

# Eco-friendly bio-dyeing of bio-treated nylon fabric using Esfand (*P. harmala*) based yellow natural colorant

Journal of Engineered Fibers and Fabrics

Volume 17: 1–15

© The Author(s) 2022

DOI: 10.1177/15589250221091265

journals.sagepub.com/home/jef



Shahid Adeel<sup>1</sup> , Mahmood ul Hasan<sup>1</sup>, Fatima Batool<sup>2</sup>,  
Meral Ozomay<sup>3</sup>, Mozhgan Hosseinezhad<sup>4</sup> , Nimra Amin<sup>5</sup>  
and Muhammad Hussaan<sup>6</sup>

## Abstract

The ongoing age is the time of sustainability, where in the current pandemic scenario, which is getting worse, needs treatment with nature rather than chemical-based products. In this study, microwaves (M.W. rays) treatments as extraction mode for Esfand (*P. harmala*) have been revived for polyamide (nylon) dyeing. The water solubilized and acid solubilized filtrates and polyamide (nylon fabrics) were treated M.W. rays up to 10 min with an interval of 2 min. Mordanting with chemicals and plant extracts before and after dyeing was done at 60°C–80°C. It has been found the application of M.W. ray treatment for 4 min., to 30 mL of extract of 8 pH containing 4 g/100 mL of Table salt as leveling agent has given desired results when employed at 55°C for 55 min. Statistical analysis of dyeing variables through R.S.M., and two way-Anova shows that the effect of these variables has been observed highly significant. Experimentally it has been observed that the application of extract for dyeing of polyamide (nylon fabric) has given good results when chemical or bio-mordanted at selected conditions. Practically, Esfand seeds has ability for *bio*-coloration of surface modified polyamide fabric (nylon fabric), and utilization of pomegranate extract as bio-mordant and tannic acid as sustainable chemical mordant has furnished colorfast shades.

## Keywords

Acacia, Esfand, FTIR, nylon fabric, peanut, SEM, sustainability, turmeric

Date received: 26 December 2021; accepted: 15 March 2022

<sup>1</sup>Department of Chemistry, Government College University Faisalabad, Faisalabad, Pakistan

<sup>2</sup>Department of Botany, Division of Science and technology, University of Education Lahore, Lahore, Pakistan

<sup>3</sup>Department of Textile Engineering, Marmara University Istanbul, Istanbul, Turkey

<sup>4</sup>Department of Organic Colorants, Institute for Color Science and Technology, Tehran, Iran

<sup>5</sup>Department of Applied Chemistry, Government College University Faisalabad, Faisalabad, Pakistan

<sup>6</sup>Department of Botany, Government College University Faisalabad, Faisalabad, Pakistan

## Corresponding authors:

Shahid Adeel, Department of Chemistry, Government College University Faisalabad, Faisalabad 38040, Pakistan.  
Email: shahidadeelappliedchemist@gmail.com

Meral Ozomay, Department of Textile Engineering, Marmara University Istanbul, Istanbul 34722, Turkey.  
Email: meral.akkaya@marmara.edu.tr

Mozhgan Hosseinezhad, Department of Organic Colorants, Institute for Color Science and Technology, No. 55, Vafamaneh St., Lavizan Exit, Sayad Shirazi North HWY, Tehran, Iran.  
Email: hosseinezhad-mo@icrc.ac.ir



## Introduction

Herbal-based green products are the part and parcel of the global community.<sup>1,2</sup> Their utilization as the pharmaceuticals has played an essential role in the introduction of world-famous medicines systems such as Chinese, Greek, and Indian Ayurvedic.<sup>3</sup> Because these green products have such potent isolates which contains not only biological characteristics beneficial for the global community but also have the ability to color the matrices such as textile, perfume, flavors, food, pharmaceuticals etc.<sup>4,5</sup> Many plants have been introduced in the world of medicines which has such functional moieties that has the ability to protect the global community from evil diseases such as cholera, flu, plague, dengue, typhoid, swine flu, bird flu, etc., of these plants.<sup>3</sup>

Esfand (*Peganum harmala* L.) commonly known as Harmal belongs to the family of Zygophyllaceae.<sup>6</sup> Its seeds extract (Figure 1(a)) contains specific alkaloids such as harmol, harmaline, and harmine (Figure 1(b)) which not only have good efficacy to cure various diseases but also exhibit antioxidants, antibacterial, antiviral, anti-tumor, and anti-inflammatory effects.<sup>6-8</sup> The smoke of its seeds is also said to be antidote for the evil spirits. Its extracts are also used as a possible substitute for Turkey Red Color (TR) to dye fabrics, and to make tattoos and inks. The alkaloids present in extract which act as the source of natural yellow dye for textiles.<sup>9,10</sup> Hence the medicinal-based plants colorants (natural dyes) are found as a possible substitute of carcinogenic synthetic dyes.<sup>11,12</sup> The utilization of harmal seeds have been first time has been explored by our research group onto cotton, silk, wool, and nylon fabrics.

Before discovery of synthetic dyes “Mauveine” by Perkin, these plants were used to color fabrics, houses, bodies, caves etc., but the synthetic dyes have caused their decline due to having variety of cheap and brighter shades.<sup>13,14</sup> With the passage of time, it has been found that their effluent load is untreatable, producing mutagenic and carcinogenic diseases and is destroying the global ecosystem.<sup>15,16</sup> The world protection associations and health conscious people have forced the industrialists to provide sustainable, green, clean, and eco-label products which have not only soothing nature but also act as remedies for diseases.<sup>17-19</sup> Previous studies revealed that their effluents could be disposed, recyclable, and even on adding in soil can play as the role of fertilizers.<sup>20</sup> Their extracts, when employed on to the fabric, the dyed fabrics show biological characteristics which are beneficial for humanity.<sup>21,22</sup> These also cover broad spectrum of colors with eye-catching shades. Hence being harmonized with nature, now the classic art of natural dyeing of fabrics, handicrafts, body painting, cosmetics, etc. is on the way.<sup>11,23</sup>

However, these colors have some issues such as poor strength and low fastness ratings.<sup>24</sup> Previously, scientists are using conventional methods to improve the colorant

strength, and are using chemical mordants to improve fastness ratings. However, these processes are less effective on the basis of cost, time, energy, and labor.<sup>25</sup> Now modern methods such as radiation methods, enzyme methods, supercritical fluid methods etc., are being employed.<sup>24,26</sup> Among these methods radiation methods particularly the utilization of microwaves rays (M.W.) are gaining fame.<sup>27</sup> These rays have ability of leveled, non-contact, and uniform heating to penetrate into the matrix.<sup>28,29</sup> These rays not only extract the colorant with a high strength but also tune the fabric surface to improve its dyeing behavior.<sup>30,31</sup> As the possible substitute of chemical mordants, now plant-based bio-mordants have been used to improve the fastness ratings with new tints.<sup>32</sup> Nylon (Polyamide) fabric is the elastic man-made fibers that contain polyamide linkage (Figure 1(c)) as functional points for bonding with colorants.

Hence keeping given advantages of M.W. rays for the extraction with high yield and bio-mordants for improving fastness ratings, this latest research studies have been designed to utilize herbal-based plants that is, harmal as source of natural dye for polyamide fiber (nylon) dyeing. Also, to author's knowledge no such a brief study under M.W., rays on nylon dyeing using Esfand (a common cheap available source of natural dye) has been done so far. Another purpose is to use herbal-based bio mordants and sustainable chemical mordants at 60°C–80°C for getting new shades with good fastness ratings.

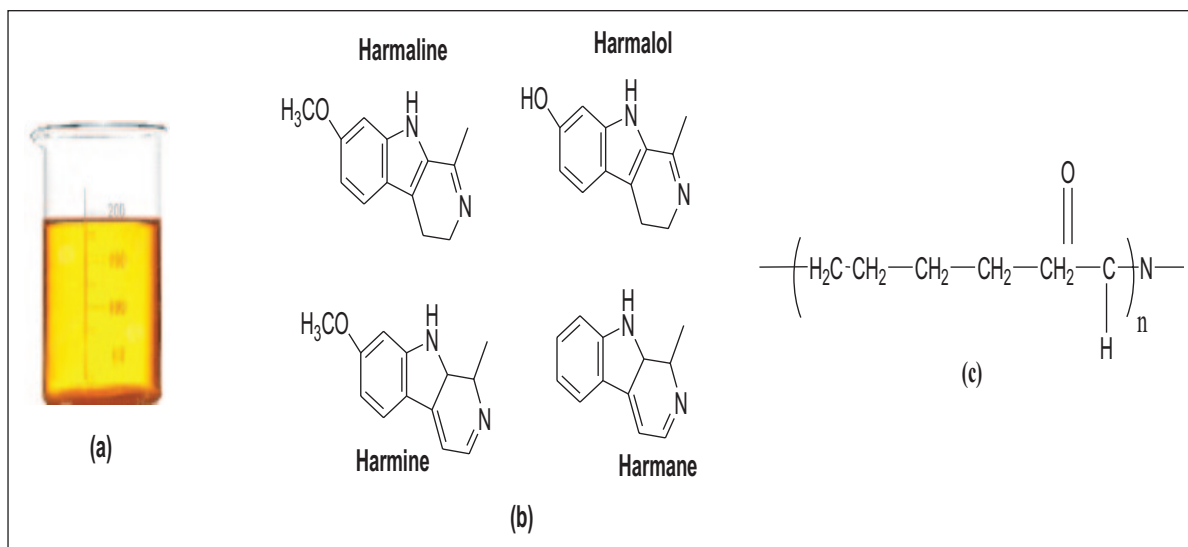
## Materials and methods

### Material collection

Using food blender, Esfand seeds (*P. harmala*) obtained from market were finely grounded and sieved for further Isolation process. Bio-mordants were also treated in same way as discussed above and stored for further shade making process. Nylon (polyamide) fabric was pretreated with neutral soap at 60°C for 30 min. to remove any direct and used for dyeing process.

### Dye extraction and irradiation process

Water solubilized extract (aqueous) was prepared by heating fine powder (4 g) at boiling with 100 mL of water. For acid solubilized extract, acidic solution was prepared by mixing 2 mL of conc. HCl with 98 mL of water (2% HCl, V/V), where the rest of extraction process employed was same. Nylon (polyamide) fabric, water (aqueous), and acid solubilized filtrates (acidic) were given MW-rays treatment up to 10 min., with an interval of 2 min. Non treated extracts were employed to dye non-treated (polyamide) nylon fabric at 80°C for 45 min keeping filtrate to (polyamide) nylon fabric ratio (F:N) of 25:1. The rest of experiments displayed in Table 1 were carried out at selected conditions.<sup>33</sup>



**Figure 1.** Harmal seeds extract (a), Alkaloids of Harmal (b) & Functional Unit of nylon (c).

**Table 1.** Microwave-assisted extraction and dyeing conditions for coloration of polyamide (nylon) fabric from *Esfand* seeds (*P. harmala*).

Fabric used	Sample code	Microwave irradiation time	Dyeing conditions
Nylon	NRE/NRNF	2–10 min.	30 mL acid extract of pH=8 using 4 g/100 mL of Table salt for 45 min. at 65°C
	RE/NRNF	2–10 min.	
	RE/RNF	2–10 min.	
	NRE/RNF	2–10 min.	
	MAD	2–10 min.	

NRE: non-M.W. Treated extract; RE: M.W. Treated extract; RNF: M.W. Treated nylon fabric; NRNF: non-M.W. Treated nylon fabric; MAD: Bio-dyeing under M.W. Treatment.

### Optimization of dyeing conditions

On selection of extraction media and M.W. rays time, a plan of 32 experiments was arranged on varying the levels of extract volume, extract pH, and salt concentration contact parameters (dyeing temperature and time). For the purpose, response surface methodology (R.S.M.) as statistical analysis has been utilized to conduct experiments for observing significance of results. The detail of 32 experiments has been displayed in Table 2.

### Shade development treatment

In bio-dyeing, treatment of fabric with either salts or natural plant materials (biomolecules) before and after dyeing at given temperature has been done to get colorfast shades. Here, 1–5 g/100 mL of chemical anchors and 1–5 g/100 mL extracts of plants have been utilized at 60°C–80°C before and after dyeing at selected conditions. The process of making colorfast shades has been taken from already published work of Adeel et al.<sup>9</sup> and Habib et al.<sup>34</sup>

### Evaluation of dyed and un-dyed fabrics

For physic chemical analysis, the nylon fabric before and after M.W. rays treatment for 4 min. was subjected to SEM (SEM-model Tescan; 5kV) and FTIR (Perkin Elmer, USA). The data after assessment was analyzed statistically using table design. All the dyed fabrics were assessed for color analysis through data color (SF 600 USA). The fabrics mordanted at given temperature before and after dyeing were assessed for fastness ratings as per ISO standards for light, washing crocking and dry cleaning.

## Results and discussion

### Selection of extraction and irradiation condition

Exploration of plants having biological and claims characteristics useful for the global health using ecofriendly, green, sustainable techniques is currently the warm topic for all walks of life.<sup>22,35</sup> The results given in Figure 2(a) show that the irradiation of aqueous extract for 6 min. have caused good color yield by rupturing of cell wall of the

**Table 2.** Central Composite Design with six center runs for optimization of dyeing variables using irradiated acidic extract of *Esfand (P. harmala)* through.

Experiment no.	A	B	C	D	E	Response	Experiment no.	A	B	C	D	E	Response
1	4	30	45	45	4	K/S	17	2	40	55	55	3	K/S
2	8	30	45	45	2	K/S	18	10	40	55	55	3	K/S
3	4	50	45	45	2	K/S	19	6	20	55	55	3	K/S
4	8	50	45	45	4	K/S	20	6	60	55	55	3	K/S
5	4	30	65	45	2	K/S	21	6	40	35	55	3	K/S
6	8	30	65	45	4	K/S	22	6	40	75	55	3	K/S
7	4	50	65	45	4	K/S	23	6	40	55	35	3	K/S
8	8	50	65	45	2	K/S	24	6	40	55	75	3	K/S
9	4	30	45	65	2	K/S	25	6	40	55	55	1	K/S
10	8	30	45	65	4	K/S	26	6	40	55	55	5	K/S
11	4	50	45	65	4	K/S	27	6	40	55	55	3	K/S
12	8	50	45	65	2	K/S	28	6	40	55	55	3	K/S
13	4	30	65	65	4	K/S	29	6	40	55	55	3	K/S
14	8	30	65	65	2	K/S	30	6	40	55	55	3	K/S
15	4	50	65	65	2	K/S	31	6	40	55	55	3	K/S
16	8	50	65	65	4	K/S	32	6	40	55	55	3	K/S

K/S: Colour Strength; A: pH; B: extract volume; C: dyeing time (min.); D: dyeing temperature ( $^{\circ}\text{C}$ ); E: salt conc. (g/100mL).

seed<sup>36,37</sup> The potent molecules interact with medium promisingly through potential mass transfer Kinetics.<sup>29</sup> Upon change of medium (Figure 2(b)) the acidic extract after exposure to microwave treatment rays for 4 min. has also given good strength onto nylon fabric. In comparison to conventional isolation process the aqueous and acidic extracts (NRE) did not give significant color strength on to unirradiated fabric (NRF). Overall, it can be seen that acidic extract (RE) irradiated for 6 min., has given excellent color strength onto surface tuned fabric (RF). This is because microwave rays are leveled heating source which penetrate the medium at molecular level, and valorize the solid-liquid interaction by transfer of heat in less time to give promising yield.<sup>38</sup> Scratching of surface of nylon (polyamide) fibers before and after M.W. treatment has also been observed (Figure 3(a) and (b)) which has added value in rising tint strength.<sup>39-41</sup> The important thing which has been found via FTIR spectra (Figure 4(a) and (b)) is that position of functional peaks of amido-linkage (binding site of nylon) has been remained the same, even after microwave treatment up to 4 min. Hence M.W. rays treatment has not only enhanced color strength in acidic medium but also without altering nylon (polyamide) fabric chemistry has improved its dye uptake potential.

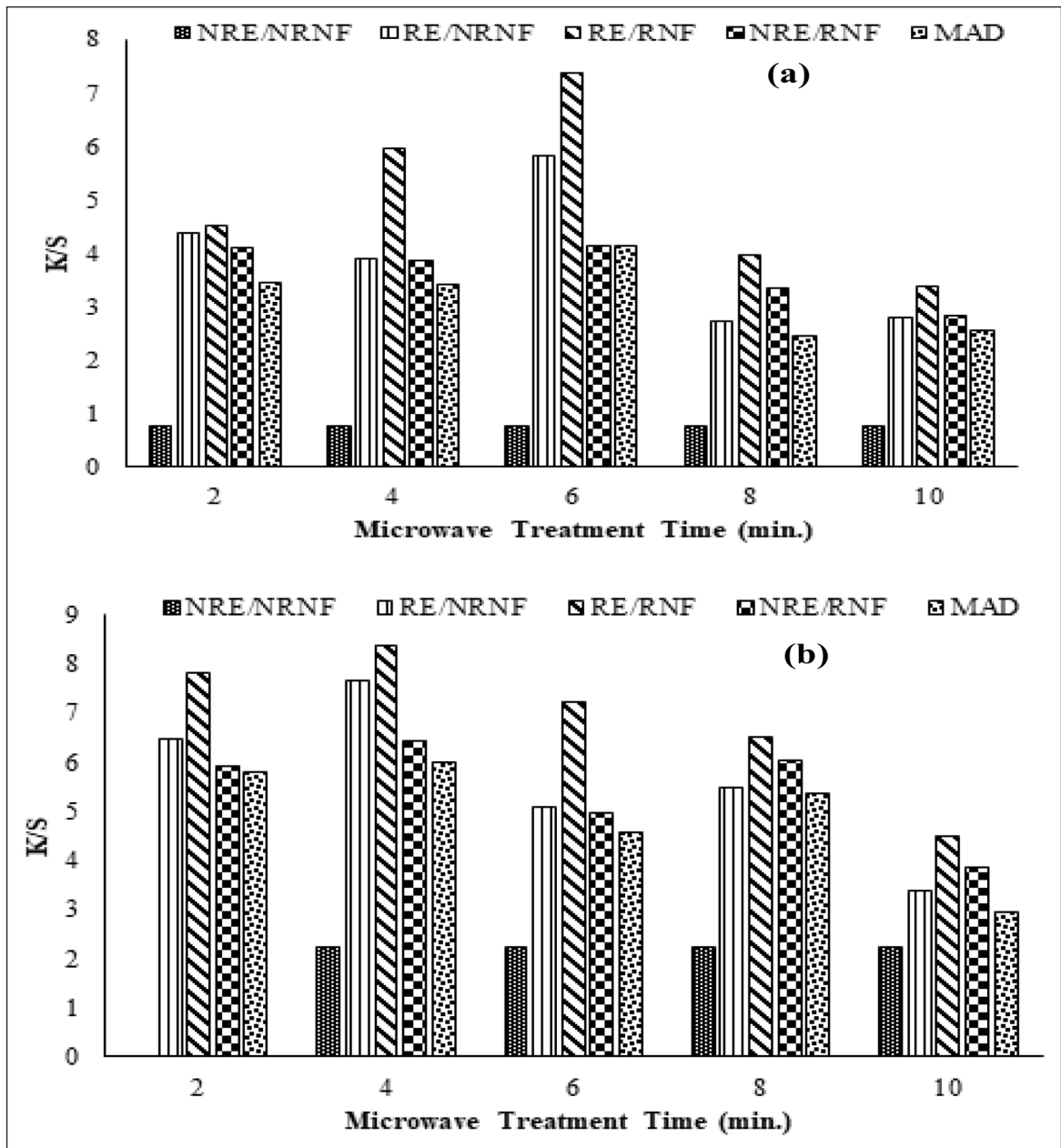
### Selection of dyeing parameters

Literature studies show that for biocoloration of nylon (polyamide) fabric, both time and heat plays their roles but M.W. rays treatment has reduced their levels. The results show that at  $55^{\circ}\text{C}$  for 55 min. the dyed fabrics were of high in strength to get excellent results (Figure 5(a) and (b)). The addition of Table salt as leveling agent that adds value

in bio-coloration by giving maximum evolution of colorant (at 8 pH) toward fabric from dye bath. The results given in Figure 5(c) shows that the utilization of 4 g/100 mL of Table salt has given good results. Hence M.W. rays treatment has reduced the level which is the proof by their study that this treatment is cost, time, and energy effective in nature<sup>28,29</sup> Statistically for the detailed study of the relationship between the experimental parameters and the K/S value, we initially considered quadratic effects and second order interaction effects of the factors through second-order polynomial model. We improved the model adequacy and the significance of effects by removing some trials which produced outlier observations and eliminated some insignificant terms from the model (Table 3). It can be seen that the role of extract pH and volume are highly significant ( $p$ -value=0.000) for K/S, where as Time is significant at 5% level of significance and Temperature and Salt are significant at 10% level of significance. The model summery shows quite good values. The final model shows high value of coefficient of determination ( $R^2=97.16\%$ ) indicating a good model fit. The results presented in Tables 3 and 4 are further reinforced by presenting contribution of each term in the model in the model by showing coefficients and their significance in the form of  $p$ -value. This has been improved by statistical analysis also (Table 2). Hence statistically it has also been found that dyeing variable should also be considered during biocoloration of nylon (polyamide) with harmful extracts.

### Optimization of shade development process

Mordanting is an essential parameter for the natural dyeing particularly the utilization of plant extract having unique



**Figure 2.** Application of Esfand water solubilized (aqueous=a) and acid solubilized (acidic=b) filtrates for bio-dyeing of polyamide (nylon) fabric before and after M.W. rays.

herbal characteristics called bio-mordants. In this study mordanting at various temperatures has been done followed by dyeing of nylon using statistically optimized dyeing conditions. The results reveal that dyeing of nylon after mordanting with  $Al^{3+}$  (2%),  $Fe^{2+}$  (3%), and T.A (3%) as pre chemical mordant at 60°C and Acacia (3%), pomegranate (2%), Turmeric (2%), and Pine nut (Pn) hull extracts as pre bio-mordant has given high color yield (Figure 6(a)). Likewise  $Al^{3+}$  (2%) and (2%) T.A,  $Fe^{2+}$  (3%) as post chemical while acacia (2%), pomegranate (2%) pinenut hull and turmeric (3%), as post-bio-mordanting at 60°C has given excellent color strength (Figure 6(b)). On mordanting

at 70°C the utilization of  $Al^{3+}$  (3%),  $Fe^{2+}$  (2%), and T.A (3%) where pomegranate (2%), Acacia (3%), Turmeric (2%), and pine nut hull extracts as pre mordants has given excellent shade strength (Figure 7(a)), whereas  $Al^{3+}$  (4%), 3% of  $Fe^{2+}$  (3%), and T.A (2%), as post chemical and the application of acacia (2%), pomegranate (2%), and pinenut hull (2%) and turmeric (3%) as post bio-mordants have given high color yield (Figure 7(b)). On mordanting at 80°C the utilization of  $Al^{3+}$  (3%),  $Fe^{2+}$  (3%), T.A (4%) as in same way pre chemical mordant where pomegranate (3%) and pine nut hull, Acacia (2%) and Turmeric and extracts as pre biomordant has given excellent shade

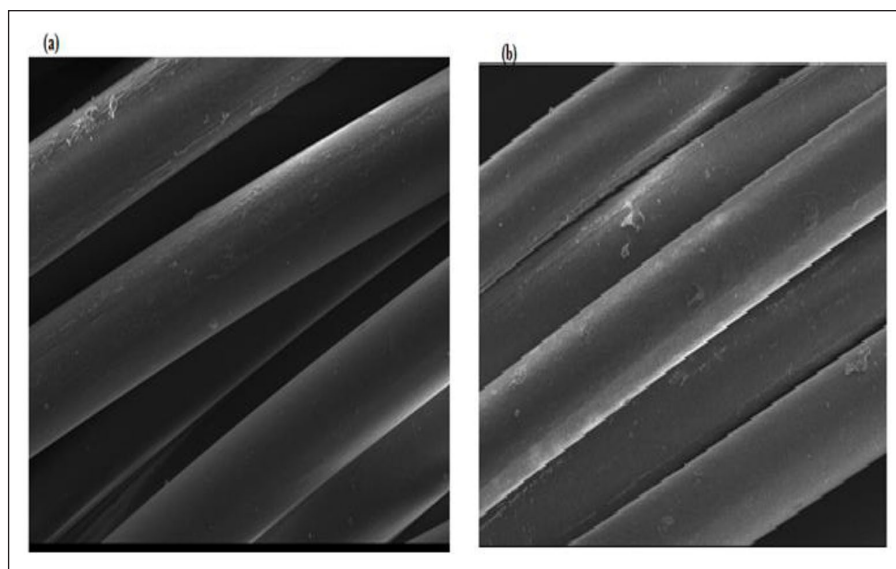


Figure 3. SEM Images of control (a = un-irradiated) and treated nylon (polyamide) fabric (b = M.W.).

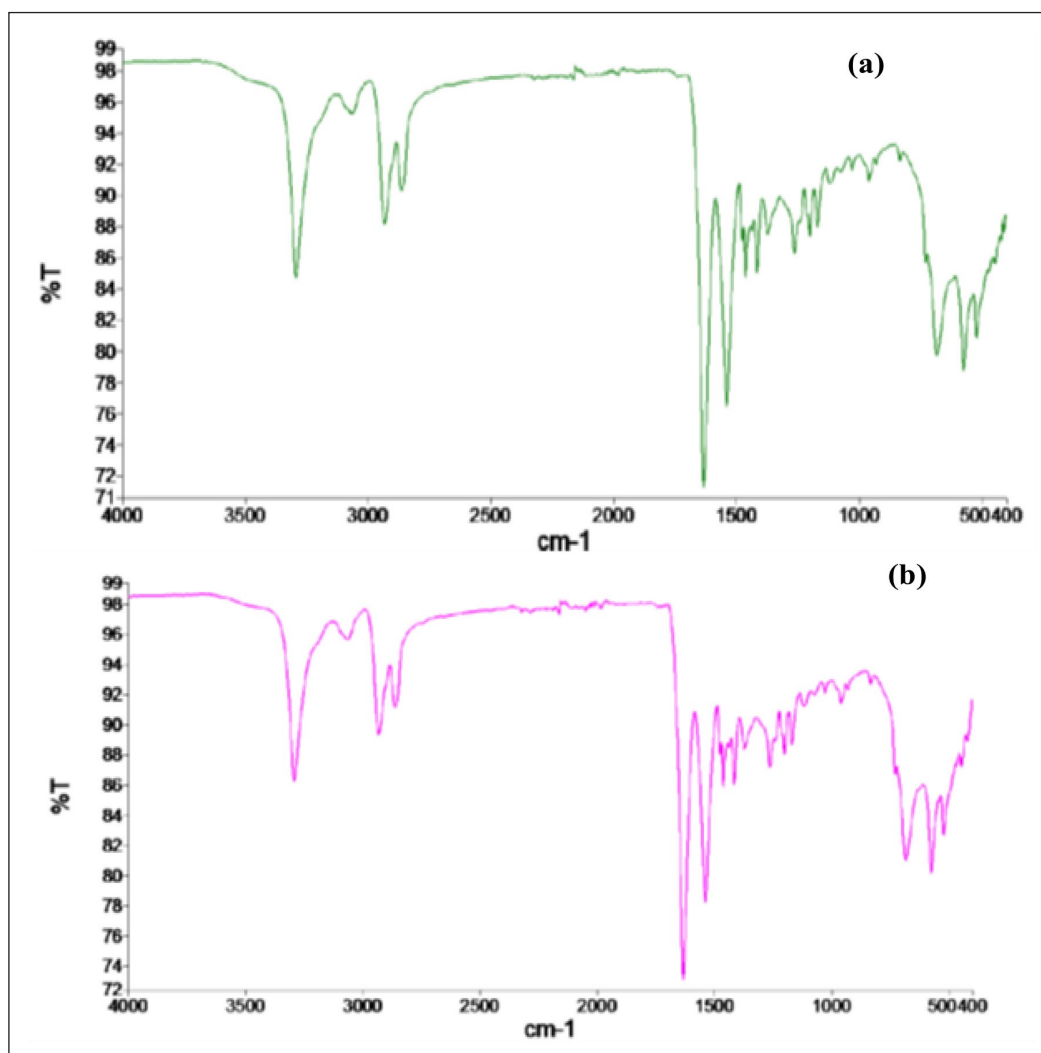
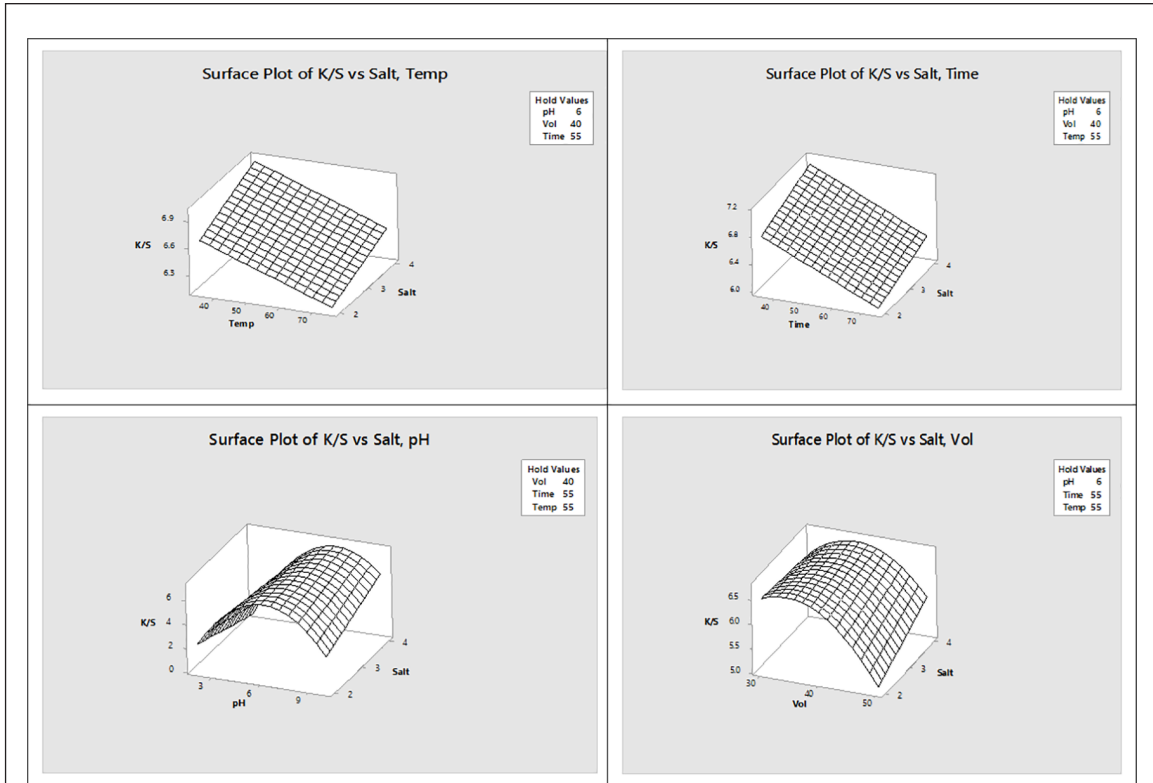
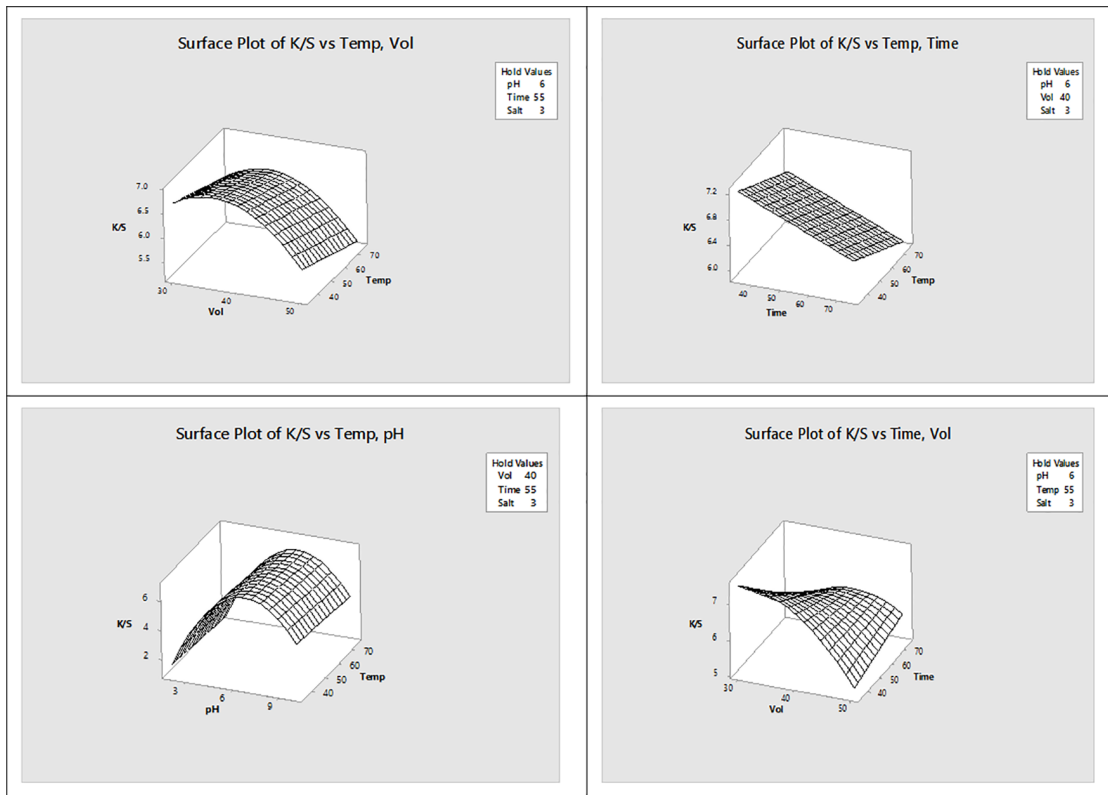


Figure 4. Spectral images of functional peaks of control (a = un-irradiated) and treated nylon (polyamide) fabric (b = M.W.).



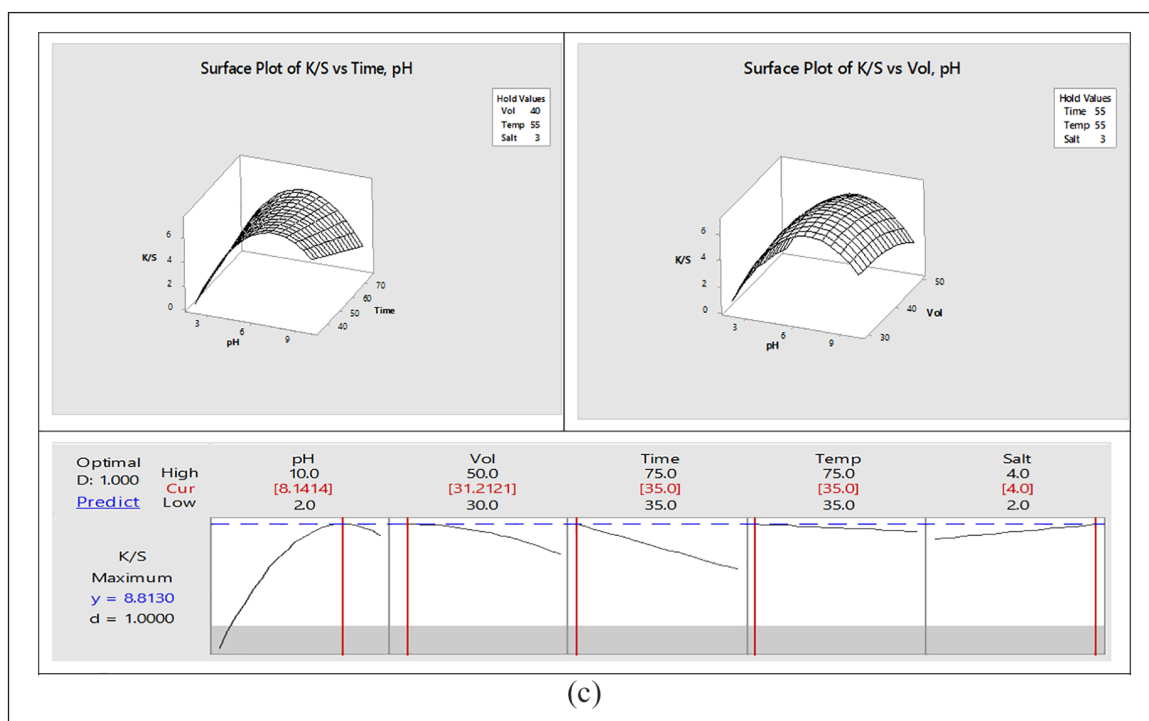
(a)



(b)

(Continued)

Figure 5. (Continued)



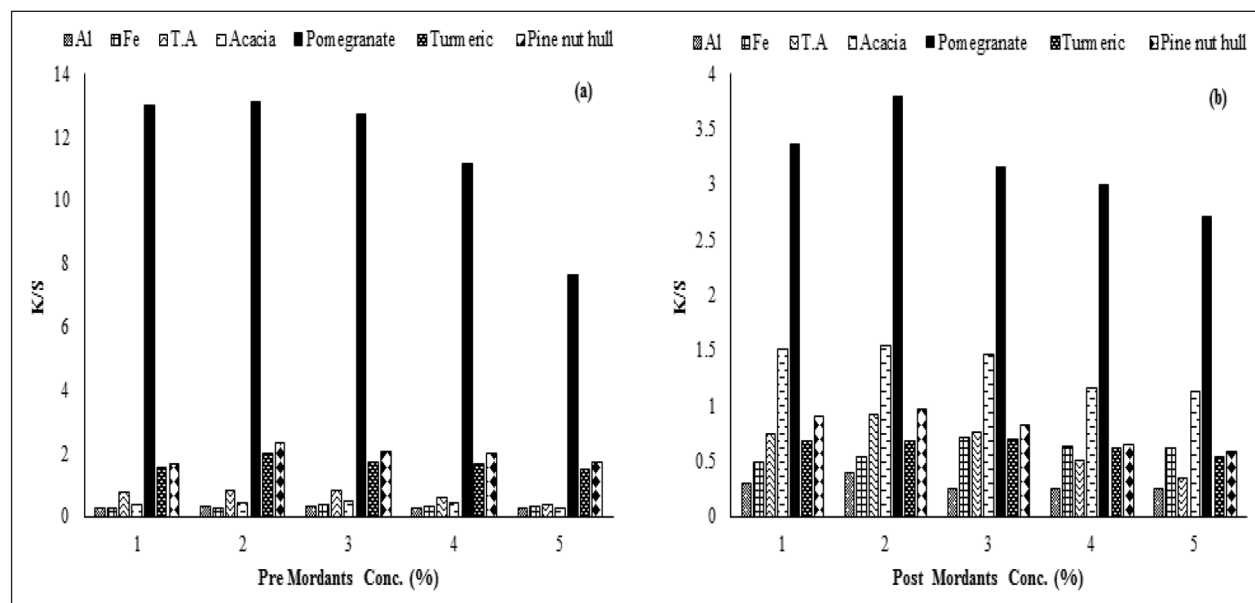
**Figure 5.** (a) 3-D response surfaces of *P. harmala* K/S versus two parameters from Salt, Temperature, Time, pH, and Volume, and holding the remaining three parameters at middle level, using Response Surface Regression. (b) 3-D response surfaces of *Esfand* K/S versus two parameters from Salt, Temperature, Time, pH, and Volume, and holding the remaining three parameters at middle level, using Response Surface Regression. (c) Graphical Representation of Optimization of *Esfand* K/S versus Temperature and salt using Response Surface Regression.

**Table 3.** Response Surface Regression: K/S versus pH, Volume, Time, Temperature, and Salt using ANOVA.

Source	DF	Adj SS	Adj MS	F-value	p-Value
Model	12	53.2379	4.4365	34.18	0.000
Linear	5	16.9937	3.3987	26.18	0.000
pH	1	11.5405	11.5405	88.90	0.000
Volume	1	3.7065	3.7065	28.55	0.000
Time	1	0.9084	0.9084	7.00	0.021
Temperature	1	0.4232	0.4232	3.26	0.096
Salt	1	0.4152	0.4152	3.20	0.099
Square	2	26.4717	13.2359	101.96	0.000
pH × pH	1	23.4513	23.4513	180.65	0.000
Volume × Volume	1	2.1814	2.1814	16.80	0.001
2-way interaction	5	9.7725	1.9545	15.06	0.000
pH × Volume	1	0.5270	0.5270	4.06	0.067
pH × Time	1	1.8218	1.8218	14.03	0.003
pH × Salt	1	4.9312	4.9312	37.99	0.000
Volume × Time	1	1.6591	1.6591	12.78	0.004
Volume × Salt	1	0.8334	0.8334	6.42	0.026
Error	12	1.5578	0.1298	—	—
Lack-of-fit	10	1.4225	0.1423	2.10	0.365
Pure error	2	0.1353	0.0676	—	—
Total	24	24	54.7957	—	—
Model summary	S	R <sup>2</sup>	R <sup>2</sup> (adj)	R <sup>2</sup> (pred)	—
	0.360298	97.16%	94.31%	84.98%	—

**Table 4.** Coded Coefficients of dyeing parameters second order polynomial regression model.

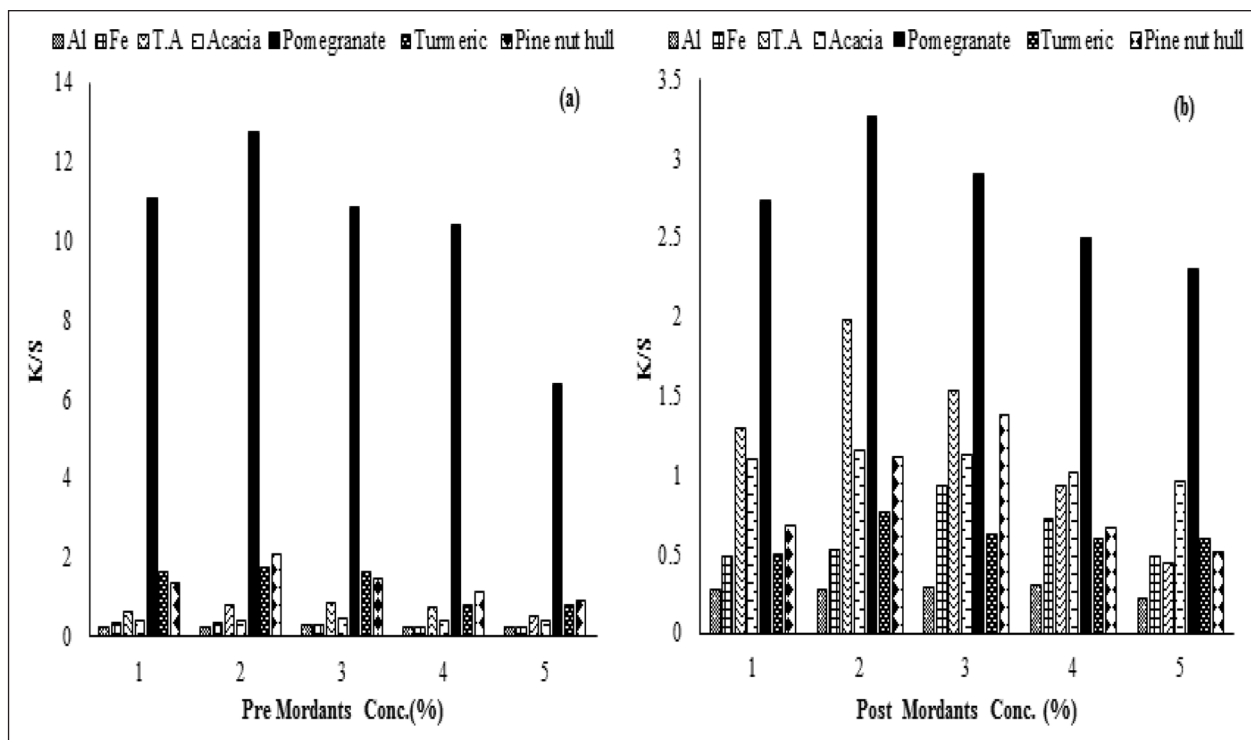
Term	Effect	Coef	SE Coef	T-value	p-Value
Constant		6.567	0.136	48.23	0.000
pH	2.774	1.387	0.147	9.43	0.000
Volume	-1.925	-0.963	0.180	-5.34	0.000
Time	-0.778	-0.389	0.147	-2.65	0.021
Temperature	-0.531	-0.266	0.147	-1.81	0.096
Salt	0.644	0.322	0.180	1.79	0.099
pH × pH	-7.766	-3.883	0.289	-13.44	0.000
Volume × Volume	-4.930	-2.465	0.601	-4.10	0.001
pH × Volume	-1.452	-0.726	0.360	-2.01	0.067
pH × Time	-2.699	-1.350	0.360	-3.75	0.003
pH × Salt	4.441	2.221	0.360	6.16	0.000
Volume × Time	2.576	1.288	0.360	3.58	0.004
Volume × Salt	1.826	0.913	0.360	2.53	0.026

**Figure 6.** Dyeing of M.W. treated polyamide (nylon) fabric after (a=pre) and before (b=post) mordanting at 60°C using acid solubilized filtrates.

strength (Figure 8(a)). Also the application of  $Al^{3+}$  (2%),  $Fe^{2+}$  (2%), and T.A. (3%) as post chemical mordant and acacia (Ac=4%), pomegranate (Pm=3%), pinenut (4%=Pn) hull, and turmeric (Tm=4%) as post bio-mordants has given good result (Figure 8(b)). Overall, in comparison at 60°C, the utilization of pomegranate (Pm=2%) as pre bio-mordant and T.A. (2%) as post chemical mordants has given good color strength, while at 70°C pomegranate (Pm=2%) as pre bio-mordant and T.A., (2%) as post-chemical has given good color strength. However, at 80°C pomegranate (Pm=3%) as pre bio-mordant and T.A., (3%) as post chemical mordant has given promising results. The color coordinates given in Tables 5 to 7 reveal that

mordanting of fabric before dyeing has produced brighter shade with reddish yellow hues. The fabric dyed after application of pomegranate (Pm=2%) at 60°C is darker ( $L^*=56.79$ ) having reddish yellow tone ( $a^*=8.5$ ;  $b^*=35.66$ ). The fabric dyed before application of Tannic acid (T.A.=2%) at 70°C is brighter in shade ( $L^*=77.13$ ) having less reddish yellow tone ( $a^*=4.56$ ;  $b^*=26$ ).

This high color strength was observed due to formation of coordinate covalent bond by metals with dye onto surface-tuned polyamide (nylon) fabric through metal-dye complex followed by dyeing at optimum temperature.<sup>42,43</sup> Here the reduction power of metal ( $Al^{3+}$ ,  $Fe^{2+}$ ) or -OH of tannic acid as functional site works were to interact



**Figure 7.** Dyeing of M.W. treated polyamide (nylon) fabric after (a = pre) and before (b = post) mordanting at 70°C using acid solubilized filtrates.

with fabric and colorant. Using plant extract, the  $-OH$  of quercetin of acacia (Ac), tannin of pomegranate (Pm), curcumin of turmeric (Tm), and catechin of pine nut (Pn) hulls has played their role by forming the additional H-bonding with amido-linkage of polyamide (nylon) and  $-OH$  of colorant.<sup>44-46</sup> Hence at optimal temperature using M.W. treated extract, the observed amount of mordants have developed not only new tints but also improved color strength with color fast shades (Figure 9(a) and (b)) is showing the suggested interaction of sustainable chemical and bio-mordants with fabric and dye.

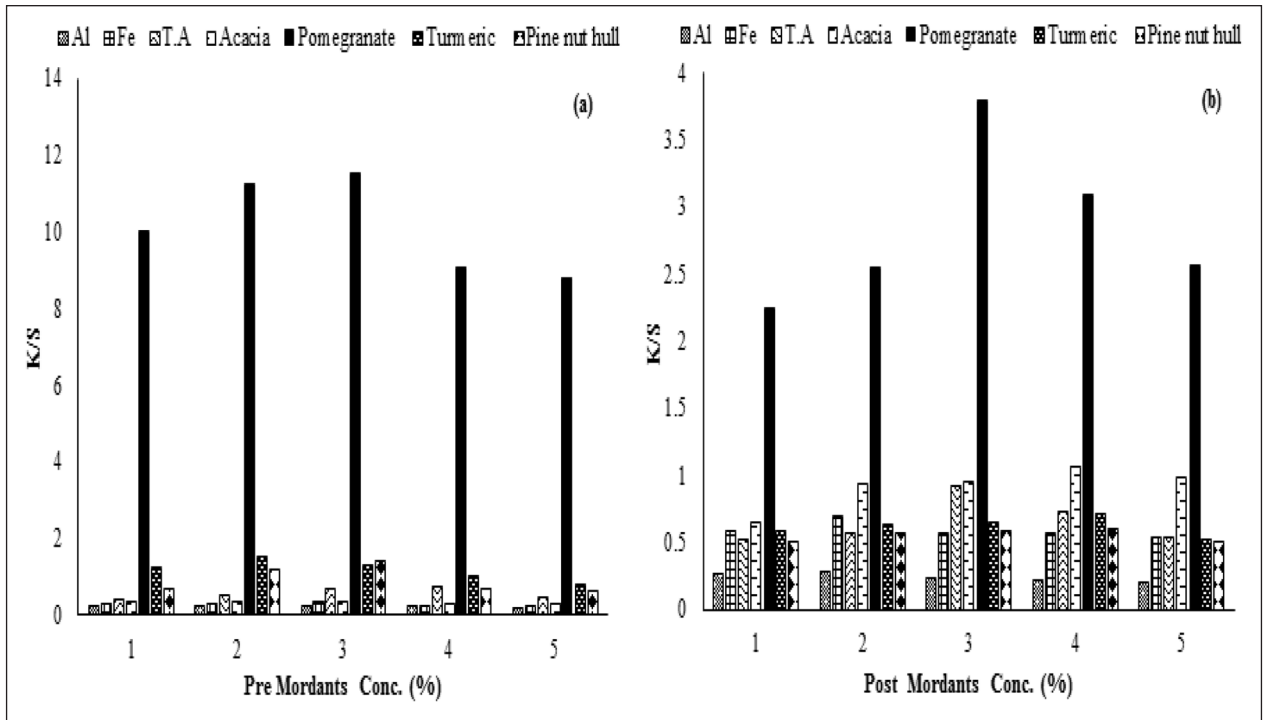
### Assessment of colorfastness ratings

The colorfastness properties ratings have been assessed at Gray-scale using standard ISO methods. The results given in Tables 5 to 7 reveal that good to excellent fastness ratings have been obtained during dyeing of surface modified polyamide (nylon) fabric with acid solubilized extract of Esfand (harmal). The firm metal complex with dye and additional H-bonding developed by  $-OH$  of bio-mordants, conjugation system presents in bio-molecules and microwave treatment to the colorant are possible aspects that have played their role in developing color fastness shades. Upon exposure to heat, light, washing, crocking less color is

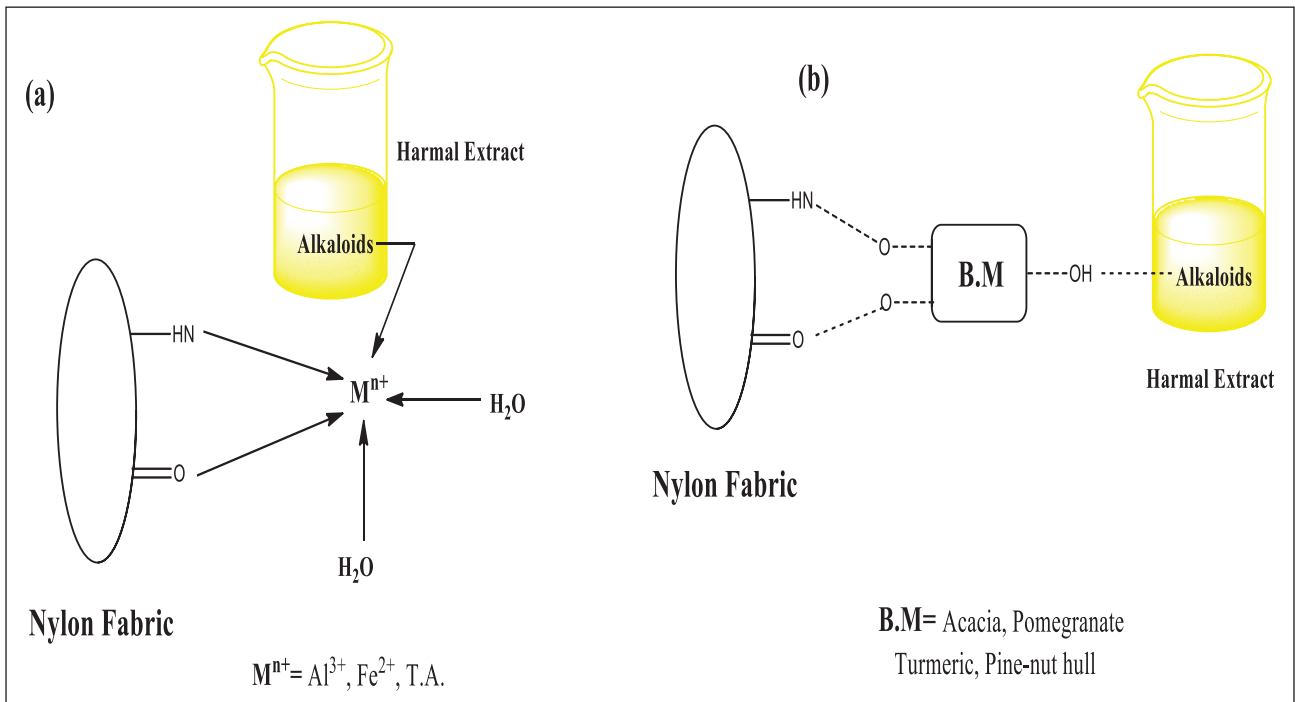
detached and good to excellent rating is found.<sup>30,47,48</sup> Overall bio-mordants have given better effects. Hence microwave treatment has an excellent potential to isolate colorant from Harmal seeds, and tune to the fabric to reduce dyeing variables. Additionally, bio-mordants as an alternative to their chemical counterparts have made process sustainable eco-friendly and soothing in nature.<sup>49-52</sup>

### Conclusion

The aftereffects of carcinogens have now forced the people to swing toward green sustainable products. Esfand or harmal seeds (*P. harmala*) as bio-dye source has been valorized under M.W. treatment and plant based antiviral and anti-oxidants molecules as bio-anchors have been utilized to make process more colorfast and sustainable. It has been found the application of M.W. ray treatment for 4 min., to 30 mL of extract of 8 pH containing 4 g/100 mL of Table salt as leveling agent has given desired results when employed at 55°C for 55 min. Statistical analysis of dyeing variables through R.S.M., and two way-Anova shows that the effect of these variables has been observed highly significant. The utilization of pomegranate extract as bio-mordant and tannic acid as sustainable chemical mordant has furnished colorfast shades.



**Figure 8.** Dyeing of M.W. treated polyamide (nylon) fabric after (a=pre) and before (b=post) mordanting at 80°C using acid solubilized filtrates.



**Figure 9.** Proposed interactions of chemical (a) and bio-mordants (b) with alkaloids of colorant and nylon fabric.

**Table 5.** Color characteristics of dyed fabric mordanted at 60°C using harmal extract.

Mordant Conc. %	L*	a*	b*	L.F	W.F		RF		DCF
					C.C	C.S	DRF	WRF	
Ctrl	60.69	10.75	29.62	3/4	3/4	3/4	3/4	3	3/4
2% Al (Pre)	80.30	7.93	7.22	5	4/5	4/5	5	4/5	4/5
2% Al (Post)	84.1	-2.03	19.98	5	4/5	4/5	5	4/5	4/5
2% Fe (Pre)	82.18	2.86	4.27	5	4/5	4/5	5	4/5	4/5
3%Fe (Post)	73.09	8.32	11.62	5	4/5	4/5	5	4/5	4/5
3% T.A (Pre)	79.71	3.81	16.47	5	4/5	4/5	5	4/5	5
2%T.A. (Post)	81.56	1.24	19.06	5	4/5	4/5	5	4/5	5
3% Ac (Pre)	78.14	3.93	9.93	5	4/5	4/5	4/5	4/5	5
2% Ac (Post)	73.14	5.89	20.08	5	4/5	4/5	4/5	4/5	5
2% Pm (Pre)	56.79	8.35	35.66	5	4/5	4/5	4/5	4/5	4/5
2% Pm (Post)	76.21	-0.33	28.38	5	4/5	4/5	5	4/5	4/5
2% Tr (Pre)	63.97	20.77	18.44	5	4/5	4/5	4/5	4	4/5
3%Tr. (Post)	68.05	15.86	20.91	5	4/5	4/5	4/5	4	4/5
2%Pn (Pre)	72.69	4.13	27.24	5	4/5	4/5	4/5	4	4/5
2%Pn (Post)	72.43	2.72	14.50	5	4/5	4/5	4/5	4/5	4/5

L.F: light fastness; W.F: washing fastness; c.c: color change; c.s: color stain; RF: Rubbing fastness; DRF: dry rubbing fastness; WRF: wet rubbing fastness; DCF: dry clean fastness; Tr: turmeric; Pn: pinenut hulls, Pm: pomegranate; Ac: acacia.

**Table 6.** Color characteristics of dyed fabric mordanted at 70°C using harmal extract.

Mordant Conc. %	L*	a*	b*	L.F	W.F		RF		DCF
					C.C	C.S	DRF	WRF	
Ctrl	60.69	10.75	29.62	3/4	3/4	3/4	3/4	3	3/4
3% Al (Pre)	80.83	5.21	7.73	5	4/5	4/5	5	4/5	5
4% Al (Post)	84.7	-2.09	14.27	5	4/5	4/5	5	4/5	5
3% Fe (Pre)	81.40	4.13	4.87	5	4/5	4/5	5	5	5
3% Fe (Post)	70.44	5.03	14.33	5	4/5	4/5	5	5	4/5
3% T.A. (Pre)	78.60	6.53	16.18	5	4/5	4/5	5	4/5	4/5
2% T.A. (Post)	77.13	4.56	26.48	5	4/5	4/5	4/5	4/5	4/5
3% Ac (Pre)	79.76	3.64	10.00	5	4/5	4/5	4/5	4/5	5
2% Ac (Post)	72.53	6.80	15.89	5	4/5	4/5	5	4/5	4/5
2% Pm (Pre)	56.54	8.48	34.49	5	4/5	4/5	5	4/5	5
2% Pm (Post)	78.71	-1.55	27.69	5	4/5	4/5	5	4/5	5
2% Tr. (Pre)	66.16	19.36	18.88	5	4/5	4/5	4/5	4	4/5
2% Tr. (Post)	78.01	2.87	13.81	5	4/5	4/5	4/5	4	4/5
2% Pn (Pre)	57.72	10.57	12.87	5	4/5	4/5	4/5	4/5	4/5
3% Pn (Post)	73.57	2.68	28.04	5	4/5	4/5	4/5	4/5	5

L.F: light fastness; W.F: washing fastness; c.c: color change; c.s: color stain; RF: Rubbing fastness; DRF: dry rubbing fastness; WRF: wet rubbing fastness; DCF: dry clean fastness; Tr: turmeric; Pn: pinenut hulls, Pm: pomegranate; Ac: acacia.

The addition of green and clean processes for the extraction of bio-colors and the application of these sustainable and eco-friendly mordants for improving shade quality has made this act of cultural heritage more value added. For future studies, such new anti-viral, antibacterial, and anti-oxidant based herbal plants should be assessed for their coloring behavior and additionally, plant based herbal mordants should be used to make process more sustainable and ayurvedic.

### Acknowledgement

We are thankful to Mr. Zafar Iqbal Manager (R &D) Noor Fatima Fabrics Faisalabad Pakistan and Mr. Muhammad Abbas Chief Executive Harris Dyes and Chemicals Faisalabad Pakistan for providing us the technical guidance at lab scale for smooth running of this M.Phil. Studies. We are also thankful to National Engineering laboratory of Modern Silk, College of Textile and Clothing Engineering, Soochow University, Suzhou, China for providing us pre-treated nylon fabric.

**Table 7.** Color characteristics of dyed fabric mordanted at 80°C using harmal extract.

Mordant Conc. %	L*	a*	b*	L.F	W.F		RF		DCF
					C.C	C.S	DRF	WRF	
Ctrl	60.69	10.75	29.62	3/4	3/4	3/4	3/4	3	3/4
3% Al (Pre)	82.63	3.66	5.88	5	4/5	4/5	5	4/5	4/5
2% Al. (Post)	84.61	-1.65	11.88	5	4/5	4/5	5	4/5	5
3% Fe (Pre)	78.50	5.23	5.42	5	4/5	4/5	5	5	4/5
2% Fe. (Post)	75.08	1.31	16.36	5	4/5	4/5	5	5	5
4% T.A (Pre)	74.72	4.18	11.15	5	4/5	4/5	4/5	4/5	4/5
3%T.A. (Post)	80.60	1.59	18.89	5	4/5	4/5	4/5	4/5	4/5
4% Pn (Post)	80.24	0.32	16.52	5	4/5	4/5	4/5	4/5	4/5
2% Ac (Pre)	80.98	2.85	8.90	5	4/5	4/5	5	4/5	4/5
4% Ac. (Post)	74.23	5.14	15.86	5	4/5	4/5	5	4/5	4/5
3% Pm (Pre)	58.75	7.63	34.49	5	4/5	4/5	4/5	4/5	4/5
3% Pm (Post)	76.13	0.02	28.32	5	4/5	4/5	4/5	4/5	4/5
2% Tr (Pre)	63.28	21.92	13.64	5	4/5	4/5	4/5	4	4/5
4% Tr. (Post)	78.77	3.59	16.08	5	4/5	4/5	4/5	4	4/5
3% Pn (Pre)	67.03	7.76	13.09	5	4/5	4/5	4/5	4/5	4/5

L.F: light fastness; W.F: washing fastness; c.c: color change; c.s: color stain; RF: Rubbing fastness; DRF: dry rubbing fastness; WRF: wet rubbing fastness; DCF: dry clean fastness; Tr: turmeric; Pn: pinenut hulls, Pm: pomegranate; Ac: acacia.

### Declaration of conflicting interests

The author(s) declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

### Funding

The author(s) received no financial support for the research, authorship, and/or publication of this article.

### ORCID iDs

Shahid Adeel  <https://orcid.org/0000-0002-5516-7882>

Mozhgan Hosseinnezhad  <https://orcid.org/0000-0003-3351-0157>

### Supplemental material

Supplemental material for this article is available online.

### References

- Liu J, Lin X and Liang H. Dyed fabrics modified via assembly with phytic acid/berberine for antibacterial, UV resistance, and self-cleaning applications. *J Eng Fiber Fabr* 2019; 14: 1–10.
- Najeebullah S, Shinwari ZK, Jan SA, et al. Ethno medicinal and phytochemical properties of genus *Allium*: a review of recent advances. *Pak J Bot* 2021; 53(1): 135–144.
- Ilyas A, Tanvir R and Rehman Y. Indo-pak medicinal plants and their endophytes: an emphasis on nutraceutical and bioactive potential. *Endophytes* 2021; 26: 51–70.
- Özomay M and Akalın M. Optimization of fastness properties with gray relational analysis method in dyeing of hemp fabric with natural and classic mordant. *J Nat Fibers*. Epub ahead of print 5 November 2020. DOI: 10.1080/15440478.2020.1837328.
- Chandru P and Praveena R. Organic hybrid pigments from *Crotalaria globosa* for cotton dyeing – a sustainable approach. *Mater Today Proc* 2021; 47(9): 1773–1776.
- Shahrajabian MH, Sun W and Cheng Q. Improving health benefits with considering traditional and modern health benefits of *Peganum harmala*. *Clin Phytosci* 2021; 7(1): 1–9.
- Shaheen HA and Issa MY. In vitro and in vivo activity of *Peganum harmala* L. Alkaloids against phytopathogenic bacteria. *Sci Hort* 2020; 264: 108940.
- Singhai A and Patil UK. Amelioration of oxidative and inflammatory changes by *Peganum harmala* seeds in experimental arthritis. *Clin Phytosci* 2021; 7(1): 1–11.
- Adeel S, Rehman FU, Zuber M, et al. Environmental friendly application of ultrasonic rays for extraction of natural colorant from Harmal (*P. Harmala*) for dyeing of bio-mordanted silk. *J Eng Fibers Fabr* 2021; 16: 15589250211063868.
- Haji A. Wool dyeing with harmal seed as a natural dye: investigation of influencing factors using response surface methodology. *J Color Sci Technol* 2019; 13(2): 131–140.
- Rather LJ, Zhou Q and Li Q. Re-use of *Cinnamomum camphora* natural dye generated wastewater for sustainable UV protective and antioxidant finishing of wool fabric: effect of Fe(II) sulfate. *Sustain Chem Pharm* 2021; 21: 100422.
- Mongkholrattanasit R, Nakpathom M and Vuthiganond N. Eco-dyeing with biocolorant from spent coffee ground on low molecular weight chitosan crosslinked cotton. *Sustain Chem Pharm* 2021; 20: 100389.
- Amutha K and Annapoorani SG. Natural dye extraction from agro-waste and its application on textiles. *Asian Dyers* 2019; 16(1): 35–39.
- Sanku LP and Padma A. Identifying the viability of natural dye sources from India: a review. *Int Res J Mod Eng Technol Sci* 2021; 2: 736–744.
- Özomay M and Özomay Z. The effect of temperature and time variables on printing quality in sublimation transfer

- printing on nylon and polyester fabric. *Eur J Sci Technol* 2021; 23: 882–891.
16. Sowndharya B and Giri DRS. Extraction of natural dye from *Tagetes erecta* Linn. flowers and evaluation of cotton fabric dyeing using different chemical mordants. *World J Sci Res* 2020; 5(1): 10–17.
  17. Özomay M, Güngör F and Özomay Z. Determination of optimum dyeing recipe with different amount of mordants in handmade cotton fabrics woven with olive leaves by grey relational analysis method. *J Text Inst*. Epub ahead of print 13 April 2021. DOI: 10.1080/00405000.2021.1908008.
  18. Thakker AM. Sustainable processing of cotton fabrics with plant-based biomaterials *Sapindus mukorossi* and *Acacia concinna* for health-care applications. *J Text Inst* 2021; 112: 718–726.
  19. Vashist P, Basak S and Ali W. Bark extracts as multifunctional finishing agents for technical textiles: a scientific review. *AATCC J Res* 2021; 8(2): 26–37.
  20. Baseri S. Natural bio-source materials for green dyeing of cellulosic yarns. *J Nat Fibers* 2021; 2021: 1–12.
  21. Verma M, Gahlot N, Singh SSJ, et al. UV protection and antibacterial treatment of cellulosic fibre (cotton) using chitosan and onion skin dye. *Carbohydr Polym* 2021; 257: 117612.
  22. Indrianingsih AW, Rosyida VT, Darsih C, et al. Antibacterial activity of *Garcinia mangostana* peel-dyed cotton fabrics using synthetic and natural mordants. *Sustain Chem Pharm* 2021; 21: 100440.
  23. Ansari NT and Iqbal S. Antibacterial efficiency of naturally occurring dyes and mordants. *Res Square* 2021; 1: 1–19.
  24. Singh M, Vajpayee M and Ledwani L. Eco-friendly surface modification of natural fibres to improve dye uptake using natural dyes and application of natural dyes in fabric finishing: a review. *Mater Today Proc* 2021; 43(5): 2868–2871.
  25. Hayat T, Adeel S, Batool F, et al. Waste black tea leaves (*Camelia sinensis*) as a sustainable source of tannin natural colorant for bio-treated silk dyeing. *Environ Sci Pollut Res Int*. Epub ahead of print 25 November 2021. DOI: 10.1007/s11356-021-17341-5.
  26. Bonifácio-Lopes T, Teixeira JA and Pintado M. Current extraction techniques towards bioactive compounds from brewer's spent grain – a review. *Crit Rev Food Sci Nutr* 2020; 60(16): 2730–2741.
  27. Amesimeku J, Fan L, Jakpa W, et al. Dyeing properties of meta-aramid fabric dyed with basic dye using ultrasonic-microwave irradiation. *J Clean Prod* 2021; 285: 124844.
  28. Ticha MB, Slama N, Dhoubi N, et al. Bark residues recovery of *Juglans regia* L for the dyeing of wool fabrics: development of microwave-assisted extraction and dyeing processes. *J Nat Fibers*. Epub ahead of print 29 March 2021. DOI: 10.1080/15440478.2021.1875370.
  29. Karadag R, Buyukakinci BY and Torgan E. Extraction and natural cotton dyeing of valonia oak and anatolian buckthorn by microwave irradiation. *J Nat Fibers* 2022; 19: 159–172.
  30. Arain RA, Ahmad F, khatri Z, et al. Microwave assisted henna organic dyeing of polyester fabric: a green, economical and energy proficient substitute. *Nat Prod Res* 2021; 35(2): 327–330.
  31. Saleh MO, El-Asasery MA, Hussein AM, et al. Microwave assisted synthesis of some azo disperse dyes part 2: eco-friendly dyeing of polyester fabrics by using microwave irradiation. *Eur J Chem* 2021; 12(1): 64–68.
  32. Hosseinneshad M, Gharanjig K, Jafari R, et al. Cleaner colorant extraction and environmentally wool dyeing using oak as eco-friendly mordant. *Environ Sci Pollut Res* 2021; 28(6): 7249–7260.
  33. Adeel S, Habib N, Batool F, et al. Eco-friendly approach towards isolation of colorant from Esfand for bio-mordanted silk dyeing. *Environ Sci Pollut Res Int* 2022; 29: 13523–13533.
  34. Habib N, Adeel S, Ali F, et al. Environmental friendly sustainable application of plant-based mordants for cotton dyeing using Arjun bark-based natural colorant. *Environ Sci Pollut Res Int* 2021; 28(38): 54041–54047.
  35. Kamboj A, Jose S and Singh A. Antimicrobial activity of natural dyes – a comprehensive review. *J Nat Fibers*. Epub ahead of print 31 January 2021. DOI: 10.1080/15440478.2021.1875378.
  36. Pinzon IAP, Razal RA, Mendoza RC, et al. Parametric study on microwave-assisted extraction of runo (*Miscanthus sinensis* Andersson) dye and its application to paper and cotton fabric. *Biotechnol Rep* 2020; 28: e00556.
  37. Suktham K, Daisuk P and Shotipruk A. Microwave-assisted extraction of antioxidative anthraquinones from roots of *Morinda citrifolia* L. (Rubiaceae): errata and review of technological development and prospects. *Sep Purif Technol* 2021; 256: 117844.
  38. Taqi A, Farcot E, Robinson JP, et al. Understanding microwave heating in biomass-solvent systems. *Chem Eng J* 2020; 393: 124741.
  39. Elnagar K, AbouElmaaty T and Raouf S. Dyeing of polyester and polyamide synthetic fabrics with natural dyes using ecofriendly technique. *J Textile* 2014; 2014: 1–8.
  40. Ebrahimi I and Parvinezadeh Gashti M. Extraction of polyphenolic dyes from henna, pomegranate rind, and *Pterocarya fraxinifolia* for nylon 6 dyeing. *Coloration Technol* 2016; 132(2): 162–176.
  41. Penthala R, Heo G, Kim H, et al. Synthesis of azo and anthraquinone dyes and dyeing of nylon-6,6 in supercritical carbon dioxide. *J CO2 Util* 2020; 38: 49–58.
  42. Vuthiganond N, Nakpathom M, Somboon B, et al. Dyeing parameters, fastness and ultraviolet protection properties of nylon dyed with mangrove bark extract. *Mater Today Proc* 2019; 17: 2062–2069.
  43. Bhavsar P, Fontana GD, Tonin C, et al. Superheated water hydrolyses of waste silkworm pupae protein hydrolysate: a novel application for natural dyeing of silk fabric. *Dyes Pigm* 2020; 183: 108678.
  44. Singh G, Mathur P, Singh N, et al. Functionalization of wool fabric using kapok flower and bio-mordant. *Sustain Chem Pharm* 2019; 14: 100184.
  45. Rani N, Jajpura L and Butola BS. Ecological dyeing of protein fabrics with carica papaya L. leaf natural extract in the presence of bio-mordants as an alternative copartner to metal mordants. *J Inst Eng (India) Ser E* 2020; 101(1): 19–31.
  46. Dhanania Y, Singhee D and Samanta AK. Optimization of dyeing process variables for eco-friendly dyeing of cotton fabric with babul bark extract as a natural dye and gallnut extract as a bio-mordant. *J Nat Fibers* 2021; 2021: 1–18.

47. Hossain S, Jalil MA, Kamal SAB, et al. A natural dye extracted from the leaves of *Mimusops Elengi* linn and its dyeing properties on cotton and silk fabrics. *J Text Inst* 2021; 112(3): 455–461.
48. Hassan MM and Saifullah K. Sustainable dyeing and functionalization of jute fabric with a Chinese sumac gall-derived gallotannin using eco-friendly mordanting agents. *Cellulose* 2021; 28: 5055–5070.
49. Ashrafi N, Gharanjig K, Hosseinnzhad M, et al. Dyeing properties and color fabrics using natural dye and mordant. *Prog Color Colorants Coat* 2018; 11(2): 79–83.
50. Shabbir M, Rather LJ, Bukhari MN, et al. First-time application of biomordants in conjunction with the *Alkanna tinctoria* root extract for eco-friendly wool dyeing. *J Nat Fibers* 2019; 16(6): 846–854.
51. Silva PMDS, França VH, Queiroz RS, et al. *Copaifera langsdorffii* Desf. bark extract: optimisation of dyeing conditions to wool and colour fastness properties. *Nat Prod Res* 2021; 2021: 1–6.
52. Adeel S, Rehman F, Pervaiz M, et al. Microwave assisted green isolation of laccic acid from lac insect (*Kerria lacca*) for wool dyeing. *Prog Color Color Coat* 2021; 14(4): 293–299.