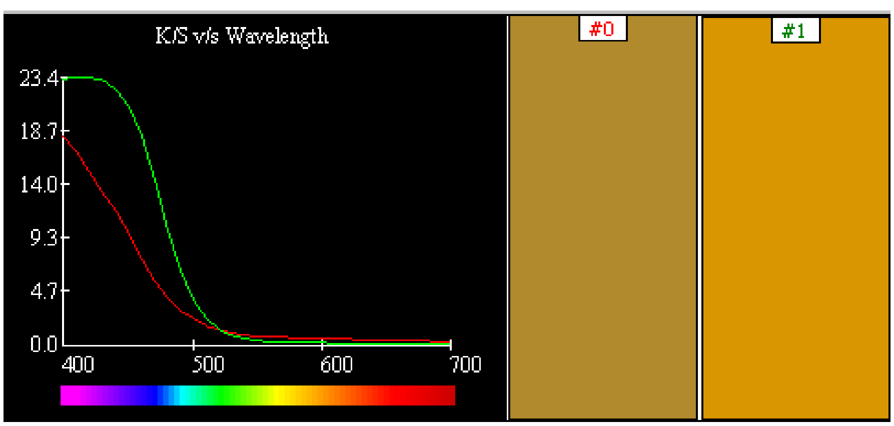


Figure- 3 *Tagetus* extract # 0 Aq extract dyed and # 1 Ethanol extract dyed cotton

The role of the metal mordanting in natural dyeing has been well demonstrated [16,17] The mordant forms an insoluble complex between dye molecule and metal, which is responsible for its insolubility and resultant affinity to the fiber, mainly silk and wool [18].

In our case also we have observed that *Tagetus* extract showed very good affinity for cellulosic as well as proteinaseous fibers when used in conjunction with metal mordants. The hue colors developed through metal mordanting were quite dark and saturated and fitted very well into the basic criteria of good natural dye formulation.

3.1. Proposed Mordant-dye Complex

Most of the natural dye extract have poor affinity for cotton fibers, their fastness is often enhanced by metal mordants, which form an insoluble complex with dye molecules. The *tagetus* dye extract also showed less affinity for cotton fibers as compared to wool and silk. The fastness was enhanced by using metal mordants, which formed an insoluble complex with dye molecules, a representative metal salt-M has been shown in figure 4.

The step wise dyeing of cotton fabric with tannic acid pretreatment, metal mordanting and dyeing with *Tagetus* flower extract, showed very good results of even dyeing. It has been proposed that the probable method of chelation of the dye molecules was occurring through the formation of bonds with metal ions. The dihydroxy flavone structure in one of the dye molecule –Patuletin has been shown to chelate with the metal mordant. This is a plausible explanation for chelation with one such flavonoid molecule. However *Tagetus* flower extract should not be considered as a dye molecule rather it should be attributed to a group of molecules which are cumulatively responsible for the dyeing process and color content.

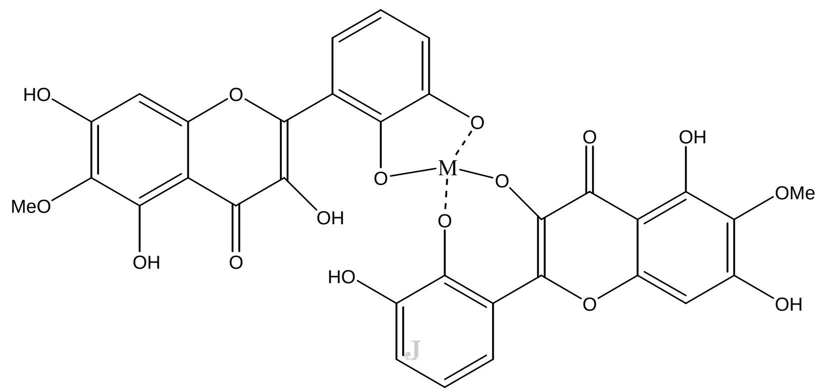


Figure-4 Proposed Dye-mordant complex

3.2. Spectral analysis of the dye and the isolated compounds 2a and 2b

UV-Visible spectrum of the yellow compound separated from *Tagetus* flower extract shows peak at 265,353 and 392 nm (0.72, 0.50 and 0.55 A respectively). The extract from *Tagetus* flower showed the following peaks in FT-IR spectrum:

Compound 2a:
^{1}H-NMR □(400 MHz, $CDCl_3$): 0.849, 0.998 (6H, s, 1'-gem-Me), 1.074 (6H, s, 1-gem-Me), 1.37 (1H, dd, J 13, 7, 2'ax-H), 1.48 (1H, t, J 12, 2ax-H), 1.626 (3H, s, 5'-Me), 1.739 (3H, s, 5-Me), 1.84 (1H, dd, J 13, 6, 2'eq-H), 1.912 (3H, s, 9'-Me), 1.970 (9H, s, 9-, 13-, 13'-Me), 2.04 (1H, dd, J 17, 10, 4ax-H), ca. 2.33-2.45 (2H, m, 6'-, 4eq-H), ca. 4.0 (1H, m, 3-H), 4.25 (1H, 3'-H), 5.43 (1H, dd, J 15.5, 10, 7'-H), 5.55 (1H, s, 4'-H), ca. 6.12 (2H, s, 7-, 8-H), ca. 6.15 (3H, m, 8'-, 10-, 10'-H), ca. 6.26 (2H, m, 14-, 14'-H), 6.36 (2H, d, J 15, 12-, 12'-H), ca. 6.55-6.71 (4H, m, 11-, 11'-, 15-, 15'-H). Major Mass fragments (m/z) 568, 551, 533,476,459 and 429 amu

Compound 2b:
^{1}H-NMR δ (400 MHz, $CDCl_3$): 3.91(OCH_3 -6), 6.91(H-8), 7.81(d, H-2'), 6.89(d, H-5'), 7.71 (dd, H-6') were some of the peaks identified.
IR: 3402, 2922, 2852, 1731, 1461, 1376, 1263, 1024, 739, 589, 530, and 422 cm^{-1}.
Mass fragments (m/z): 332, 314, 289 amu.

3.3. Color measurements K/S was measured for cotton, silk fabrics and wool yarn and CIElab values are shown in Tables 3-5.

Table-3 L*, a*, b* , C and H values for Cotton Fabric dyed with *Tagetus erecta*

Method	Mordant	L*	a*	b*	C	δE	K/S
	Controlled (Dye)	56.62	-0.88	44.06	44.07	--	72.64
	Controlled(TA+Dye)	55.97	-2.07	42.30	42.35	2.23	74.86
Pre-mordanting	Alum	59.45	5.56	51.79	52.09	10.44	69.98
	Copper sulphate	52.16	6.44	39.43	39.95	9.49	131.65
	Stannous chloride	62.84	11.09	58.50	59.54	19.75	136.70
	Ferrous sulphate	41.68	2.09	15.25	15.39	32.59	187.77
	Pot. dichromate	52.45	14.64	40.47	43.04	16.47	138.84
	Stannic Chloride	63.56	4.01	56.87	57.01	15.36	86.01

Table 3 shows the colorimetric values of dyed cotton fabric with *Tagetus* after pretreatment with different metal mordants, the dyeings with different mordants imparted a shade change from yellow-yellowish green-brown. Tannic acid pretreatment prior to mordanting enhanced the formation of dye-mordant complex in the case of cotton fabrics. Different mordants were used in 1-2 % keeping in mind the toxicity factor of some mordants. Varied hues of color were obtained from premordanting the cotton with $FeSO_4$, $SnCl_2$, $CuSO_4$, $SnCl_4$, $K_2Cr_2O_7$ and alum were dyed by aqueous extract of *tagetus* flower as shown in the figure-5, the different mordants not only cause difference in hue color and significant changes in K/S values but also L* values and brightness index values. The best values were obtained with ferrous sulphate K/S were measured for cotton, silk fabrics and wool yarn as shown in figures-5,6 and 7 and CIElab values are shown in table 3-5.

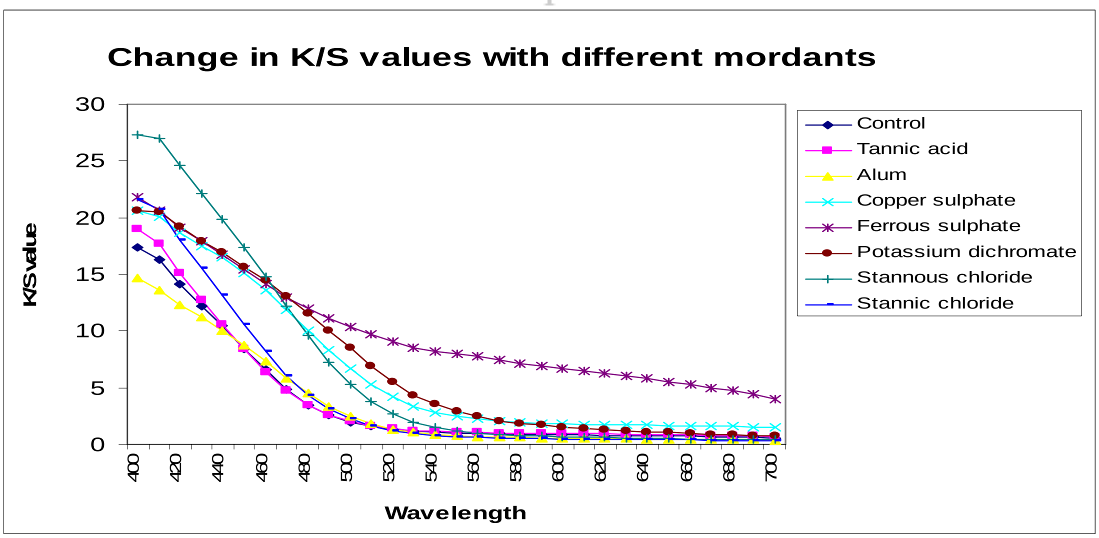


Figure-5 Change in K/S values with different mordants for cotton fabrics

Table- 4 L*, a*, b* , C and H values for Silk Fabric dyed with *Tagetus erecta*

Method	Mordant	L*	a*	b*	C	δE	K/S
	Controlled	57.35	-3.91	34.70	34.93	--	52.04
Pre-mordanting	Alum	64.64	7.38	55.13	55.62	24.45	126.07
	Copper sulphate	56.38	2.59	37.56	37.65	7.18	72.44
	Stannous chloride	69.06	10.62	63.29	64.18	34.14	132.91
	Ferrous sulphate	45.64	3.77	13.10	13.63	25.74	271.63
	Pot. dichromate	55.23	9.81	36.42	37.72	14.00	53.34
	Stannic Chloride	64.99	-1.48	48.21	48.23	15.70	54.01

Table 4 shows the colorimetric values of dyed silk fabric with *Tegetus* after pretreatment with different metal mordants, the dyeings with different mordants imparted a shade change from pure yellow to yellowish brown. Different mordants are used in 1-2 % keeping in mind the toxicity factor of some mordants. Varied hues of color can be obtained from premordanting the silk with $FeSO_4$, $SnCl_2$, $CuSO_4$, $SnCl_4$, $K_2Cr_2O_7$ and alum were dyed by aqueous extract of *tagetus* flower as shown in the figure-6, the different mordants not only cause difference in hue color and significant changes in K/S values but also L* values and brightness index values. The best values are obtained with ferrous sulphate.

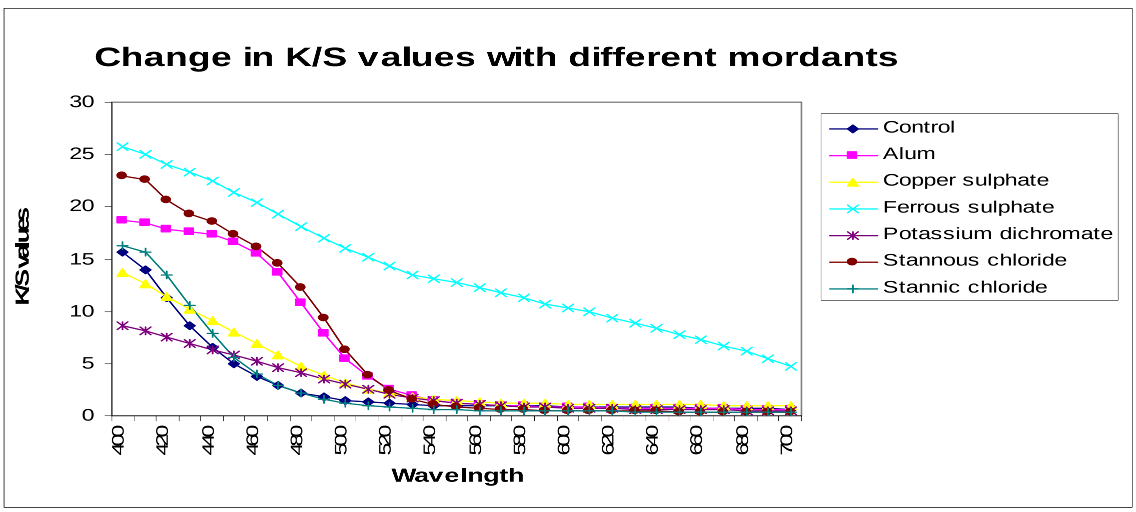


Figure-6 Change in K/S values with different mordants for silk fabrics

Table-5 L*, a*, b* , C and H values for Wool Yarn dyed with *Tagetus erecta*

Method	Mordant	L*	a*	b*	C	δE	K/S
	Controlled	52.14	-2.00	43.34	43.38	--	132.50
Pre-mordanting	Alum	58.03	10.80	60.83	61.78	22.47	84.88
	Copper sulphate	37.18	2.03	23.82	23.90	24.92	67.08
	Stannous chloride	59.31	12.80	65.44	66.68	27.55	102.12
	Ferrous sulphate	32.85	2.45	13.46	13.68	35.84	256.54
	Pot. dichromate	36.90	5.55	22.96	23.62	26.54	81.18
	Stannic Chloride	67.98	13.50	80.56	81.69	43.33	234.06

Table 5 shows the colorimetric values of dyed wool yarn with *Tagetus* after pretreatment with different metal mordants, the dyeings with different mordants imparted a shade change from light yellow to brown. Varied hues of color can be obtained from premordanting the wool yarn with $FeSO_4$, $SnCl_2$, $CuSO_4$, $SnCl_4$, $K_2Cr_2O_7$ and alum were dyed by aqueous extract of *tagetus* flower as shown in the figure-7, the different mordants not only cause difference in hue color and significant changes in K/S values but also L* values and brightness index values. The best values are obtained with ferrous sulphate. Thus in all the three cases iron provided dark shades.

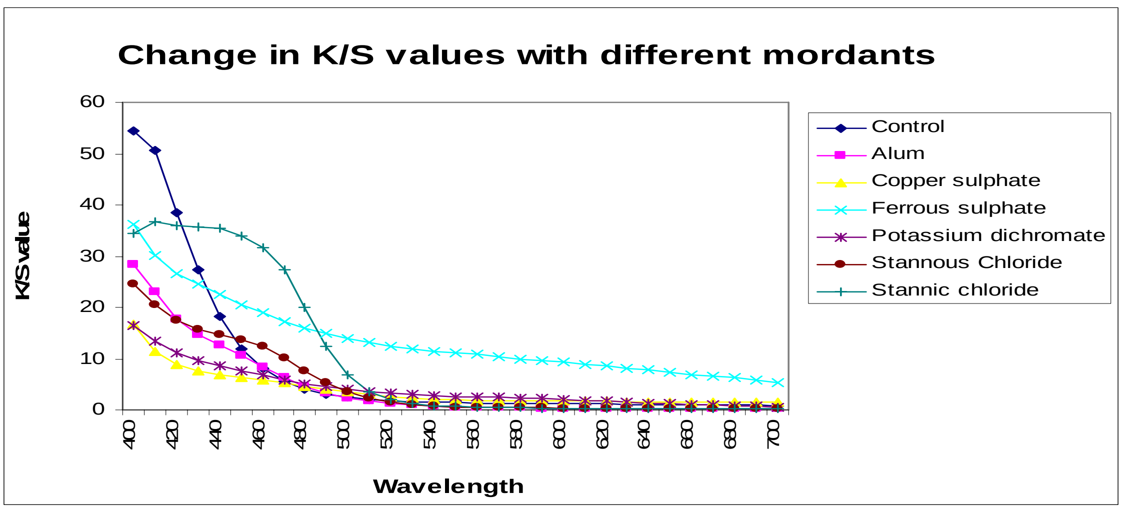


Figure-7 Change in K/S values with different mordants for wool yarn

3.4. Effect of mordanting conditions

It was observed that the pre mordanting technique with metal mordants imparted good fastness properties to the cotton, wool and silk fabric. Controlled samples without mordant were also prepared for comparison. Therefore, in pre mordanting technique, the dyed fabrics were mordanted with stannic chloride, stannous chloride, ferrous sulphate, copper sulphate, potassium dichromate and alum. The mordant activity of the six cases followed the sequences.

The order of K/S values is as following: Fe → Cr → Sn(II) → Cu → Sn(IV) → Al in cotton for *Tagetus erecta,* the absorption of color by cotton fabric was enhanced when metal mordants were used. Fe (II) provides best chelation as shown in table-3, figure-8.

The order of K/S values is as following: Fe → Sn(II) → Al → Cu → Sn(IV) → Cr in silk for *Tagetus erecta,* the absorption of color by silk fabric was enhanced when metal mordants were used. Fe(II) provides best chelation as shown in table-4, figure-9.

The order of K/S values is as following: Fe → Sn(IV) → Sn(II) → Al → Cr→ Cu in wool for *Tagetus erecta,* the absorption of color by wool yarn was enhanced when metal mordants were used. Fe (II) provides best chelation as shown in table-5, figure-10.

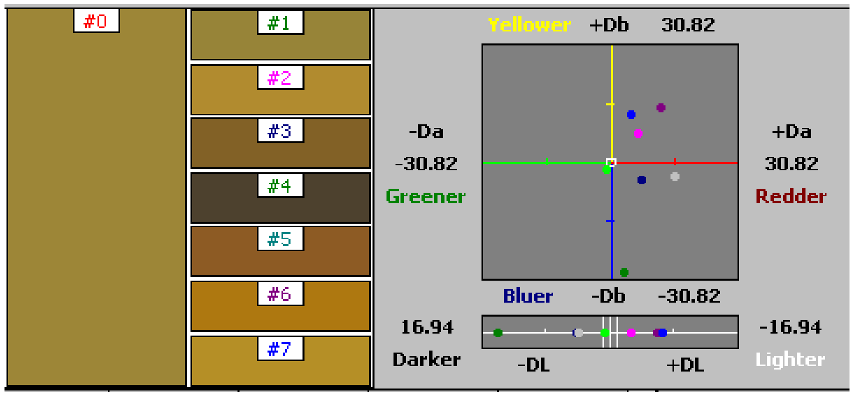


Figure-8 shows the colorimetric values of dyed cotton fabric with *Tagetus*

Figure-8 shows the colorimetric values of dyed cotton fabric with *tagetus* after pretreatment with different metal mordants(#0-7 in the order of unmordanted, tannic acid, alum, copper sulphate, ferrous sulphate, potassium dichromate, stannous chloride and stannic chloride). The dyeings with different mordants imparted a shade change from Khaki to light brown to dark brown having blackish tinge.The lightness value decreased for iron, chromium and stannous mordanted cotton sample and shade of depth retained their brightness, while the highest was obtained with stannic chloride having dullness.

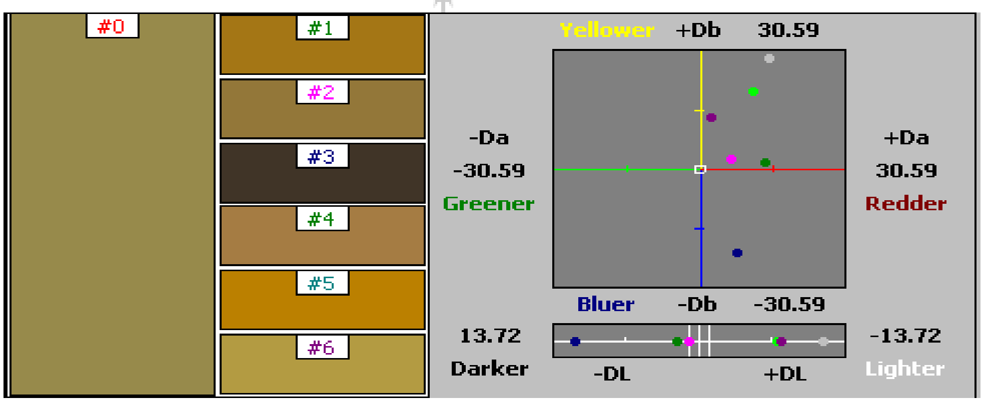


Figure-9 shows the colorimetric values of dyed silk fabric with *Tagetus*

Figure-9 shows the colorimetric values of dyed silk fabric with *tagetus* after pretreatment with different metal mordants (#0-6 in the order of unmordanted, alum, copper sulphate, ferrous sulphate, potassium dichromate, stannous chloride and stannic chloride). The dyeings with different mordants imparted a shade change from dull yellow dark mustard, brown to dark brownish black. The lightness value decreased for iron, stannous and alum mordanted silk samples and shade of depth became dull and dark, while the highest was obtained with stannic chloride and potassium dichromate having good brightness.

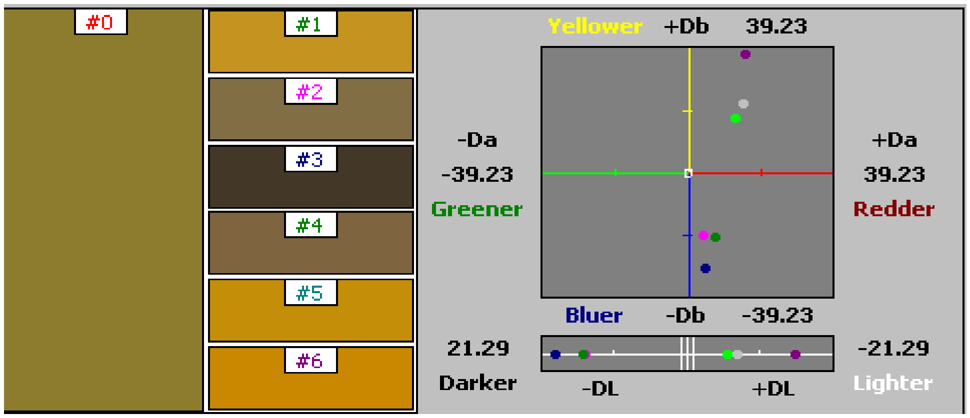


Figure-10 shows the colorimetric values of dyed wool yarn with *Tagetus*

Figure-10 shows the colorimetric values of dyed wool yarn with *tagetus* after pretreatment with different metal mordants (#0-6 in the order of unmordanted, alum, copper sulphate, ferrous sulphate, potassium dichromate, stannous chloride and stannic chloride). The dyeings with different mordants imparted a shade change from mustard yellow to moss green, brown and black. The lightness value decreased for iron, copper and chromium mordanted wool samples and shade of depth became dull and dark, while the highest was obtained with stannic chloride having good brightness. The shades of the dyed wool samples were the brightest among cotton, silk and wool samples

3.5. Fastness testing The dyed samples were tested according to Indian Standard methods and the results for alum, ferrous sulphate and copper sulphate mordanted have been tabulated below in Table 6. It shows that these mordants have caused improved fastness properties in all the three types of materials. Similar observations were made in the case of other three mordants as well. Marked improvement was noticed in the case of washing and light fastness. Thus the dye can be recommended for commercial use.

Table-6 Fastness properties of dyed cotton, silk fabrics and wool yarn under conventional heating with different metal modanting with *Tagetus erecta*

Dyeing methods	**Wash–perspiration–rubbing–light**					
	WF[a]	**Per$_{acidic}$**	**Per$_{basic}$**	**Rub$_{dry}$**	**Rub$_{wet}$**	**LF**[b]
Cotton(control)	2-3	2	2	2-3	2-3	2
Cotton(Alum)	4	4	3–4	3-4	3-4	4
Cotton(Fe SO_4)	4-5	4	4	4	4	4-5
Cotton(Cu SO_4)	4	4	4	4	4	4

Dyeing methods	Wash–perspiration–rubbing–light					
	WF[a]	Per_{acidic}	Per_{basic}	Rub_{dry}	Rub_{wet}	LF[b]
Silk(control)	2	2	2-3	2-3	2	2
Silk(Alum)	4	4	4	4	4	4
Silk(Fe SO_4)	5	4-5	4-5	4-5	4-5	4-5
Silk(Cu SO_4)	4-5	4	4	4	4	4-5
Wool (control)	3-4	3-4	3	3	3	3-4
Wool(Alum)	4	4	4	4	4	4
Wool(Fe SO_4)	5	4-5	4-5	4-5	4-5	5
Wool(Cu SO_4)	4-5	4	4	4	4	4-5

[a] wash fastness [b] light fastness.

Through the UV-Visible absorption of the dyebath before and after dyeing we have tried to show that dye uptake by mordanted fabric is better than the controlled samples. The enhancement in dye uptake with metal mordanting has been shown in table-7. Individual effect of metal mordanting, on cotton, silk and wool can be seen in table-7. The results clearly show marked enhancement as compared to the controlled samples. Overall enhancement of dye uptake due to metal mordanting has been found to be ranging from 45-52 % in the case of cotton, 38-46 % in silk and 37-51 % in wool with respect to the controlled samples.

Table 7 Dye exhaustion of *Tagetus* for cotton, silk and wool with various conditions

Metal mordant	cotton	silk	wool
Controlled	24%	28%	29%
Alum	45%	39%	37%
Ferrous sulphate	52%	46%	51%
Copper sulphate	35%	42%	47%
Stannous chloride	32%	38%	38%
Stannic chloride	38%	38%	39%
Pot. dichromate	48%	44%	48%

4. Conclusion

Tagetus erecta flower a temple waste has been shown to have good dyeing prospects through this study. It grows abundantly in India and Sri Lanka and can be explored for industrial natural dyeing. The ethanolic extraction followed by removal of ethanol and finally using the extract in aqueous medium has provided brighter shades as compared to direct aqueous extract. We have shown repeatability of the process and the consistency in the color content and therefore recommend for Industrial application. Metal mordant in conjunction with *Tagetus erecta* flower extract was found to enhance the dyeability and fastness properties effect in case all three types of material. Thus the net enhancement of dye uptake due to metal mordanting has been found to be ranging from 45-52 % in the case of cotton, 38-46 % in silk and 37-51 % in wool with respect to the controlled samples. The higher percentage of color strength in the case of silk and wool makes *Tagetus erecta* best suited for proteinaseous materials although cotton dyed samples have acceptable brightness and fastness properties. The dyeing process developed by us is for the ease of industrial application.

Acknowledgement

The author, S. Wijayapala, expresses her sincere thanks to the Indian Institute of Technology, Kanpur for the training provided.

References:

1. Vasudevan P, Kashyap S and Sharma S. Tagetes: A Multipurpose Plant, Bioresource Technology 1997; 62(1-2): 29-35.
2. Bosma TL, Dole JM and Maness NO. Crop Ecology, Management& Quality, Optimizing Marigold (*Tagetus erecta*.L.) Petal and Pigment Yield, Crop Science 2003; 43: 2118-24.
3. Mihalick J E and Donnelly K M. Using metals to change the colors of natural dyes, Journal of Chemical Education 2006; 83(10): 1550-51.
4. Pan N C, Chattopadhyay S N and Day A. Dyeing of jute with vegetable dyes, Journal of the Textile Association 2003; 63(5): 243 -45.
5. Pan N C, Chattopadhyay S N and Day A. Dyeing of jute fabric with natural dye extracted from marigold flower, Asian Textile Journal 2004; 13(7): 80-82.
6. Mahale G, Bhavani K, Sunanda R K and Sakshi. Standardising dyeing conditions for African marigold, Man-Made Textiles in India 1999; 42(11): 453-58.
7. Karolia A and Dilliwar S. Natural yellow dyes from marigold flowers for leather, Colourage 2004; 51(6): 31-2, 34-6,38.
8. Agarwal R, Pruthi N and Singh S S. Effect of mordants on printing with Marigold flowers dye, Natural Product Radiance 2007; 6: 306-09.
9. Li J. Method for preparing lutein crystal from dried marigold flower, Faming
10. Zhuanli Shenqing Gongkai Shuomingshu 2006; 9.
11. Guinot P, Gargadennec A, Valette G, Fruchier A and Andray C., 2008. Primary Flavonoids in Marigold Dye: Extraction, Structure and Involvement in the Dyeing Process, Phytochemical Analysis 2008; 19: 46-51.
12. Trotman, E.R. Dyeing and chemical technology of textile figures. 6th Ed. John Wiley and Sons, NewYork 1984.
13. Vankar PS, Tiwari V and Shanker R. Dyeing Cotton, Wool and Silk with Tectona leaves dye, Asian Textile Journal 2003; 86-89.
14. Indian Standards Institution (BIS), Handbook of Textile Testing, Manak Bhawan, p 539, 550, 553, 569, New Delhi, 1982.
15. Kubelka P. New Contributions to the Optics of Intensely Light-Scattering Materials. Part I, Journal of Optical Society of America 1948; 38 (5): 448 -51.
16. Kubelka P. New Contributions to the Optics of Intensely Light-Scattering Materials. Part II, Nonhomogeneous Layers, Journal of Optical Society of America 1948; 44 (4): 330-35.
17. Sarkar A and Seal C M. Color strength and color fastness of flax fabrics dyed with natural colorants, Clothing and Textiles Research Journal 2003; 21(4): 162-66.

18. Popoola AV. Dyeability of cellulosic fibers using dyestuff from African rosewood Pterocarpus erinaceous, Journal of Applied Polymer Science 2000; 77: 746-51.

19. Tiedemann E J and Yang Y. Fiber safe extraction of red mordant dyes from hair fibers, Journal of the American Institute of Conservation 1995; 34: 195-206.