Performance Evaluation and Statistical Analysis of Color Parameters for the Modified and Dyed Cotton Fabric

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Abstract: In this study, the reactive composite (N-methylolated-PAam-GG) onto cotton fabrics in the presence of a catalyst (i.e., ammonium sulfate and ammonium chloride) had been prepared, and it was fixed with various conditions of time and temperature, and then dyeing these modified cotton fabrics with the madder as a natural dye. Spectral reflectance, color coordinates, and color differences were measured. Then, the Kubelka-Munk equation was used to determine and evaluate the fixation onto dyed modified cotton fabrics and then study the statistically analyzed results using one-way ANOVA. The existing relationships had been detected with the change of both times (3 and 5 min.), temperature (120, 140 and 160°C) with two catalysts used (i.e., ammonium sulfate and ammonium chloride) by the conventional method for the assessment of natural dye onto the modified cotton fabrics if a known concentration of the same dye, is available for comparison. We found that the modified cotton fabrics were fixed with ammonium sulfate at 140°C for 5 min. exhibited a higher color fixed with the madder as a natural dye in all color parameters, color coordinates, and color difference.

Keywords: Kubulka-Munk; color coordinates; natural dye; Guar gum; paaam-GG composite; CIE.

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1. Introduction

The light absorption of a solute in solution obeys the Beer-Lambert law. That is, the absorbance of the solution at a given wavelength has a constant, linear functional relationship to the concentration of the absorbing species. Beer Lambert’s law is not applicable in opaque media, and the Kubelka-Munk theory has been widely applied to describe the depth of color of a dye on a textile substrate. This has proven to be a useful tool for dye users and dye manufacturers, allowing for an objective assessment of the depth of a specific shade of dye on textile substrates [1]. This has significant commercial implications; the dye house manager must achieve a shade at the lowest cost of dye while meeting the required fastness criteria. Furthermore, at the maximum absorption wavelength, K/S values have been used to obtain a quantitative assessment of the amount of dye on the fabric [2].

Guar gum (GG), appears with chemically structured long straight chains of α-D mannopyranosyl units linked by (1-4) β-glycosidic linkages with the ratio 2:1. Commonly used natural guar gum because of its cost-effectiveness, thickening effect, nontoxic, biodegradability, biocompatibility, high viscosity, and water-solubility, and now used in many industries [3] such as sizing; finishing agents in the textile and paper industries as a binder stabilizer and thickener in the cosmetics; food industries and as a fracturing fluid additive in mining and hydraulic fracturing processes [4,5]. However, the substitution of GG is a difficult
task due to insolubility to offered dispersion without degradation, so with this aim, try to modify GG with reactive polyblends prepared below then dyed with natural dye (i.e., Indian madder), which is one of the prominent dye bearing plants belonging to the family Rubiaceae [6].

The main textile fiber is cotton, and it has special fiber properties, like high intensity, Durability, softness, strong biodegradability, and dyeability. It has been used in textiles and their Production for many centuries [7]. Color plays a major part in the use of fibers made of cotton. Researchers are studying various methods and technologies for the preparation of cotton fibers to enhance the ability of natural dyes to dye them. In several studies, ultrasound energy, enzymes, mineral mordants, and biomordants have been used to enhance the coloring potential of cotton fiber with various natural colorants[8-13]. To improve the affinity of cotton fiber for natural dyes, pretreatment of cotton with chitosan was used [14-17]. Anionic and cationic active compounds, cross-linking agents, and ozone treatment have been used as pretreatments in other studies to enhance the dyeing ability of cotton with natural dyes [18-26]. The use of radiation technologies such as plasma therapy, microwave, gamma, and UV radiation has been shown to improve the fatigue and fixation of natural cotton dyes [27-33].

2. Materials and Methods

2.1. Materials.

Mill scoured, bleached, and mercerized plain weave cotton fabric (100%) weight 120 gm/m², the thickness was 0.2 mm, the number of yarn/cm in warp direction 30 and weft 31 were supplied by El-Naser-Company for spinning weaving and dyeing. Mahalla El-Kubra Egypt. Guar gum (GG) was purchased from S.D. fine chemicals (Mumbai, India). Acrylamide, potassium bromates (KBrO₃), thiourea (TU), sulfuric acid, sodium hydroxide, triethanolamine (TEA), and formaldehyde (HCHO) were laboratory grade chemicals. Natural dye (Indian madder-Rubiacordifolia) was supplied by Misr scientific, Dokki-Giza, Egypt.

2.2. Synthesis and pretreatment of cotton fabrics.

Modified cotton fabrics [34] were prepared by immersing them in a solution containing (40% methylolation based on the weight content of reactive composite (N-methlylol -PAam-GG) in the presence of catalysts (0.5% ammonium sulfate or ammonium chloride), then all samples were padded followed by roasting for (3 and 5 minutes) at various temperature (120, 140, and 160 °C). At 90 °C, the treated samples were washed for 10 minutes in a solution containing 0.5gm/l nonionic detergent. Finally, the fabric was dried and prepared for dyeing with natural dye extracted from madder roots.

2.3. Dye extraction procedure.

Dried Indian madder (Rubiacordifolia) was used to obtain natural dye (as an anthraquinone dyeing compound), which was conventionally extracted from powdered (Rubiacordifolia) [35] that gives the highest yield, about (50 % based on fiber weight) of madder root powder, which was held overnight in aqueous solution for 12 hours, and then cooked in distilled water for 30 minutes at a liquor ratio of (1:4) with continues stirring The modified cotton fabric was then used for subsequent dyeing.
2.4. Optical density of dye extracted.

Shimadzu (VIS) Double Beam Spectrophotometer with standard illuminant C (1174.83) model V-530 and bandwidth 2.0 nm with precision ± 0.05 percent in the UV-visible range (250-700nm) was used to determine the optical density of the extracted solution after filtration by evaluating its optical density. With a UV spectrophotometer, we calculate the optical density of the collected solution at various concentrations. From (Figure 1), we observed that one peak was detected at an optical density of 2.586 at 530 nm. That means we are in the red color range for this natural dye used.

![Figure 1. Absorption spectrum of Madder (Natural dye).](image)

2.5. Dyeing with natural dye extracted from Madder root.

Modified cotton fabrics (MCF) were dyed using exhaustion Madder as a natural dye. The dye bath material to liquor ratio (M:L) was kept at 1:40, the temperature of the bath was raised to 100°C, and the dyeing operation was continued for 60 min. followed by washing with nonionic detergent; finally, all dyed samples were dried.

2.6. Color measurements.

The color strength [36] values and CIE. L, a, and b color parameters (L*(lightness), a*(red-green axis), b*(yellow-blue axis), c*(chroma), h*(hue angle)) were evaluated using Data color 650 Spectrophotometer with illuminant D65 and 10observer. The color intensity as K/S values for all dyed samples was calculated through Kubelka–Munk equation:

$$\frac{K}{S} = \frac{1 - R^2}{2R}$$

where K is the absorption coefficient, R is the reflectance of the dyed sample, and S is the scattering coefficient.

All measured samples showed a maximum absorption wavelength value (λ max at 480 nm). The color difference (ΔE*) is calculated by:

$$\Delta E^* = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2}$$

where ΔL* denotes the difference between lightness values, Δa* denotes the difference between red/green values, and Δb* denotes the difference between yellow/blue values of the batch and standard [37,38].

2.7. Statistical analysis.

The analysis of variance assumes equal variance across the samples. All data were subjected to Levene’s tests of homogeneity [39] of variance (α = 0.05). The equal variance
cannot be assumed for reflectance comparisons, while equal variances were found for \( L^* \), \( a^* \), \( b^* \), \( C^* \), and \( h \). All of the experiments were conducted in duplicate. All data analyzed and expressed as mean values ± 10 standard deviations. This was done to ensure the high precision of metrological measurements all over the work when using our calibrated instruments in our institute by either standard primary apparatus or certified reference materials used especially for this purpose, the uncertainty of the material variability was estimated by using one-way analysis of variance ANOVA [40-42].

3. Results and Discussion

3.1. Spectral reflectance.

The spectral reflectance curves (values of wavelength) and their corresponding 95% of confidence limits for all samples modified cotton fabric with (PAam-GG) which dyed with natural dye anthraquinone colorants extracted from roots of Indian madder with material-to-liquor (M:L) ratio of 1:20, The reflectance of modified and dyed cotton fabric is dependent on the wavelength of madder dye, the reflectance was low and increased across the entire wavelength range with a different catalyst, temperature and time.

![Figure 2. Spectral reflectance of the modified cotton fabric (PAam-GG) dyed with madder with (NH\(_4\))_2Cl catalyst with different temperatures (a) 3min. (b) 5 min.](https://nanobioletters.com/)

The spectral reflectance (%) is dependent on wavelength (\( \lambda \)) in the presence of two catalysts (ammonium chloride and ammonium sulfate) which roasting with various temperatures (120, 140 and 160°C) and time (3 and 5 min.) as shown in both figures (2) and (3). The reflectance was low and increased across the entire wavelength range in the short wavelengths. The spectral reflectance was very similar for all samples used, and also, we note an increasing moderately in the spectral reflectance (%) in the presence of ammonium chloride while in the presence of ammonium sulfate more increasing in the spectral reflectance (%); this is because the new bond formed between modified cotton fabric (PAam-GG) and madder as a natural dye.
3.2. Color coordinators.

The color differences for all samples dyed with natural dye in the presence of a catalyst (ammonium chloride or ammonium sulfate) at different temperatures (120, 140 and 160°C) and curing time from 3 to 5 minutes were calculated uncertainty values. The uncertainty value of the variability was estimated by using a one-way analysis of variance ANOVA [40-42]. The obtained values appear in (Tables 1 and 2).

Table 1. Values of ANOVA- one-way analysis for using (NH₄Cl) as a catalyst.

| Color parameter | Temp. | 120°C | 140°C | 160°C |
|-----------------|-------|-------|-------|-------|
| Time            | F-value | P-value | F-circuit | F-value | P-value | F-circuit | F-value | P-value | F-circuit |
| L* Blank        | 0.174  | 0.688  | 5.318  | 0.174  | 0.688  | 5.318  | 0.174  | 0.688  | 5.318  |
| 3min.           | 2.898  | 0.127  | 5.318  | 2.898  | 0.127  | 5.318  | 2.898  | 0.127  | 5.318  |
| 5min.           | 0.277  | 0.013  | 5.318  | 0.277  | 0.013  | 5.318  | 0.277  | 0.013  | 5.318  |
| a* Blank        | 0.091  | 0.771  | 5.318  | 0.091  | 0.771  | 5.318  | 0.091  | 0.771  | 5.318  |
| 3min.           | 0.333  | 0.580  | 5.318  | 0.333  | 0.580  | 5.318  | 0.333  | 0.580  | 5.318  |
| 5min.           | 0.032  | 0.862  | 5.318  | 0.032  | 0.862  | 5.318  | 0.032  | 0.862  | 5.318  |
| b* Blank        | 0.080  | 0.784  | 5.318  | 0.080  | 0.784  | 5.318  | 0.080  | 0.784  | 5.318  |
| 3min.           | 0.159  | 0.700  | 5.318  | 0.159  | 0.700  | 5.318  | 0.159  | 0.700  | 5.318  |
| 5min.           | 1.220  | 0.302  | 5.318  | 1.220  | 0.302  | 5.318  | 1.220  | 0.302  | 5.318  |
| C* Blank        | 0.122  | 0.736  | 5.318  | 0.122  | 0.736  | 5.318  | 0.122  | 0.736  | 5.318  |
| 3min.           | 0.893  | 0.372  | 5.318  | 0.893  | 0.372  | 5.318  | 0.893  | 0.372  | 5.318  |
| 5min.           | 0.373  | 0.558  | 5.318  | 0.373  | 0.558  | 5.318  | 0.373  | 0.558  | 5.318  |
| h* Blank        | 0.955  | 0.357  | 5.318  | 0.955  | 0.357  | 5.318  | 0.955  | 0.357  | 5.318  |
| 3min.           | 0.167  | 0.694  | 5.318  | 0.167  | 0.694  | 5.318  | 0.167  | 0.694  | 5.318  |
| 5min.           | 2.430  | 0.136  | 5.318  | 2.430  | 0.136  | 5.318  | 2.430  | 0.136  | 5.318  |

The data from Tables 1 and 2 show that the F-value < F-crit. and P-value > 0.05, so we conclude that all samples were homogeneous. Where: P-value is the probability of observing test statistic value, F is Fisher Snedecor distribution, and F critical is a function of the degrees of freedom of the numerator and the de-numerator and significance level [43,44].

Table 2. Values of ANOVA- one-way analysis for using (NH₄SO₄) as a catalyst.

| Color parameter | Temp. | 120°C | 140°C | 160°C |
|-----------------|-------|-------|-------|-------|
| Time            | F-value | P-value | F-circuit | F-value | P-value | F-circuit | F-value | P-value | F-circuit |
| L* Blank        | 0.174  | 0.688  | 5.318  | 0.174  | 0.688  | 5.318  | 0.174  | 0.688  | 5.318  |
| 3min.           | 0.962  | 0.355  | 5.318  | 0.962  | 0.355  | 5.318  | 0.962  | 0.355  | 5.318  |

The data from Tables 1 and 2 show that the F-value < F-crit. and P-value > 0.05, so we conclude that all samples were homogeneous. Where: P-value is the probability of observing test statistic value, F is Fisher Snedecor distribution, and F critical is a function of the degrees of freedom of the numerator and the de-numerator and significance level [43,44].
The presence of two catalysts [(NH₄)4SO₄ and NH₄Cl] then roasting with various time and temperature then dyed with madder as a natural dye, as shown in (Table 3,4) we note an increase in all parameters with catalyst (NH₄)4SO₄ and NH₄Cl then roasting with various time and temperature then dyed with madder as a natural dye, with temperature increase from 120 to140°C and then decrease slowly or slightly stable to 160°C, and also, L* values also increase from 3 to 5 minutes. Similar actions for all CIE values (a*,b*, c* and h* values). The highest C* and the lowest value of h* suggest that dye absorption was excellent with samples of high saturation and yellowness, respectively.

| Color parameter | Temp. | 120°C | 140°C | 160°C |
|-----------------|-------|-------|-------|-------|
|                 | Time  | F-value | P-value | F-circuit | F-value | P-value | F-circuit | F-value | P-value | F-circuit |
| a*              | 5 min | 1.000   | 0.347  | 5.318   | 0.533   | 0.486   | 5.318   | 0.110   | 0.749   | 5.318   |
|                 |       | 0.091   | 0.771  | 5.318   |         |         |         |         |         |         |
|                 | 3 min | 0.171   | 0.690  | 5.318   | 0.019   | 0.895   | 5.318   | 1.085   | 0.328   | 5.318   |
|                 | 5 min | 1.280   | 0.291  | 5.318   | 2.133   | 0.182   | 5.318   | 0.536   | 0.485   | 5.318   |
| b*              | 5 min | 0.080   | 0.784  | 5.318   |         |         |         |         |         |         |
|                 | 3 min | 0.356   | 0.578  | 5.318   | 0.012   | 0.917   | 5.318   | 0.022   | 0.887   | 5.318   |
|                 | 5 min | 0.222   | 0.886  | 5.318   | 0.202   | 0.665   | 5.318   | 0.327   | 0.583   | 5.318   |
| C*              | 5 min | 0.159   | 0.701  | 5.318   | 2.250   | 0.172   | 5.318   | 0.425   | 0.533   | 5.318   |
|                 | 3 min | 0.008   | 0.931  | 5.318   | 1.618   | 0.239   | 5.318   | 1.199   | 0.305   | 5.318   |
| h*              | 3 min | 1.589   | 0.243  | 5.318   | 1.017   | 0.343   | 5.318   | 0.533   | 0.486   | 5.318   |
|                 | 5 min | 1.017   | 0.343  | 5.318   | 0.303   | 0.597   | 5.318   | 1.142   | 0.316   | 5.318   |

The uncertainty value of color parameters (L* =57.18 ± 0.047,a*=20.52 ± 0.048,b*= 8.39 ± 0.049, c* = 22.17 ±0.047 and h* = 2.31 ± 0.047) for the unmodified cotton fabric dyed with madder (as a natural dye). The uncertainty value of color parameters for all modified cotton fabric with (PAam-GG) composite in the presence of two catalysts [(NH₄)2SO₄ and NH₄Cl] then roasting with various time and temperature then dyed with madder as a natural dye, as shown in (Table 3,4) we note an increase in the uncertainty value of L* values increase with temperature increase from 120 to140°C and then decrease slowly or slightly stable to 160°C, and also, L* values also increase from 3 to 5 minutes. Similar actions for all CIE values (a*,b*, c* and h* values). The highest C* and the lowest value of h* suggest that dye absorption was excellent with samples of high saturation and yellowness, respectively.

| Temp. | Time  | L*      | a*      | b*      | C*      | h*      |
|-------|-------|---------|---------|---------|---------|---------|
| 120°C | Blank | 57.18 ± 0.047 | 20.52 ± 0.048 | 8.39 ± 0.049 | 22.17 ± 0.047 | 2.31 ± 0.047 |
|       | 3 min | 57.30 ± 0.050 | 20.61 ± 0.047 | 13.90 ± 0.047 | 24.86 ± 0.048 | 1.25 ± 0.047 |
|       | 5 min | 59.54 ± 0.048 | 23.74 ± 0.048 | 16.52 ± 0.047 | 28.92 ± 0.060 | 1.19 ± 0.047 |
| 140°C | 3 min | 58.34 ± 0.047 | 20.85 ± 0.047 | 14.50 ± 0.047 | 25.40 ± 0.047 | 1.19 ± 0.047 |
|       | 5 min | 62.30 ± 0.048 | 23.89 ± 0.047 | 18.11 ± 0.047 | 29.98 ± 0.047 | 1.06 ± 0.047 |
| 160°C | 3 min | 58.20 ± 0.048 | 20.80 ± 0.053 | 14.40 ± 0.047 | 25.30 ± 0.047 | 1.21 ± 0.047 |
|       | 5 min | 62.27 ± 0.047 | 23.70 ± 0.047 | 18.00 ± 0.047 | 29.76 ± 0.047 | 1.05 ± 0.047 |

Color parameters for all samples with a different catalyst [(NH₄)2SO₄ and NH₄Cl] as shown in Table (3,4), we note an increase in all parameters with catalyst (NH₄)2SO₄ and NH₄Cl but the case of ammonium sulfate more than ammonium chloride because of new bond formed between natural dye (i.e., madder) and modified cotton fabric by PAam-GG. We note similar behavior in all terms of CIE (a*,b*, c*, and h* values). The optimum results were for modified cotton fabric (PAam-GG) dyed with madder as a natural dye with catalyst (NH₄)2SO₄ at 140°C for 5 min.

3.3. Color difference.
The color differences between modified cotton fabric (PAam-GG) dyed with madder as a natural dye in CIELAB units with different catalysts [(NH\(_4\)]\(_2\)SO\(_4\) and NH\(_4\)Cl] as appears in Figure (4a,4b), we note ammonium sulfate gives the best result than ammonium chloride with the same conditions because of natural dyes obtained from madder has a good affinity.

\[ \Delta E^{*}_{ab} = \left[ (\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2 \right]^{1/2} \]

\( \Delta E^{*}_{ab} \) is a measure of color difference between all samples with a different catalyst [(NH\(_4\)]\(_2\)SO\(_4\) and NH\(_4\)Cl] as shown in Figure (3a,3b). It shows the difference as a single value for color (reflected by (\(\Delta a^*\))^2+(\(\Delta b^*\))^2) and lightness [reflected by(\(\Delta L^*\))^2], because of different interferences, the color discrimination is usually worse, so we used the color difference to do that.

**Table 5. Uncertainty of color difference (\(\Delta E\)).**

| Catalyst     | Temp.(°C) | 120°C | 140°C | 160°C |
|--------------|-----------|-------|-------|-------|
|              | Time(min.)| F-value| P-value| F-circuit | F-value| P-value| F-circuit | F-value| P-value| F-circuit |
| (NH\(_4\)]\(_2\)SO\(_4\) | 3min.     | 1.110  | 0.323  | 5.318  | 1.847  | 0.211  | 5.318  | 0.004  | 0.949  | 5.318   |
|              | 5min.     | 0.022  | 0.887  | 5.318  | 0.188  | 0.676  | 5.318  | 0.305  | 0.596  | 5.318   |
| NH\(_4\)Cl   | 3min.     | 5.468  | 0.048  | 5.318  | 2.847  | 0.130  | 5.318  | 0.140  | 0.718  | 5.318   |
|              | 5min.     | 1.611  | 0.240  | 5.318  | 0.049  | 0.830  | 5.318  | 0.022  | 0.886  | 5.318   |

As shown in Table 5, the F-value < F- crit. and P-value > 0.05, so we conclude that all samples were homogeneous. Where: P-value is the probability of observing test statistic value, F is Fisher Snedecor distribution, and F- critical is a function of the degrees of freedom of the numerator and the de-numerator and significance level [18,19, 44]. As a result, appears from Table5, the color difference for samples modified with (PAam-GG) dyed with madder as natural dye increase with temperature variation from (120°C to 140°C then slightly decrease at 160°C) with increasing curing time(3 to 5 min.) with catalyst ammonium sulfate more than ammonium chloride at the same condition.

**4. Conclusions**

In this study, the modified cotton fabric with (PAam-GG) composite in the presence of two catalysts [(NH\(_4\)]\(_2\)SO\(_4\) and NH\(_4\)Cl)] then roasting with various time and temperatures then dyed with the madder was able to match with the natural dye used properly. It was evaluated after the study of homogeneity and stability for all color parameters and color differences.
within the limitations of this study. And also, the corresponding shade of the dye appears on unmodified and the modified cotton fabrics dyed in acceptable color differences.

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Conflicts of Interest

The authors declare no conflict of interest.

References

1. Derbyshire A. N.; Marshall W. I. Value analysis of dyes - a new method based on color measurement.” J. Soc. Dyers 1980, 96, 166 –177, https://sdc.org.uk/kv/value-analysis-of-dyes-a-new-method-based-on-colour-measurement/.
2. Burkiniash, S. M.; Wills, A. E. The dyeing of conventional and microfiber nylon 6.6 with reactive dyes. Dyes Pigment 1997, 34, 243–253, https://doi.org/10.1016/S0143-7208(96)00074-5.
3. Cui, W.; Mazza, G.; Biliaderis, C. G. Chemical Structure, molecular size distributions and rheological properties of flaxseed gum, J. of Agriculture and food chemistry 1994, 42, 1891-1895, https://doi.org/10.1021/jf00045a012.
4. Chudzikowski, R. J. Guar gum and its applications. J. of the Society of Cosmetic Chemists 1971, 22, 43, https://citeseerx.ist.psu.edu/viewdoc/download?doi=10.1.1.560.2090&rep=rep1&type=pdf
5. Sudhir, S.; Kulbhushan, G.; Sangamesh, V. A.; Sultan, H. B.; Brian, S.; Dawn, V. L. Growth and Yield of Guar (Cyamopsis tetragonol L.) Genotypes under Different Planting Dates in the Semi-Arid Southern High Plains. American Journal of Plant Sciences 2016, 7, 1246-1258, http://dx.doi.org/10.4236/ajaps.2016.78120.
6. Deshkar, N.; Tiloo, S.; Fande, V. A. A comprehensive review of Rubia cordifolia Linn. Pharmaco. Rev. 2008, 2, 124-134, https://www.phcogrev.com/sites/default/files/PhcogRev-2-3-124.pdf.
7. Parvinzadeh M. The effects of softeners on the properties of sulfur-dyed cotton fibers. J. of Surfact. and Deterg. 2007, 10, 219–223, https://doi.org/10.1007/s11743-007-1034-6.
8. Vankar, P. S.; Shanker, R.; Mahanta, D.; Tiwari, S. C. Ecofriendly Sonicator dyeing of cotton with rubiacordifolialinn. Using biomordant , Dyes and Pigm. 2008, 76, 207-212, https://doi.org/10.1016/j.dyepig.2006.08.023.
9. Adeeel S.; Zuber M.; Fazal U. R.; Zia K.M.;(2018) : Microwave-assisted extraction and dyeing of chemical and bio-mordanted cotton fabric using harmal seeds as a source of natural dye , Environ. Sci. Pollution Res.,25, 11100-11110. DOI: 10.1007/s11356-018-1301-2
10. Vankar P. S.; Shanker R.; Verma, A.;(2007) : Enzymatic natural dyeing of cotton and silk fabrics without metal mordants, J. Cleaner Prod., 15(15), 1441-1450. doi:10.1016/j.jclepro.2006.05.004
11. Ticha M. B.; Haddar W.; Meksi N.; Guesmi A.; Mhenni M. F.;(2016) : Improving dyeability of modified cotton fabrics by the natural aqueous extract from red cabbage using ultrasonic energy, Carbohydrate Poly.,154, 287-295. http://dx.doi.org/10.1016/j.carbpol.2016.06.056
12. Haddar W.; Baaka N.; Meksi N.; Ticha M.; Guesmi A.; Mhenni M. F.;(2015) : Use of ultrasonic energy for enhancing the dyeing performances of polyamide fibers with olive vegetable water, Fiber Poly., 16, 1506-1511, DOI 10.1007/s12221-015-4931-8
13. Dumitrescu, I.; Mitran, E. C.; Varzaru, E.; Constantinescu, R.; Iordache, O. G.; Stefanescu, D.; Pislaru, M. Multi-functional effects of textiles dyed with madder roots powder (rubiatinctoria), Ind. Text. 2018, 69, 451-457, http://dx.doi.org/10.3355/IT.069.06.1576.
14. Haji, A. Improved natural dyeing of cotton by treatment and chitosan coating; optimization by response surface methodology. Cellulose Chem. Technol. 2017, 51, 975-982.
15. Haji, A.; Khajeh, M. M.; Hashemizad, S. Plasma and chitosan treatments for improvement of natural dyeing and antibacterial properties of cotton and wool. Vlakna Text. 2016, 23, 86-89.
16. Özgür, M.Ü.; Gümrükü, G.; Turak, F.; Saroğlu, C. Dyeing cotton strips with natural dyes for improved fastness by dyeing with liquids containing tannic acid and water-soluble metal salts, 4th Aacd Congress, Adnan Menderes University, Turkey, 2004.
17. Teli, M. D.; Sheikh, J.; Shastrakar, P. Exploratory investigation of chitosan as mordant for eco-friendly antibacterial printing of cotton with natural dyes. J. Text. 2013, 2013, 1-6, https://doi.org/10.1155/2013/320510.

18. Vankar, P. S.; Shanker, R.; Srivastava, J. Ultrasonic dyeing of cotton fabric with aqueous extract of eclipta Alba. Dyes Pigm. 2007, 72, 33-37, https://doi.org/10.1016/j.dyepig.2005.07.013.

19. Haji, A.; Nasiriboroumand, M.; Qavamnia, S. S. Cotton dyeing and antibacterial finishing using agricultural waste by an eco-friendly process optimized by response surface methodology. Fiber Poly.2018, 19, 2359-2364, https://doi.org/10.1007/s12221-018-8657-2.

20. Benli, H.; Bahtiyari, M. D. Combination of ozone and ultrasound in retreatment of cotton fabrics prior tonatural dyeing. J. Clean. Proud. 2015, 89, 116-124, http://dx.doi.org/10.1016/j.jclepro.2014.11.007.

21. Vankar, P. S.; Tiwari, V.; Singh, L. W.; Potsangbam L. Sonicator dyeing of cotton fabric and chemical characterization of the Colorant from Melastoma malabathricum, Pigm. Res. Technol. 2009,38, 38–42, https://doi.org/10.1016/j.03699420910923562.

22. Vankar, P. S.; Shanker, R.; Dixit, S.; Mahanta, D.; Tiwari, S. C. Sonicator dyeing of cotton with the leaves extract Acer pectinatum Wallich, Pigm. Res. Technol. 2008, 37, 308–313, https://doi.org/10.1016/j.0369942081091981.

23. Vankar, P. S.; Shanker, R.; Dixit, S. D.; Tiwari, S. C. Sonicator dyeing of modified cotton, wool and silk with mahonianapaulensis dc. And identification of the colorant in mahonia. Ind. Crop Prod. 2008, 27, 371-379, https://doi.org/10.1016/j.indcrop.2007.12.009.

24. Vankar, P. S.; Shanker, R.; Dixit, S. Chemical characterisation of extract derived from Daphne papyraceae and sonicator dyeing of cotton, silk and wool with the extract. Pigm. Res. Technol. 2009, 38,181–187, https://doi.org/10.1016/j.03699420910957042.

25. Padma, R.S.; Vankar, S. Ecofriendly ultrasonic natural dyeing of cotton fabric with enzyme pretreatments. Desalination 2008, 230, 62-69, https://doi.org/10.1016/j.desal.2007.11.016.

26. Baaka, N.; Mahfoudhi, A.; Haddar, W.; Mhenni, M. F.; Mighri, Z. Green dyeing process of modified cotton fibers using natural dyes extracted from Tamarix aphylla (L.) karst. Leaves. Nat. Product Res. 2017, 31, 22-31, https://doi.org/10.1080/14786419.2016.1207072.

27. Zahid, M.; Bhatti, I. A.; Adeel, S.; Saba, S. Modification of cotton fabric for textile dyeing: Industrial mercerization versus gamma irradiation. J. Text. Instit. 2017, 108, 287-292, https://doi.org/10.1080/00405000.2016.1165398.

28. Rehman, F.; Adeel, S.; Hanif, R.; Muneer, M.; Zia, K. M.; Zubr, M.; Jamal, M. A.; Khosa, M. K. Modulation of marigold-based lutein dye and its dyeing behavior using UV radiation. J. Natural Fib 2017, 14, 63-70, https://doi.org/10.15440/478.2016.1146642.

29. Hussaan, M.; Iqbal, N.; Adeel, S.; Azeem, M.; TariqJaved, M.; Raza, A. Microwave-assisted enhancement of milkweed (calotropisprocera l.) leaves as an ecofriendly source of natural colorants for textile. Environ. Sci. Pollution Res. 2017, 24,5089-5094, https://doi.org/10.1007/s11356-016-8162-3.

30. Adeel, S.; Gulzar, T.; Azeem, M.; Fazalur, R.; Saeed, M.; Hanif, I.; Iqbal, N. Appraisal of marigold flower based lutein as natural colorant for textile dyeing under their fluence of gamma radiations. Radiation Phys. Chem. 2017, 130, 35-39, https://doi.org/10.1016/j.radphyschem.2016.07.010.

31. Gulzar, T.; Adeel, S.; Hanif, I.; Rehman, F.; Hanif, R.; Zubr, M.; Akhtar, N. Eco-friendly dyeing of gamma ray induced cotton using natural quercetin extracted from acacia bark (a. Nilotica). J. Nat. Fiber 2015, 12,494-504, https://doi.org/10.1080/15440478.2014.964445.

32. Rehman, F.U.; Adeel, S.; Shahid, M.; Bhatti, I. A.; Nasir, F.; Akhtar, N.; Ahmad, Z. Dyeing of γ-irradiated cotton with natural flavonoid dye extracted from irradiated onion shells (allium cepa) powder. Radiation Phys. Chem. 2013, 92, 71-75, https://doi.org/10.1016/j.radphyschem.2013.07.002.

33. Gorjanc, M.; Mozetič, M.; Vesel, A.; Zaplotnik, R. Natural dyeing and UV protection of plasma treated cotton. Europ. Phys. Jour. A (EPJ A) 2018, 72, 41-46, https://doi.org/10.1140/epjae2017-80680-9.

34. Zahran, M.K.; Mahmoud, R.I. Synthesis and Characterization of Polycrylamide Guar Gum Graft Copolymer. Journal of Textile Association 2006, 273-277.

35. Baer, P.I.; Marriet, F.W.; Panik, Y. A Practical Introduce to the dyeing and Finishing of wool Fabrics. The Society of Ion Dyers and Colorists, Bradford, UK. 1986, https://www.bookdepository.com/Practical-Introduction-Dyeing-Finishing-Wool-Fabrics-Ian-Bearpark/9780901956446.

36. Commission Internationale de l’Eclairage. CIE Technical Report: Colorimetry. CIE Pub No. 15.3. Vienna, Austria: CIE Central Bureau; 2004.

37. Park, J. Instrumental Color Formulation: A Practical Guide, The Society of Dyers and Colorists. West Yorkshire 1993, https://www.waterstones.com/book/instrumental-colour-formulation/james-park/society-of-dyers-and-colourists/9780901956545.

38. Punrattanasin, N.; Nakpathom, M.; Somboon, B.; Narumol,N.; Rungruangkitkrai, N.; Mongkolrattanasit, R. Silk fabric dyeing with natural dye from mangrove bark (Rhizophoraapiculdatablume) extract. Industrial Crops and Products 2013, 49,122-129, https://doi.org/10.1016/j.indcrop.2013.04.041.

39. International Organization for Standardization (ISO 5725 -2). Accuracy (trueness and precision) of measurement methods and results -- Part 2: Basic method for the determination of repeatability and reproducibility of a standard measurement method, 1994.
40. David, M. D.; Christopher, D. B.; Cetinkaya, R. M. *Open Intro Statistics* (3rd Ed.). Open Intro. Retrieved 11 Nov. 2017.
41. Daya S. One-way analysis of variance, *Evidence-Based Obstet. Gynecol.* 2003, 5, 153–155.
42. Cortina, J. M.; Nouri, H. *Effect size for ANOVA designs*. Thousand Oaks, CA: Sage Publications. Effect Size for ANOVA Designs (Quantitative Applications in the Social Sciences. https://ro.scribd.com/document/388001645/Effect-Size-for-ANOVA-Designs-pdf.
43. International Organization for Standardization, (ISO/IEC 17043). Proficiency testing by internal laboratory comparisons – Part 1: Development and operation of proficiency testing schemes, Geneva. 2nd.
44. International Organization for Standardization (ISO 3534-1). Statistics — Vocabulary and symbols — Part 1: General statistical terms and terms used in probability. International Organization for Standardization (ISO 3534-1 :2006). Statistics —Vocabulary and symbols — Part 1: General statistical terms and terms used in probability.