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Dyeing Of Textiles With Natural Dyes - An Eco-Friendly Approach

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Abstract: Numerous plant species are found to have an important role in the day-to-day life of the ethnic and local people. However, it is a matter of concern that the indigenous knowledge of extraction, processing and practice of using of natural dyes has diminished to a great extent among the new generation of ethnic people due to easy availability of cheap synthetic dyes. Thus by keeping in view of above, the present study has been undertaken so as to revive the age-old are of dyeing with natural dyes. In the present work, the flowers of *Clitoria ternatea* (Clitoria flowers) and *Targetes erecta Linn* (Marigold) and *Punica granatum* (pomegranate) peel were used for the extraction of dye, dyeing of the selected fabrics at optimized conditions, using combination of mordants and evaluate the resultant colour fastness of the selected dyed samples to washing, rubbing, and light. Also the antimicrobial properties of the dyes were evaluated.
Key words: Natural dye, Mordants, colour fastness, eco-friendly dye.

Introduction

A renewed international interest has arisen in natural dyes due to increased awareness of the environmental and health hazards associated with the synthesis, processing and use of synthetic dyes. Textile processing industry is one of the major environmental polluters as the effluent from these industries contains a heavy load of chemicals including dyes used during textile processing. There are two main ways to limit the environmental impact of textile processing. One is to construct sufficiently large and highly effective effluent treatment plants, and the other way is to make use of dyes and chemicals that are environment friendly[1].

Natural dyes, when used by themselves have many limitations of fastness and brilliancy of shade. However, when used along with metallic mordants they produce bright and fast colours. Therefore, instead of using unsustainable technology for producing colours one can use mild chemistry to achieve almost similar results. The rich biodiversity of our country has provided us plenty of raw materials, yet sustainable linkage must be developed between cultivation, collection and their use[2]. Natural dyes can produce special aesthetic qualities, which, combined with the ethical significance of a product that is environmentally friendly, gives added value to textile production as craftwork and as an industry.

Thus by keeping in view of above, the present study has been undertaken so as to revive the age-old are of dyeing with natural dyes. In the present work, the flowers of *Clitoria ternatea* (Clitoria flowers) and *Targetes erecta Linn* (Marigold) and *Punica granatum* (pomegranate) peel were used for the extraction of dye, dyeing of the selected fabrics at optimized conditions, using combination of mordants and evaluate the resultant colour fastness of the dyed samples to washing, rubbing, and light. Also the antimicrobial properties of the dyes were evaluated.

Materials and methods

Source: *Punica granatum* (pomegranate), *Clitoria ternatea* (Clitoria flowers) and *Targetes erecta Linn* (Marigold) flowers. The dyeing of cotton/ synthetic fabric was carried out in three stages; Extraction of dyes from the plant sources, Mordanting and Dyeing.

Pre-Treatment of fabrics (Scouring)

Cotton and synthetic fabrics were washed in a solution containing 2 g/L commercial (Tide) detergent at 50º C for 25 min, keeping the material to liquid ratio at 1:40. The scoured material was thoroughly washed with tap water and dried. Pieces of 10 cm X10 cm were cut and used for the experiments[3].

Extraction of dye

Various experiments were conducted for the maximum extraction of natural dye from Pomegranate peels, Marigold flowers and Ciltoria flowers. The samples were collected and washed thoroughly with water to remove any dirt. After thorough drying at room temperature, the samples were ground into powder with the help of grinder and the powdered samples were used for the extraction of dyes. In order to find out the optimum extraction conditions, experiments were conducted in aqueous extraction at various range of pH (2-8) and temperature (28 $\pm$ **2** and 100 **°C**) with M:L ratio 1:10.

Mordanting

Two chemical mordant's namely ferrous sulphate and copper sulphate were used and lemon was used as a natural mordant at 2% concentration. Mordanting was carried out in three stages: Pre-mordanting, Simultaneous mordanting and Post-mordanting

Pre-mordanting

In this method the scored fabrics were first treated with mordant and then dyed using extracts for each plant separately. The fabrics were treated with each of the mordant mentioned above at the concentration of 1:20 M:L ratio for 30minutes at 28 $\pm$ 2 ^{0}C. Then the mordanted fabric was used for dyeing.

Simultaneous mordanting

In this method the fabrics were immersed in equal mixture of the mordant and the dye extract for 30 min at 28 $\pm$ ^{0}C followed by washing and drying of the dyed fabrics.

Post-mordanting

In case of post-mordanting, the dyed fabric was treated with mordants at 28 $\pm$ ^{0}C for 60 min with M:L ratio 1:20.

Dyeing

Experiments were performed in which dyeing was done at 28 $\pm$ ^{0}C and for 30 min.

Quality assurance tests of dyed fabric

Most dyes are organic compounds and are, therefore, vulnerable in varying degree to the action of destructive agents. A number of tests are necessary to cover all the important properties of any one dye because good fastness to one inference is not necessarily accompanied by equal fastness to other conditions. For characterization and evaluation, following tests were performed with selected dyed abrics: Washing fastness, Rubbing fastness and Light fastness.

Washing Fastness

Dyed sample was placed between two pieces of non dyed white samples (control). These three pieces were held together by stitching round the edges. The pre heated soap solution (Tide, at 60°C) in the ratio of 1:50 i.e 0.5g/25 mL water, was taken in a vessel added 1.0 g of sandwiched fabric for 30 minutes Then the specimen was removed and rinsed in cold water. The colour fastness is usually rated by the presence of the colour in control sample [4].

Rubbing fastness

The rub fastness of the dyed fabrics were carried out by rubbing the fabrics manually and checking for fading of color [5, 6, 7].

Light Fastness

The fabric was exposed to sun light for 24 h. The colour fastness to light was evaluated by comparison of colour change of the exposed portion to the unexposed original material [4].

Antimicrobial screening test

Susceptibility of the bacterial strains to the natural dyes was investigated using the disc diffusion method. Cultures used in the study were *E. coli* and *Klebsiella sp (*gram negative*)* and *S. aureus* and *Bacillus sp* (gram positive). LB Agar medium (15 mL) was poured into each sterile petri dish. Then the surface of the agar medium was inoculated with 100 µl cultures. Dyes were sterilized by filtration through 0.45 µm membrane filters. Sterilized discs of 5 mm were each impregnated with 1.0 mg of natural dyes. Discs were placed on agar plates, and the plates were incubated at 37°C for 24 h. The inhibition zones formed on the medium were evaluated in mm. All experiments were performed in duplicate[8].

In the next set of experiments the antimicrobial activity of dyed fabrics was tested. 1 g of test fabric (undyed fabric as control) was introduced into the 100 ml nutrient broth inoculated with the desired microbe and incubated at 37°C overnight for 24 h. Then the biomass of the culture was separated by centrifugation and weighed. The reduction in the amount of the biomass between the control and the test sample was calculated which is an index of the antimicrobial activity of dyed fabrics [9].

Results and discussion

***Punica granatum* (Fig 1)** is from the family Punicacea. It grows in all warm countries of the world and was originally a native of Persia. The rind of pomegranate contains a considerable amount of tannin, about 19% with pelletierine The main coloring agent in the pomegranate peel is granatonine which is present in the alkaloid form N-methyl granatonine [10].

Butterfly pea (*Clitoria ternatea*) (Fig 1) is a perennial creeper plant to the family Fabaceae. The flowers are available almost throughout the season. The most striking characteristic about this plant are its deep blue flowers. Major flavonol glycosides, 3-O-(2"-O-alpha-rhamnosyl-6"-O-malonyl-betamalonyl)-beta-glucoside and 3-O-(2",6"-di-O-alpha- rhamnosyl)-beta-glucosid were isolated from the blue- flowered petals [11].

Marigold (*Tagetus* *erecta*) (Fig 1) belongs to the family Asteraceae. The principle colouring component of marigold flower is lutein, a fat-soluble carotenoid**,** which is responsible for the yellow to orange colour of the dye[12].

Extraction of dye

The yield of the dye per 100 g of the plant specimen obtained under various extraction conditions are summarised in the Table 1.Yield of the dye can be improved by using techniques like rotary evaporator for concentration of the dye. The amount of dye extracted from rind of pomegranate was 22.5% by Goodarzian & Ekrami [10] and 19.2% by Kulkarni *et al* [13]. Extraction of dye from butterfly pea employing aqueous extraction method for 3 h resulted in 4.605 mg L^{-1} by Sinha *et al* [14]. Extraction of dye from Marigold flower employing aqueous/organic solvent extraction method for 3 h/ 1.5 h resulted in 2.4 and 3.55 g per 100 g of the dry flower respectively was shown by Padma *et al* [15]. From Table 1, it was observed that the extraction of dye was a function of pH. The organic solvent extraction resulted in poor yielding not in detectable range, the reason is yet to be analysed.

In acidic pH solution, the colour of butterfly pea extract displayed a red colour; but in alkaline pH, the colour changed to greenish blue. The change in colour and extraction rate of butterfly pea solution depends on the change in equilibrium of four anthocyanin species in its petals according to the prevailing pH. At lower pH, the red colour signified the presence of anthocyanins and in increasing pH, the colour intensity transformed to blue colour due to the presence of quinonoidal base and the yellow colour was for the chalcone[16].

Tagetes flower **Pomegranate peel** **Clitoria flower**

Fig 1 Plant sources

Table 1 Yield of the dye obtained under various extraction conditions

Dye	**Extraction Medium**	**Temp (± 2)**	**Time**	**pH Yield of dye (mg/100 g of specimen)**				
Clitoria				**2**	**4**	**6**	**7**	**8**
		28^{0}C	24 h	200	**300**	260	230	230
	Aqueous	**100 ^{0}C**	60 min	30	70	40	40	20
Marigold		**28^{0}C**	24 h	150	300	400	**580**	16
		100 ^{0}C	60 min	100	130	140	150	140
Pomegranate peel		**28^{0}C**	24 h	150	200	270	330	**520**
		100 ^{0}C	60 min	50	70	80	80	70

Mordanting and Dyeing

Mordants play very important role in imparting color to the fabric. The mordants used in combination in different ratios gave varying shades. Better colour strength results are dependent on the metal salt used[17]. Strong co-ordination tendency of Fe enhances the interaction between the fiber and the dye, resulting in high dye uptake[3]. Ferrous sulphate and Copper sulphate have the ability of forming coordination complexes (Co-ordination numbers are 6 and 4 respectively).

Functional groups such as amino and carboxylic acid on the fiber can occupy the unoccupied sites on interaction with the fiber. Thus, a ternary complex is formed by the metal salt on which one site is with the fiber and the other site is with the dye. The mordanted cotton cloth was immediately used for dyeing because some mordants are light sensitive[3,13]. The chromatophore of the dye makes it resistant to photochemical attack, but the auxochrome may alter the fastness [3]. The resistance of a dye or pigment to chemical or photochemical attack is an inherent property of the dye chromophore. The results of the present experiments on dying with Marigold, Clitoria and Pomogranate peel are shown in the Figures 2, 3 and 4 respectively.

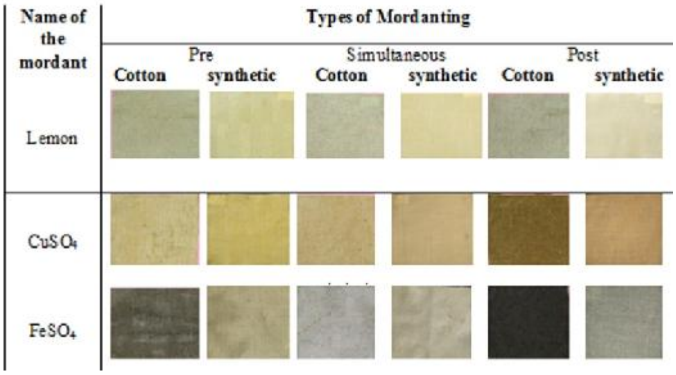

Fig 2 Dying with Marigold flower extract

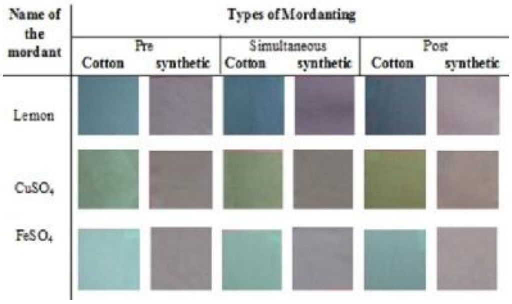

Fig 3 Dying with Clitoria flower extract

Fig 4. Dying with Pomegranate peel Extract

Quality assurance tests of dyed fabric

Fastness properties of the fabric dyed with extract from marigold flower were reported in this work.

i) Light Fastness

Good light fastness was observed in fabrics dyed with the dye extracted from marigold flower (Fig 5). This is due to the formation of complex with the metal which protects the chromatophore from photolytic degradation. The substitution pattern of dyes seems to play an important role in determining their light fastness. A particular substituent may increase the electron density around the reaction site of the molecule facilitating oxidation, or it may reduce the electron density with a resultant increase in case of reduction. It can be seen that the structure of Marigold flower having two hydroxyl groups, gives good fastness.

ii) Washing and Rubbing Fastness

Wash fastness of the dye is influenced by the rate of diffusion of the dye and state of the dye inside the fiber[3]. The dye extracted from marigold exhibits good to excellent washing fastness (Fig 6). Complexing with mordant has the effect of insolubilizing the dye, making it colour fast. A particular substituent may increase the electron density around the reaction site of the molecule facilitating oxidation, or it may reduce the electron density with a resultant increase in case of reduction. Samples dyed with marigold extract by using copper sulphate as a mordant have an excellent light fastness followed by ferrous sulphate. Good rub fastness was exhibited by the fibers dyed using the dye extracted from the marigold flower. Complexing the fiber with mordant, has the effect of insolubilizing the dye, making it color fast. The fabrics dyed with marigold flower exhibit good fastness properties[5].

The whole process of extraction and dyeing is ecologically safe. The obtained results have shown the dyeing potential of all three natural plant sources as source for dyeing. There is need for proper knowledge, documentation and assessment of dye- yielding plants as well as the dying techniques so as to increase the use of natural dyes [13].

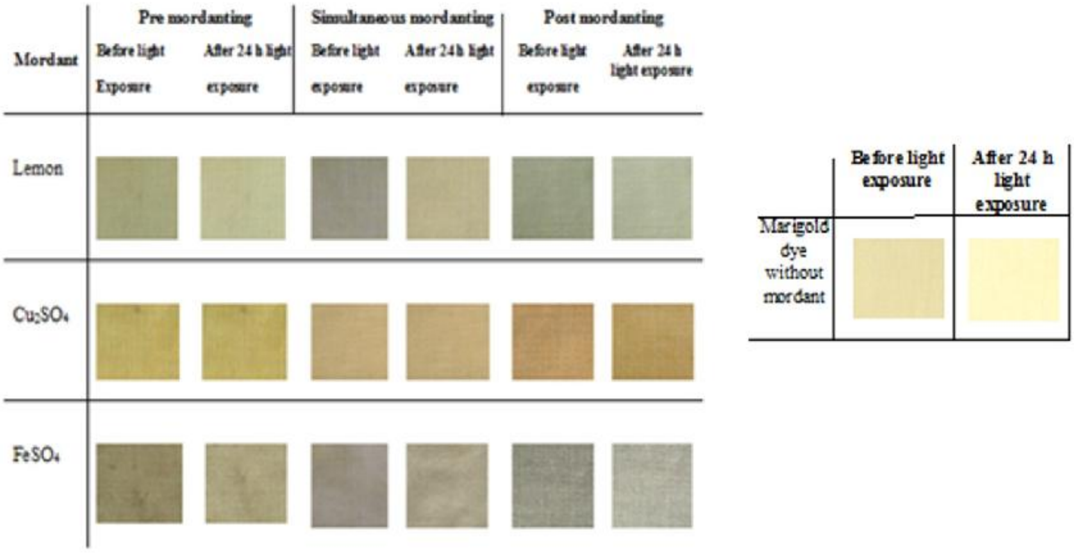

Fig 5 Light fastness - Marigold dye

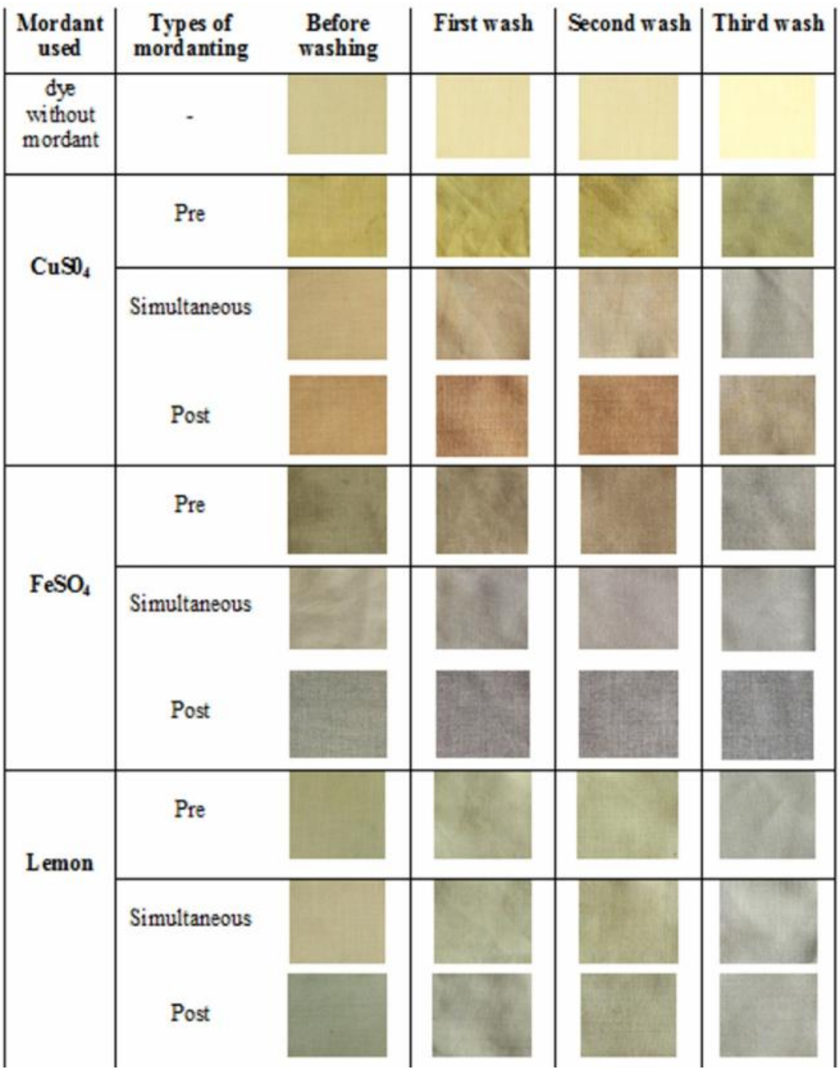

Fig 6 Wash fastness and Rubbing fastness - Marigold dye

Antibacterial property of the dyes

Many of the plants used for dye extractions are classified as medicinal, and some of these have been shown to posses' significant antimicrobial effect. The antimicrobial activities of some of these dyes are reported as potent owing to the existence of phenol, tannin and quinone in their extracts. The antimicrobial effects of some plants used in dye industries contribute to the longer life of the products [17].

Three natural dyes were screened for their antimicrobial activity against four test bacteria *E. coli* and *Klebsiella sp (*gram negative*)* and *S. aureus* and *Bacillus sp* (gram positive) and the screening showed that *P. granatum* was effective against all the test bacteria followed by Clitoria and Marigold (Table 2).

Similar work on antimicrobial activity of natural dyes were done by Singh *et al* [14] & Gupta *et al* [18]. Table 2 has also indicated that natural dyes had significant antibacterial activities towards the gram-positive bacteria and had less inhibition effect on the gram-negative test bacteria (*E. Coli* and *Klebsiella sp*.). The cell wall structure of the gram-negative bacteria is constructed essentially with LPS that avoids the accumulation of the antimicrobial agents on the cell membrane [19]. The above reports have confirmed our results.

Antimicrobial activity of natural dyes on substrate (fabric)

Different test fabrics showed different in reduction of test bacterial growth ranging from 22 – 57% as seen **in Table3.** Han & Yang [9] also observed an inhibition rate of 70% against *S. aureus* when 0.01% of curcumin was applied to the fabric and also 70% inhibition rate against *E. coli* with 0.05% of curcumin. The results from these experiments indicated that these natural dyes had antimicrobial activity both on solutions and substrate. However, further research is needed to determine the effect of dye structure on inhibition.

Table 2 Antimicrobial activity of natural dyes against test bacteria

Dye sample	**Diameter of zone of inhibition (mm)**			
	E.coli	***S. aureus***	***Klebsiella sp***	***Bacillus sp***
Pomegranate peel	8	10	8	12
Clitoria	8	9	7	10
Marigold	6	9	6	9

Table 3. Antimicrobial activity (% reduction) of textile materials dyed with natural dyes on test bacterial growth

Dye Sample	**Test fabric (1 g)**	**Test bacteria**	
		E.coli	*Staphylococcus aureus*
		(% Reduction in Biomass)	
Marigold	Cotton	44	50
	Synthetic	25	22
Pomegranate peel	Cotton	44	36
	Synthetic	56	36
Clitoria	Cotton	37	07
	Synthetic	50	57

Conclusion

The present scenario is focused more towards the utilization of the vast diversity of natural resources of colour pigments for their use in food materials, pharmaceuticals and textiles, in place of their synthetic counterparts. This trend is aimed at safeguarding human health as well as protecting and prolonging life on earth. Detailed scientific studies with natural dyes have established that in most cases their properties are comparable to those of synthetic dyes.

Therefore, if natural dyes have to be commercialized, they need to conform to the same stringent standards of performance that are applied to synthetic dyes. It thus follows that much more research and developmental effort needs to go in this area. The traditional practices may have to be substituted by modern, more scientific practices in order to overcome some of the so-called disadvantages of this dye [3].

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