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Dyeing of jute with annatto seeds and other natural dyes

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Jute fabric samples have been dyed with aqueous solution of annatto (A) seeds along with three other natural dyes, viz. onion (O) skin, pomegranate (P) rind and marigold (M) flower petals. Varying proportions of binary mixture of these dyes have been used to dye jute fabrics which are mordanted with myrobolan and natural alum before dyeing. The dyeing performance is evaluated by measuring the colour strength and other related parameters along with the fastness properties of the dyed jute fabrics. The dyed samples are further treated with chitosan and n-cetyl trimethyl ammonium bromide (CTAB) to improve wash fastness. The treatments improve the wash fastness by 1 grade. The treatment of dyed jute fabrics with benzotriazole improves the light fastness by 1 grade. The compatibility of annatto dye with respect to other three dyes has also been assessed by plotting two different curves, viz. ΔC^* *vs* ΔL^* and K/S *vs* ΔL^* for progressive depth of shade for each set of binary mixtures. These results are compared with a new parameter called Colour Difference Index (CDI). The results of CDI are in tandem with the results found from the plots, implying that the three binary pairs of natural dyes are comparable in different degrees.

Keywords: Alum, Annatto seeds, Jute, Marigold, Myrobolan, Natural dyeing, Onion skin, Pomegranate rind

1 Introduction

Jute being a lignocellulosic bast fibre is traditionally used in packaging materials. Nowadays several ways are being explored to diversify the usage of jute for its greater marketability. Though 60% of jute fibre is composed of cellulose, the fibre is different from cotton-like cellulosic fibres in respect of dyeing characteristics, like affinity and absorption owing to the presence of other chemical groups, such as -COOH, -CHO, etc. This makes the study of different aspects of dyeing of jute important to explore. As the diversified jute products are growing in demand, its demand potential can be increased when this natural fibre is dyed with an eco-friendly natural dye.

Natural dyeing has several shortcomings regarding its reproducibility of shades, limited range of shades, low fastness, etc. These shortcomings can be mitigated if a combination of few selected natural dyes can be used to create a desired shade. So, the compatibility of such binary mixtures of natural dyes needs to be studied. To the best of our knowledge, there are only a few discerning research articles available on the studies of compatibility of natural dyes, and on compatibility of jute[1-3]. This aspect of natural dyeing should be studied in detail as combinations of dyes can produce a gamut of shades, which is required for practical purposes. Compatibility of a binary pair of dyes can be assessed by various methods, though the most common method is plotting of ΔC^* *vs* ΔL^* or K/S *vs* ΔL^* values for two sets of progressive shades of the dyed samples. In the present study, compatibility of annatto seeds, a popular natural dye in combination with three other natural dyes, viz. onion (O) skin, pomegranate (P) rind and marigold (M) flower petals has been studied using conventional plotting method as well as using a new method calculating Colour Difference Index (CDI) and Relative Compatibility Ratio (RCR) of the dyed jute fabrics.

2 Materials and Methods

2.1 Materials

Bleached jute fabric of the specification, 47 ends/dm warp density, 42 picks/dm weft density, 7.65 lb/spy warp count, 8.23 lb/spy weft count, and 260 g/m^2 area density, was used.

Four natural resources, viz annatto (A) seeds, marigold (M) flower petals, onion (O) skins and pomegranate (P) rind, were used as sources of natural dyes. The main colorant components found in annatto

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are bixin & norbixin[4] [Fig. 1(a) & (b)], while lutein[5] [Fig. 1(c)] is the main colorant of marigold flower petals. Onion skins have quercetin[6] [Fig. 1(d)] and pomegranate rinds have granatonine[7] [Fig. 1(e)] as their respective main colouring components.

The mordants, natural alum [aluminium potassium sulphate, $KAl(SO_4)_2.12H_2O$] and myrobalan (*Terminalia chebula*) were procured from a local shop. The chemical structure of chebulinic acid, the main chemical compound present in myrobolan, is shown in Fig. 1(f). Auxiliary chemicals LR-grade acetic acid and sodium carbonate (for adjustment of *p*H) and non-ionic detergent were procured from M/s Ma Kali Chemicals, Kolkata.

2.2 Methods

2.2.1 Extraction of Natural Dye

Annatto seeds, onion skins, marigold petals and pomegranate rinds were pulverised to powder in a mechanical grinder and the powder was put into aqueous medium under optimized conditions (Table 1) for the maximum colour extraction. The aqueous extract was filtered and filtrate was used as natural dye.

2.2.2 Mordanting of Jute Fabric

Bleached jute fabrics were pre-mordanted (mordanting done before dyeing process) using myrobolan and then natural alum applied in sequence. Both the mordants were applied individually with 10% add-on onto the fabric.

2.2.3 Dyeing of Pre-mordanted Jute Fabric

Jute fabrics already mordanted were dyed with the binary pairs of two natural dyes in different ratios (100:0, 75:25, 50:50, 25:75 and 0:100), applying overall 20% (owf) concentration of the dyes (based on the mass of powdered dye) at 100°C using MLR 1:40 for 30 min, adding 10 g/L sodium chloride as electrolyte. In each case, the dyed sample was rinsed in cold water followed by soaping using 2 g/L of non-ionic detergent dissolved in water (1:50 MLR) at 60°C for 15 min. Finally, it was again rinsed in cold water and dried in air.

2.2.4 Application of Chitosan, CTAB and UV-absorber

Dyed jute fabrics were treated with aqueous solutions of 1% chitosan and 2% chitosan separately in acidic medium using acetic acid by pad-dry method. Similarly, 1% CTAB, 2% CTAB and 1%

Table 1 — Optimized conditions for extraction of colour from selected natural dyes

Dye	Time min	Temperature during extraction, °C	Initial MLR	*p*H
Annatto[8]	60	90	1:30	11
Marigold[9]	45	90	1:25	11
Onion[10]	60	90	1:30	9
Pomegranate[11]	80	90	1:20	5

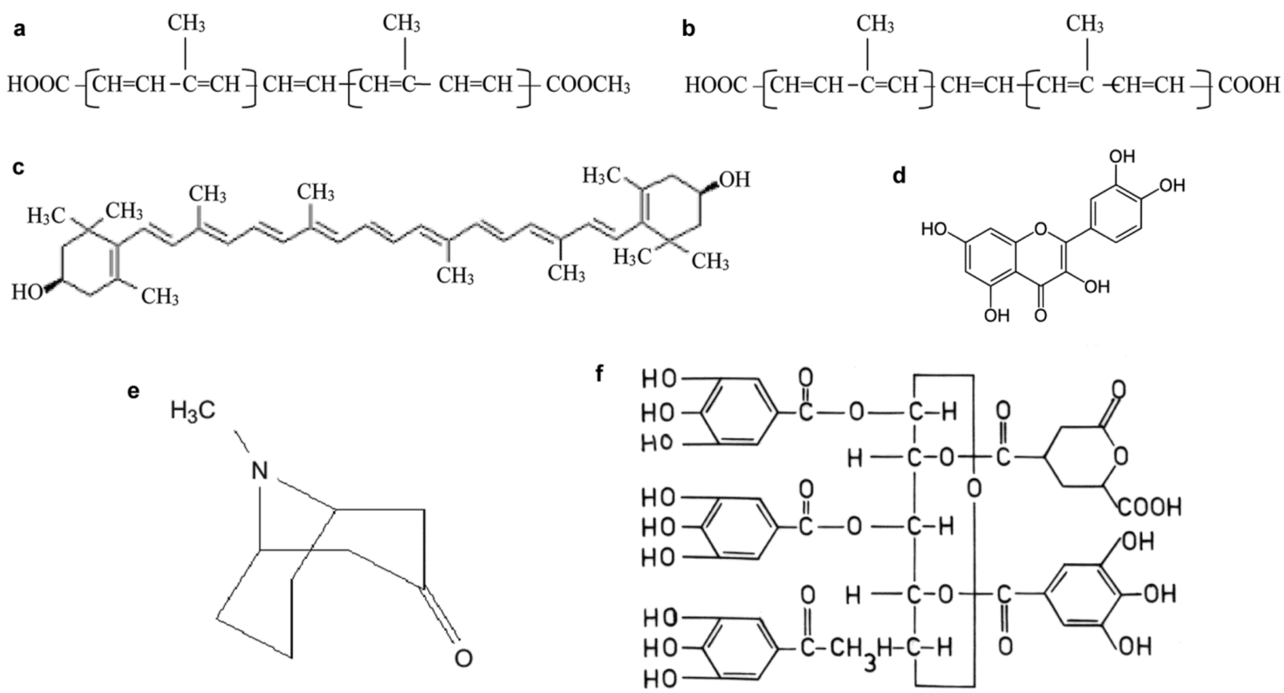

Fig. 1 — Chemical structures of (a) bixin, (b) norbixin, (c) lutein, (d) quercetin, (e) granatonine and (f) chebulinic acid

benzotriazole were applied by pad-dry method in presence of 0.5% sodium hydroxide (alkaline medium), keeping 100% wet pick-up in the jute fabric followed by dry-cum-heat treatment at 100°C for 15 min in laboratory stenter.

2.2.5 Testing of Dyed Jute Fabric

Colour strength (*K*/*S* values) and other colour-related parameters like ΔL^* (difference in lightness), Δa^* (difference in redness/greenness), Δb^* (difference in blueness/yellowness), ΔC^* (difference in psychometric chroma), ΔH^* (difference in hue), ΔE^* (total colour difference) and LABD Metamerism Index (MI) of the dyed jute fabrics were measured using reflectance spectrophotometer (Make Premier Colorscan; Model S-5100A) Kubelka-Munk equation and CIE-Lab equations, 1976. Colour fastness to washing, rubbing and light of the dyed jute fabrics were measured using ISO 105-C10:2006 (reviewed in 2020), ISO 105-X12:2016 (reviewed in 2021) and ISO 105-B01:2014 (reviewed in 2020) respectively. CDI is evaluated on the basis of earlier findings[1].

2.2.6 Compatibility Tests for Selected Binary Mixtures of Dyes

(i) Conventional Method[1,2,12,13]

The following binary pairs of natural dyes in (50:50) proportions using 20% (owf) dye extracts were used to dye the pre-mordanted jute fabrics:

- M1 – Annatto seeds + Onion skins (A+O)
- M2 – Annatto seeds + Pomegranate rinds (A+P)
- M3 – Annatto seeds + Marigold flower petals (A+M)

To carry out compatibility tests of the mentioned binary pairs of dyes, pre-mordanted jute fabrics were dyed in two different sets (Set I and Set II) of progressing depth of shades for each pair of dyes taken in equal (50:50) proportion.

The dyeing process variables in Set I and Set II were maintained as followed in the earlier works[1, 2, 11]. Here, in Set I, the first dyed fabric samples was taken out of dyebath at 60°C and the process of taking of samples from bath was continued in the intervals of 10 min, keeping the rate of dyeing as 10°C/min and the final sample was taken out 30 min after reaching 100°C. In a nutshell, the altogether 8 dyed samples were taken out of bath at 60°C, 70°C, 80°C, 90°C, 100°C, 100°C (remained for 10 min), 100°C (remained for 20 min) and 100°C (remained for 30 min).

In Set II, five jute samples were dyed with 4% (owf), 8% (owf), 12% (owf), 16% (owf) and 20% of each pair of dyes taken in equal (50:50) proportion at 100°C for 30 min. Here, samples were dyed with 4 percentage point increments of dye concentrations.

All the dyed samples in Set I and Set II were rinsed and dried using the procedure as mentioned in section **2.2.3**. The colorimetric properties of the dyed samples were measured in the same way as mentioned in section **2.2.5.** The compatibility was assessed by degree of closeness and overlapping of ΔC^* *vs* ΔL^* or *K*/*S vs* ΔL^* curves of selected binary pairs.

(ii) Non-conventional CDI-based Method[2,11]

An alternate method to judge compatibility is already established in the earlier work[2, 11]. Here, Colour Difference Index (CDI) of all selected pairs was calculated and from the values of CDI, Relative Compatibility Rating (RCR) and compatibility grade were evaluated. The formula for CDI is as follows:

$$\mathrm{CDI} = (\Delta E^* \times \Delta H^*) / (\Delta C^* \times \mathrm{MI})$$

3 Results and Discussion

3.1 Colour Strength of Dyed Jute Fabric

Table 2 shows the colour strength (*K*/S values) at a common wavelength of 420 nm and other relevant colour parameters (ΔL^*, Δa^*, Δb^*, ΔC^*, ΔH^*, ΔE^*, MI) of the dyed jute fabric samples along with calculated values of CDI for each dyed sample. The highest Δa^* values are observed for M1 (A+O) combination of dyes in all proportions, thus implying that the shades obtained in M1 are of redder than those of other two combinations of dyes. Similarly, the highest Δb^* values are obtained for M2 (A+P) combinations of dyes. This means that the shades obtained from M2 pair of dyes are greener than the shades of other two pairs.

The highest change in chroma (ΔC^*) is observed for M2 (A+P) pair of dyes in all proportions, while the highest change in hue (absolute values of ΔH^*) is found for M1 pair of dyes in all proportions. The change in hue angles (ΔH^*) of the dyed samples are found negative for all dyed samples. The decreasing order of the absolute values of ΔC^* and ΔH^* of the dyed samples are:

ΔC^* - M2 > M3 > M1
ΔH^* - M1 > M3 > M2

The highest value of MI is found for M1 pair of dyes in 75:25 proportion and the decreasing order of the absolute values of MI of the dyed samples in 50:50 proportion is M2 > M1 > M3.

Table 2 — Colour strength and related colour parameters of jute dyed with binary pairs of natural dyes after mordanting

Dye combinations	*K/S* at 420 nm	Δ*L**	Δ*a**	Δ*b**	Δ*C**	Δ*H**	Δ*E**	M.I.	CDI
Control (semi-bleached jute fabric)	0.26	-	-	-	-	-	-	-	-
For 100:0 proportion									
M1	4.6	-14.1	16.0	14.6	19.0	-24.5	25.9	3.1	10.8
M2	4.6	-14.1	16.0	14.6	19.0	-24.5	25.9	3.1	10.8
M3	4.6	-14.1	16.0	14.6	19.0	-24.5	25.9	3.1	10.8
For 75:25 proportion									
M1	5.2	-17.0	9.6	10.0	12.0	-17.1	22.0	3.2	9.9
M2	4.2	-16.2	10.2	17.1	18.8	-12.8	25.7	3.1	5.7
M3	5.6	-23.9	9.6	15.2	16.8	-13.0	30.0	3.1	7.5
For 50:50 proportion									
M1	5.4	-18.4	9.4	11.7	13.4	-15.2	23.8	3.1	8.7
M2	4.1	-17.2	8.9	18.0	19.2	-10.1	26.5	2.9	4.8
M3	6.1	-25.5	8.2	14.3	15.5	-10.9	30.4	2.8	7.6
For 25:75 proportion									
M1	5.5	-19.5	9.6	11.0	12.9	-16.2	24.4	3.1	9.9
M2	4.0	-19.1	8.6	19.0	20.1	-9.0	28.3	2.8	4.5
M3	6.3	-26.9	7.7	14.0	15.0	-10.1	31.3	2.5	8.4
For 0:100 proportion									
M1	5.5	-29.4	8.0	18.1	19.1	-8.4	35.5	2.4	6.5
M2	4.0	-20.2	7.9	19.8	20.7	-7.5	29.4	2.5	4.3
M3	7.1	-30.3	6.6	12.7	13.5	-8.6	33.6	2.5	8.5

Table 3 — Colour fastness of jute fabrics dyed with binary mixtures of dyes

Dye combination	Colour fastness to															
	Washing										Light		Rubbing	
	Untreated		1% chitosan		2% chitosan		1% CTAB*		2% CTAB		Untreated	1% benzotriazole	(Untreated)	
	LoD[#]	ST[^]	LoD	ST	LoD	ST	LoD	ST	LoD	ST			Dry	Wet
For 75:25 proportion														
M1	3	3-4	3-4	4	4	4-5	3-4	4-5	4	4-5	3	4	4	3-4
M2	3-4	4	4	4-5	4-5	4-5	4	4-5	4	4-5	3	4	4	3
M3	3	4	3-4	4	4	4-5	3-4	4-5	4	4-5	2-3	3-4	4	3
For 50:50 proportion														
M1	3	3-4	3-4	4	4	5	3-4	4-5	4	5	3	4	4	3
M2	4	4-5	4	4-5	4	4-5	4	4-5	4	4-5	3	4	4	3
M3	3	4	3-4	4	4	4-5	3-4	4-5	4	4-5	3	4	4	3
For 25:75 proportion														
M1	3	4	3-4	4-5	4	5	3-4	4-5	4	5	3	4	4	3
M2	4	4-5	4	4-5	4	4-5	4	4-5	4	4-5	3	4	4	3
M3	3	4	3-4	4-5	4	5	3-4	4-5	4	5	3	4	4	3

[#]LoD = Loss of depth of shade; [^]ST = staining of colour on adjacent bleached jute fabric; *CTAB = n-cetyl trimethyl ammonium bromide

3.2 Colour Fastness

Table 3 shows the fastness values of jute fabrics dyed with three different binary pairs of natural dyes in three different proportions.

The table shows that the binary pair M2 (A+P) achieves the best wash fastness than the other two pairs in proportions. Its loss of depth of shade (LoD) of 4 and staining of 4-5 in (50:50) and (75:25) proportions which are the highest among the untreated samples are satisfactory fastness properties. When treated with chitosan and CTAB, the colour fastness to wash of the dyed jute fabrics is improved, except the pair M2 in (50:50) and (75:25) proportions. While treatment with 1% chitosan improves the wash fastness by ½ grades, the treatment with 2% chitosan changes the wash fastness by 1 grade. Similar trend of results is obtained in case of treatment with CTAB, a cationic fixing agent.

Generally, CTAB is treated with 0.5% sodium hydroxide (alkaline medium), which dissociates as Na^+ ion and OH^- ion. Most of the natural dyes studied here contain phenolic –OH group, which in alkaline medium creates dissociation of the dye as the dye anion. Thus, CTAB being a cationic dye-fixer compound easily forms a larger sized complex with above-said dye anion and precipitates as insoluble ligand. This is why the colour fastness to washing is improved by this application[15].

Chitosan is reactive for the functionality of its primary amino group and the primary and secondary hydroxyl groups present in it. The primary amino group of chitosan in an acidic medium form a cationic derivative generating NH_3^+ functionality in an acidic medium and therefore it acts as a natural cationic dye-fixer to enhance washing fastness of anionic natural dyes. This is why the colour fast to washing is improved by the application of chitosan in an acidic medium[15].

Though the colour fastness to light of the dyed samples is poor to average (2/3 to 3), these are improved by the application of 1% benzotriazole, UV-absorber. Benzotriazole absorbs some portions of UV rays, thus reducing the fading and decolouration of dye molecules and as well as lignin component of jute fibre[14].

In all binary pair for all proportions of mixtures, the rub fastness (dry and wet) is good. So, dyed fabrics do not need further chemical treatment for improvement. The good rub fastness may be attributed for the absence of any unfixed dyes left after washing and soaping and dye molecules are fixed inside the fibre with ionic or hydrogen or coordinated bond formations.

3.3 Compatibility Test

The compatibility of dyes is important for obtaining a desired shade and expected colour values of dyed materials. Due to the variance in chemical composition, a natural dye compatible with a natural dye may be incompatible with other natural dyes. Two methods for testing compatibility are used in the present work. The first one is the conventional method involving the comparison of two sets of plots ΔC^* *vs* ΔL^* and K/S *vs* ΔL^* for two sets of progressive depth of shade. However, there is a need to assess the compatibility in an easier and less time-consuming way, so a new method which is reported earlier has also been used in the present work as non-conventional method. This method involves the computation of CDI values, and RCR is evaluated from the spread of CDIs for various binary pairs.

Relative compatibility rating of a binary mixture of dyes is higher when the difference of the maximum and minimum values of its CDIs is lower; the lower the difference, the higher is the compatibility rating. Though the CDI-based method is proven with scientific evidence in several published articles, two methods have been compared here for better understanding.

Figure 2 shows the plots of ΔC^* *vs* ΔL^* [plots (a) – (c)] and K/S *vs* ΔL^* [plots (a)' – (c')] for two sets (Set I and Set II) of dyed jute fabrics for three separate pairs (M1, M2, M3) of natural dyes and Table 4 shows the CDI, RCR and compatibility grades of three binary pairs.

As for M1 (A+O) pair, the curves of ΔC^* *vs* ΔL^* for Set I and Set II show similar trend with a separation and the curves of K/S *vs* ΔL^* for Set I and Set II run in the same manner. The curves do not intersect at any point. These curves imply poor compatibility for M1 binary pair. This inference is buttressed by the relative compatibility rating of 2 (Table 4), which gives the compatibility grade as "poor".

As for M2 (A+P) pair, the curves of ΔC^* *vs* ΔL^* for Set I and Set II run with a gap but the curves of K/S *vs* ΔL^* for Set I and Set II are about to intersect run in the same manner. The pair of dyes shows the RCR of 2-3 (fair) meaning closer similarity with the K/S *vs* ΔL^* curves.

In the case of M3 (A+M), the plots of ΔC^* *vs* ΔL^* for Set I and Set II run very closely and almost

Table 4 — Colour Difference Indices and Relative Colour Rating of dyed samples

Dye combination	CDI			CDI_{Max} - CDI_{Min}	RCR	Compatibility grade
	75:25	50:50	25:75			
M1	9.9	8.7	9.9	9.9 – 8.7 = 1.2	2	Poor
M2	5.7	4.8	4.5	5.7 – 4.8 = 0.9	2-3	Fair
M3	7.5	7.6	8.4	8.4 – 7.5 = 0.9	2-3	Fair

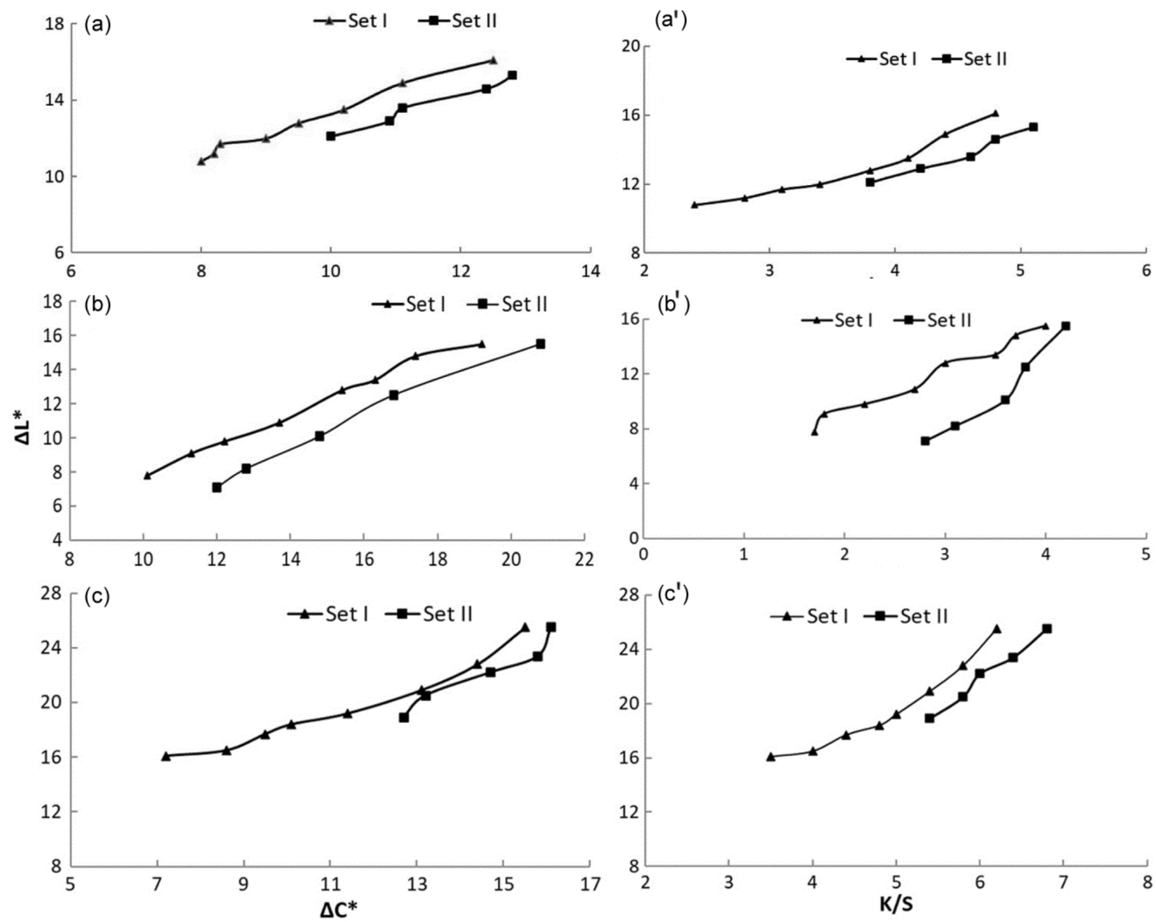

Fig. 2 — Plots of ΔC *vs* ΔL and *K/S vs* ΔL for dyeing of three binary pairs of natural dyes on pre-mordanted jute fabrics [M1 – a and a'; M2 – b and b'; and M3 – c and c']

intersected at a point and the curves of *K/S vs* Δ*L** for Set I and Set II show a similar approach. In the RCR system, this pair of dyes shows grade 2-3 (fair), implying close similarity with the curves.

4 Conclusion

The present study establishes the compatibility of three different binary pair of natural dyes to apply on jute. It is found that the decreasing order of the change in chroma values of the pairs is (A+P)> (A+M) > (A+O) and the decreasing order of the change in hue values of the pairs is (A+O) > (A+M) > (A+P). The wash fastness values for (A+P) pair for (50:50) and (75:25) proportions are the best results. The after treatment with chitosan and CTAB improves the wash fastness by 1 grade. The application of 1% benzotriazole improves the light fastness by 1 grade for all three binary pairs.

The results of the non-conventional compatibility tests are found in good agreement with the results of the conventional method of plot comparisons. As per RCR system, the compatibility grades of (A+O), (A+P) and (A+M) pairs are poor, fair and fair respectively. This present work of compatibility of the particular binary pairs of dyes can be useful for industrial application of these pairs to dye jute products.

This study will help the dyers to produce different compound shades with the use of binary mixtures of suitable compatible natural dyes to obtain optimum colour yield and maximum colour fastness rating to satisfy customers' requirements. Thus, knowledge of compatibility of dyes will help in producing desirable compound shades with mixtures of compatible dyes.

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