Egypt. J. Chem. **Vol. 64,** No. 7 pp. 3751 - 3759 (2021)

Egyptian Journal of Chemistry

http://ejchem.journals.ekb.eg/

Microwave-assisted Dyeing of Wool Fabrics with Natural Dyes as Eco-Friendly Dyeing Method: Part I. Dyeing Performance and Fastness Properties

Morsy A. El-Apasery [1,*], Abdelhaleem M. Hussein [2], Nahed M. Nour El-Din[3], Mohamed O. Saleh[2], and Abu-Bakr A. El-Adasy[2]

[1] *Dyeing, Printing and Auxiliaries Department, Textile Industries Research Division, National Research Centre, Cairo 12622, Egypt*

[2] *Department of Chemistry, Faculty of Science, Al Azhar University, Assiut 71524, Egypt*

[3]*Ecology and Range Management Department., Ecology and Dry Lands Agriculture Division, Desert Research Center, 1 Mathal El-Mataria St, 11753 Matariya, Cairo, Egypt*

Abstract

In this study different Mixtures of natural plants (***Artemisia monosperma*** Del**.** and ***Phlomis floccose*** L.*)* in different proportions and different concentrations of the mixture are used to obtain new natural dyes and applying them to woolen fabric using microwave radiation and then studying the effect of extraction time and the pH of the dyeing bath on the strength of color and then studying the fastness of these dyes against light and Washing and perspiration

1. Introduction

Artemisia monosperma Del**.**

Desert plants have received much attention during recent decades as important natural resources industrialization of natural Dyeing. In the Egyptian desert, the herb ,***Artemisia monosperma*** Delis a perennial fragrant plant which grows widely and wildly in Arabian deserts, and it have many uses such as insects'struggle[1]. ***Artemisia monosperma*** Del.is a perennial medicinal shrub where it has medicinal application, and then it was used extensively in folk medicine[2]. The most important compounds that have a biological effect that can be extracted from ***Artemisia monosperma*** Del**.** are sterols, terpenes, flavonoids, saponins and tannins[3]. ***Artemisia monosperma*** Del**.** has been used in many medical applications, including that it was used as an anti-worm and was also used as an antiseptic. ***Artemisia monosperma*** Del.is a natural antioxidant because it contains polyphenols compounds which stimulate hydrogen atom donation to free radicals, electron transfer and by metal chelating [4]. Where sterols, terpenes, flavonoids, saponins are reported as natural dyes[5] and tannins is mordant dyes[6] then ***Artemisia monosperma*** Del**.** can be used as a natural dyes.

Phlomis floccose L.

Phlomis floccose L. is a medicinal shrub found in the desert of Marsa Matrouh, Egypt [7]. Folk therapy in Egypt used ***Phlomis floccose*** L.as a sedative, stimulant, immune-suppressive, hemorrhoids, free radical scavenger, astringent, hemorrhoid, anti-diarrheal, anti-inflammatory, anti-microbial, anti-malarial, anti-diabetic, anti-mutagenic, anti-cough, anti-nociceptive, for mending of wound and to treat ulcers, gastric, intestinal and abdominal pains [8, 9]. An Egyptian study showed that silver nanoparticles can be obtained from methanol extract of ***Phlomis floccose*** L. and based on that, the study indicated that the ***Phlomis floccose*** L**.** acts as an antioxidant and cytotoxic [10]. Studies reported that ***Phlomis floccose*** L.can be used to struggle insects and

*Corresponding author e-mail: elapaserym@yahoo.com.; (Morsy Elapasery).
Receive Date: 12 April 2021, Accept Date: 21 April 2021
DOI: 10.21608/EJCHEM.2021.72134.3588

agricultural pests because it contains germacrene-D and *β*-caryophyllene with its chemical constituents [11]. The most chemical constituent of ***Phlomis floccose*** L. is flavonoids [12] which make it be able to be used as natural dye.

2. Experimental

- **Natural dyes sources.**

1- ***Artemisiamonosperma***Del.WasPlant materials, in present investigation, were collected from northern and western Sinai.

Scientific classification of ***Artemisia monosperma*** Del.

Kingdom: plantae
Clade: Tracheophytes
Clade: Angiosperms
Clade: Eudicots
Clade:Asterids
Order: Asterales
Family: Asteraceae
Subfamily: Asteroideae
Genus:*Artemisia* Del.
Species: *monosperma*

2- ***Phlomis floccose*** L. was collected from Wadi El-Natron desert – Egypt and dried in air.

Scientific classification of ***Phlomis floccose*** L**.**
Kingdom: plantae
Clade: Tracheophytes
Clade: Eudicots
Clade: Asterids
Order: Lamiales
Family: Lamiaceae
Subfamily: Lamioideae
Genus: *Phlomis* L.
Species: *floccosa*

- **Substrates**

Scoured and bleached wool fabric was purchased from Misr for Spinning and Weaving Company, Mahalla El-Kobra, Egypt.

Microwave assisted extraction

The process of extraction was carried out by using microwave. A Samsung oven model MS404MADXXBB. Three different mixtures of dried *Artemisia monosperma* Del.and *Phlomis floccose* L.(50% *Artemisia monosperma* Del.:50 % *Phlomis floccose* L.) [Dye 1], (75% *Phlomisfloccosa*L: 25% *Artemisia monosperma* Del.) [Dye 2] and (25% *Phlomis floccose* L.: 75% *Artemisia monosperma* Del.) [Dye3] were irradiated in different concentrations (2: 10 gm/L) in aqueous solution.

- **Microwave dyeing method**

The extracted dye was applied to wool fabrics with M: L ratio 1:100. Dyeing procedure was carried out different dye bath pH. A Samsung oven model MS404MADXXBB was utilized and the dyeing time was 1 hour.

- **Color *measurements***

Reflectance Spectrophotometer is the device used to measure the intensity of color on dyed wool fabrics and light reflection technology has been applied for that. The dye yield was determined on the dyed fabric using a spectrophotometer. Kubelka-Munk's Equation (1).Equation was applied to find out the intensity of the color of dye, which is symbolized by the symbol K/S[13].

K/S = [(1- R) 2 / 2R] - [(1- Ro) 2 / 2Ro] (1)

Where R is the reflectance of dyed samples, K is the absorption coefficient, S is the scattering coefficient, and Ro = decimal fraction of the reflectance of the undyed fabric.

- ***Fastness properties***

1- *Color fastness to washing*

The color stability to washing was determined by method of ISO 105-C02:1989 [14]. Aspecimen of the dyed fabric was placed between two bleached slices, one of them is cotton fabric and the other of the wool fabric, and attached with them by hand stitching, and then it was soaked in an aqueous solution consisting of 5 g/L of nonionic detergents at a liquor ratio of 1:50 for 30 minutes at of 60 °C and then sample was rinsed thoroughly with manual squeeze, and then sample was allowed to dry. Gray scale was applied to assess the color fastness to wash.

2- Color fastness to perspiration

Using L-histidine monohydro-chloride monohydrate (0.5 g), sodium chloride (5 g), and sodium dihydrogen orthophosphate dihydrate (2.2 g) in one liter of distilled water, a solution of artificial sweat was obtained. For have acidic sweat pH was adjusted to 5.5 by 0.1 N of NaOH and by the same way an alkaline sweat was obtained by adjusting pH of solution at 8.0.The fastness test was performed according to the following procedure:

Between two different parts of uncolored patterns a sample of dyed fabric (5×4 cm) was sutured. The samples were soaked in both solutions undergoing agitation and pressing for 15-30 min to have perfect wetting. Sample undergo a load of 4-5 kg while the sample was placed between two plates of plastic or glass. Then these plates were placed at a temperature of 37 °C vertically for four hours, and then a gray scale change technique was used to assess the color fastness to perspiration

3- ***Color fastness to light***

The method of measuring the stability of color fastness to light is ISO 105-B02:1988 test technique where the dyed sample placed for 35 hours to a carbon arc lamp and then using the blue color scale to investigate color change of the tested samples.

4- ***Color fastness to rubbing***

By applying the technique of ISO 105-X12:1987 test the color fastness to crocking was determined. When dyed fabric undergoes rubbing, it is can be estimate if dye move from dyed fabric to another one or not. The wet and dry rubbing had been estimates.

3. Results and discussion

Factors affecting dye extraction:

Effect of dye source concentration and extraction time

Different concentrations of the mixed dried powder were prepared; concentrations were (2: 10) gm / litter. Extraction time was studied from 30 minutes to 120 minutes.

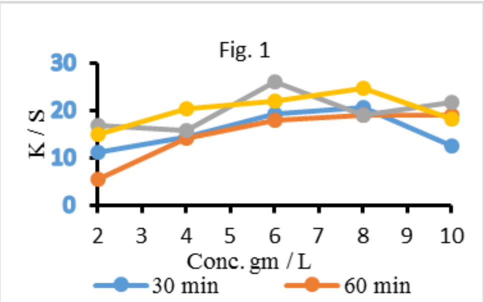

Fig. 1 Effect of dye source concentration and extraction time on K/S of Dye 1.

The time period of the extraction was varied in order to study the maximum possible extraction of dye. The optimum value determined by K/S values.

Table 1 show that optimum time for extraction of Dye 1 is 90 min. and concentration of source of the dye should be 6 gm / L.

Table 2 shows that Dye 2 gives preferable value of K/S at 30 min. Time of extraction with concentration of dye source 10 gm / L.

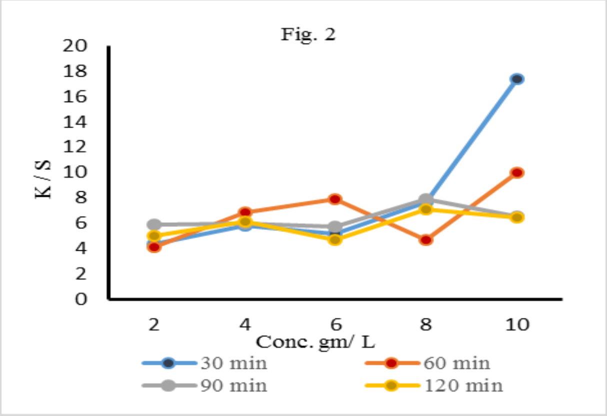

Fig. 2 Effect of dye source concentration and extraction time on K/S of Dye 2.

By the same behavior Dye 3 give best value of K/S at 30 min extraction time and using dye source concentration 10 gm/L which detailed in table 3.

.

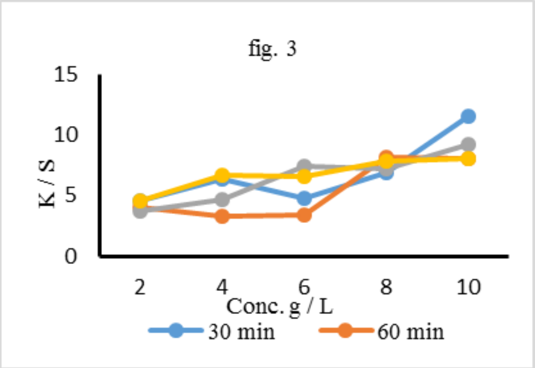

Fig.3. Effect of dye source concentration andextraction time on K/S of Dye 3.

It can be noted that K/S value are decreased after certain point. The decrease in the color intensity values after reaching a certain point may be due to the increasing in the extraction time by microwave, leads to the degradation of the dye resulting from the extraction process [15-18]. From table 1 data dye1 gives the maximum value of the color intensity at a concentration of 6 gm/L, and then the value of the color intensity decreases, may be that behavior due to some dyes reach a certain concentration and then the values of the color intensity decrease while the concentration increased [19]

Table 1. Effect of dye source concentration and extraction time on K/S of Dye 1.

concentration	K / S			
	Refluxing time per minutes			
gm/L	30	60	90	120
2	11.22	5.51	16.73	14.82
4	14.34	14.20	15.72	20.27
6	19.35	17.95	25.95	22.06
8	20.48	18.90	18.90	24.51
10	12.45	19.01	21.65	18.22

Table 2. Effect of dye source concentration and extraction time on K/S of Dye 2

concentration	K / S			
gm/L	Refluxing time per minutes			
	30	60	90	120
2	4.37	4.13	5.87	5.06
4	5.84	6.83	5.97	6.16
6	5.16	7.93	5.72	4.67
8	7.64	4.73	7.91	7.12
10	17.35	10.03	6.54	6.50

Table 3
Effect of dye source concentration and extraction time on K/S of Dye 3.

concentration	K / S			
gm/L	Refluxing time per minutes			
	30	60	90	120
2	4.56	4.00	3.69	4.61
4	6.36	3.33	4.63	6.64
6	4.82	3.44	7.43	6.62
8	6.85	8.12	7.18	7.86
10	11.54	8.00	9.23	8.04

Table 4 Effect of dye bath pH and dyeing time on K/S of Dye 1

Dye bath pH	Dying time/ min	K / S	L*	a*	b*	c	h
pH 3	15	3.95	76.95	0.69	15.59	15.61	87.46
	30	5.21	68.97	3.86	19.86	20.23	79.01
	45	2.77	74.13	2.63	15.07	15.50	80.09
	60	3.65	69.89	4.50	17.24	17.82	75.37
pH 5	15	7.36	69.81	1.85	21.30	21.38	85.03
	30	8.55	69.84	2.67	23.52	23.67	83.50
	45	11.17	65.98	3.78	23.71	24.01	80.93
	60	12.28	64.02	5.04	25.22	25.72	78.70
pH 8	15	3.58	75.41	- 0.69	17.81	17.02	92.21
	30	6.04	72.28	0.22	19.44	19.44	89.35
	45	6.17	70.32	0.83	19.48	19.50	87.65
	60	5.50	69.40	1.15	19.30	19.33	86.60
pH 11	15	2.82	74.91	0.45	20.04	20.05	88.71
	30	3.54	72.18	1.86	23.02	23.10	85.38
	45	4.41	69.77	2.90	24.32	24.49	83.19
	60	4.87	67.88	3.44	34.85	25.09	82.12

Table 5 Effect of dye bath pH and dyeing time on K/S of Dye 2.

Dye bath pH	Dying time / minutes	K / S	L	a	b	c	h
pH 3	15	4.22	70.77	2.80	20.27	20.46	82.14
	30	6.17	70.90	3.05	21.74	21.96	82.03
	45	6.70	68.15	3.15	20.59	20.78	81.29
	60	8.86	65.96	4.59	24.49	24.92	79.37
pH 5	15	2.71	75.53	- 1.14	15.29	15.34	94.28
	30	4.24	72.93	- 0.01	19.82	19.82	90.02
	45	4.56	71.80	0.80	18.89	18.91	87.57
	60	6.70	64.41	1.66	19.84	19.91	85.23
pH 8	15	3.09	69.44	0.85	20.31	20.32	87.59
	30	3.89	72.32	0.23	21.34	21.34	89.39
	45	6.85	64.93	1.01	28.75	28.76	88.00
	60	4.67	66.35	1.76	21.52	66.35	85.33
pH 11	15	2.39	74.62	0.80	20.12	20.13	87.73
	30	2.62	71.86	0.91	19.97	19.99	87.40
	45	3.23	71.63	2.39	23.65	23.77	84.22
	60	3.79	68.48	3.05	23.25	23.45	82.35

Table 6 Effect of dye bath pH and dyeing time on K/S of Dye 3.

Dye bath pH	Dying time / minutes	K / S	L	a	b	c	h
pH 3	15	10.49	68.78	6.20	23.55	24.35	75.26
	30	12.95	70.80	5.09	24.89	25.40	78.44
	45	11.77	64.57	4.32	22.11	22.53	78.94
	60	16.89	65.03	5.58	26.36	26.95	78.06
pH 5	15	5.29	73.53	0.06	17.46	17.46	89.80
	30	6.61	70.67	0.59	19.27	19.27	88.24
	45	5.99	69.67	1.29	20.47	20.51	86.39
	60	5.86	67.23	1.46	19.56	19.62	85.73
pH 8	15	3.14	74.64	- 0.46	19.10	19.10	91.38
	30	6.29	65.65	1.19	19.07	19.10	86.44
	45	3.10	72.46	0.06	19.34	19.34	89.81
	60	4.53	68.15	0.74	21.15	68.15	88.00
pH 11	15	2.10	76.39	- 0.20	17.59	17.59	90.67
	30	1.83	77.67	- 0.50	15.84	15.85	91.82
	45	4.69	70.74	3.58	25.22	25.48	81.92
	60	3.84	71.55	2.88	24.43	24.59	83.28

Table 7:Fastness properties of dyed fabrics

Dye no	**Washing fastness**			**Perspiration fastness Acidic**			**Alkaline**			**Rubbing fastness**		**Light fastness**
	Alt	**SC**	**SW**	**Alt**	**SC**	**SW**	**Alt**	**SC**	**SW**	**Wet**	**Dry**	
1	5	5	5	5	5	5	5	5	5	5	5	5
2	5	5	5	5	5	5	5	5	5	5	5	5
3	5	5	5	5	5	5	5	5	5	5	5	5

Dyeing

Effect of pH Level of Dye Bath and time of dyeing process

One of the most important factors affecting the quality of the dyeing process and the increase in the dye affinity with the fibers is dye bath pH. Where, when using any method of dyeing wool, it is noticeably affected by the pH value of the dyeing bath, which affects the textile of the wool's ability to dye [20, 21].

Dye intensity is higher at acidic pH which leads to the dye-ability decreases at pH values higher than 5 due to the variation of protonated number of terminal amino groups of wool fibers, and therefore the ionic interaction decreases [29.30].

The color of dyeing on wool fabrics is expressed in terms of CIELAB coordinates were measured: (L) lightness; (a) , whose value represents the degree of redness (positive) and greenness (negative); (b), whose value represents the degree of yellowness (positive) and blueness (negative); (c) chroma; hue angle from 0 to 360° (h). The color properties of wool fabric dyed with mixture of ***Phlomis floccosa*** Land ***Artemisia monosperma*** Del. extracts are shown in Table 4, 5 and 6a, b. All the dyed fabrics showed brownish shade in their hue, which was confirmed by their slightly low positive a* but highly positive b*. The positive values of [b] (yellow-blue axis) indicated that the color hues of the dyes D1, D2 and on the wool fabric shifted in the yellowish direction. (L) Highly positive values indicate that color shift to brightness.

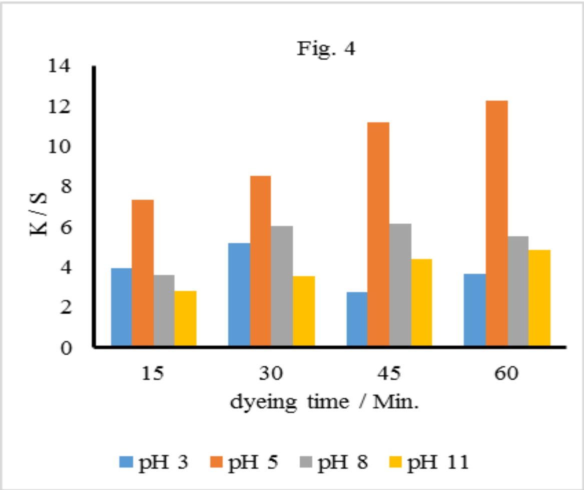

Fig. 4. Effect of dye bath pH and dyeing time on K/S

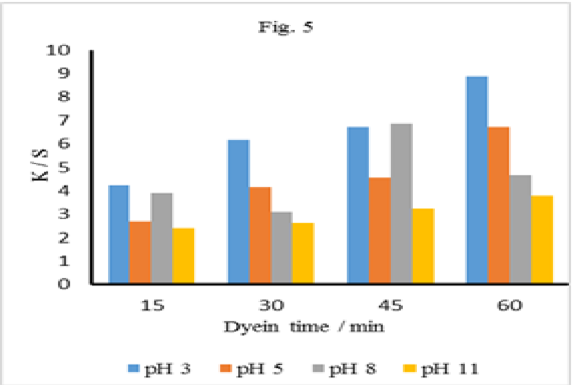

Fig. 5. Effect of dye bath pH and dyeing time on K/S of Dye 2

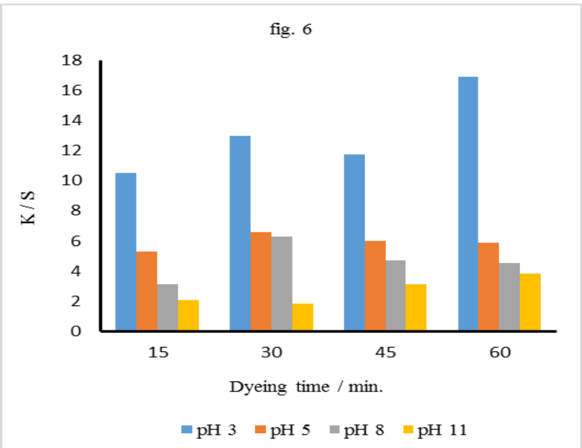

Fig. 6. Effect of dye bath pH and dyeing time on K/S of Dye 3

Fastness properties

The results of washing, light, and rubbing fastness of the dyed wool fabrics were shown in Table 7. The color fastness investigations show that all tested patterns have excellent light fastness ratings of 5 on blue scale. From data of table 7 it is clear that the dyed wool fabrics have excellent wash fastness and perspiration fastness where it is record ratings of 5 on gray scale. And it is clear from results in table 7 that the three natural dyes have excellent rubbing fastness.

Conclusion

The current study shows that, mixing of (***Artemisia monosperma*** Del. and ***Phlomis floccose*** L.*)* that are grown in Egypt can be used as a dye for coloring textiles. The natural dyes of our study have excellent washing, light and rubbing fastness.

References

(1) Hammoda,H. M.; AboulEla, M. A.; El-Lakany, A.M.; El-Hanbali,O.; Zaki1, C. S. ;Ghazy. N. M.New constituents of Artemisia monosperma Del. Pharmazie,**63**, 611–614 (2008).

(2) Sadek, M. A.;Moram,G. S.; Barakat ,H. A.; Nassar,S. S. Anti-tumor, anti-inflammatory and anti-oxidant effects of wormwood (Artemisia monosperma A.) oil extract on the risks of colon cancer induced in rats. World Journal of Pharmaceutical Research, **5**, 305 – 333, (2016).

(3) Al-Watban, A.;Salama, H.M.H. Physiological effects of allelopathic Activity of Artemisia monosperma on common bean (Phaseolus vulgaris L.). International Research Journal of Plant Science, **3**, 158-163, (2012).

(4) Canadanovic-Brunet, J. M.; Djilas, S. M.;Cetkovic, G. S.;Tumbas.V. T.Free-radical scavenging activity of wormwood (Artemisia absinthium L) extracts. Journal of the Science of Food and Agriculture,**85**, 265–272, (2005).

(5) Chairman, K.;Jayamala, M.; Christy,R. V.;Singh,A. R. Phytochemical Screening and Antimicrobial Activity of Curcuma longa Natural Dye.General Med, **3**, (2015).

(6) Guo, L.; Yang, Z.; Tang, R.; Yuan, H. Preliminary Studies on the Application of Grape Seed Extract in the Dyeing and Functional Modification of Cotton Fabric,Biomolecules,**10**, (2020).

(7) Lotfy, R. A; Abd El-Moaty, H. I.Anti-bacterial activities of some active constituents isolated from *Phlomisfloccosa*D. don.Egyptian J. Desert Res., **66**, 69-78, (2016).

(8) Ghareb,H. E.; Ibrahim, S. D.; Hegazi, G. A. Micropropagtion and DNA-barcoding of the endangered endemic *Phlomis*aureaplant of Saint Katherine., POJ,**13**, 65-77, (2020).

(9) Wafa, N.; Sofiane,G. ; Mouhamed, K.The antioxidant and antimicrobial activities of flavonoids and tannins extracted from *Phlomis*bovei De Noé. European Journal of Experimental Biology,**6**, 55-61, (2016).

(10) Abdelhady, N. M.;Badr, K. A.Comparative study of phenolic content, antioxidant potentials and cytotoxic activity of the crude and green synthesized silver nanoparticles′ extracts of two *Phlomis*species growing in Egypt. Journal of Pharmacognosy and Phytochemistry, **5**, 377-383, (2016).

(11) R. El Mokni, S. Majdoub, I. Chaieb, I. Jlassi, R. K. Joshi, S. Hammami., Chromatographic analysis, Biomedical Chromatography.,**33**., 1-10., (2019).

(12) Lotfy, R. A. ; Abd El-Moaty, H. I. Chemical composition and antioxidant activity of the essential oil and flavonoids of *Phlomisfloccosa*D. DON.Egyptian J. Desert Res, **65**, 125-135., (2015).

(13) M.Ashkar, S.;A.El-Apasery, M.; Touma, M. M.;Elnagdi, M. H. Synthesis of Some Novel Biologically Active Disperse Dyes Derived from 4-Methyl-2,6-dioxo-1-propyl-1,2,5,6-tetrahydro-pyridine-3-carbonitrile as Coupling Component and Their Colour Assessment on Polyester Fabrics**.** Molecules,**17**, 8822–8831, (2012).

(14) Al-Etaibi, A. M.; El-Apasery, M. A. Dyeing Performance of Disperse Dyes on Polyester Fabrics Using Eco-Friendly Carrier and Their Antioxidant and Anticancer Activities**.** *Inter. J. Environ. Res. Public Health*, **16**, 4603, (2019).

(15) Zhang, Z.;Lv, G.; Pan, H.; Fan, L.Optimisation of the microwave-assisted extraction process for six phenolic compounds in Agaricusblazeimurrill. International Journal of Food Science and Technolog, **47**, 24–31, (2012).

(16) Vinatoru, M.; Mason, T.J.;Calinescu, I. Ultrasonically assisted extraction (UAE) and microwave assisted extraction (MAE) of functional compounds from plant materials. Trends in Analytical Chemistry, **97**, 159-178., (2017).

(17) Summoogum-Utchanah, S. L.;Joyram, H. An investigation on the potential of extracting natural dyes from Beetroot and Turmeric.IJRET,**04**, 401- 416, (2015).

(18) Xiaokang,W. ;Lyng,J. G.; Brunton,N. P. ; Cody,L.;Jacquier,J.; Harrison,SM..;Papoutsis, K.Monitoring the effect of different microwave extraction parameters on the recovery of polyphenols from shiitake mushrooms: Comparison with hot-water and organic-solvent extractions. Biotechnology Reports, **27**, (2020)**.**

(19) Ansari,N.T.; S. Iqbal, S. Barhanpurkar.,Ecofriendly Dyeing with Senegaliacatechuusing biomordant.IJCRT,**6**, (2018).

(20) Elshemy, S.Unconventional Natural Dyeing Using Microwave Heating with Cochineal as Natural Dyes.RJTA,**15**, 26 – 36,(2011).

(21) Mehrparvar, L.;Safapour, S.; Sadeghi-Kiakhani, M.;Gharanjig, K.A cleaner and eco-benign process for wool dyeing with madder, Rubia tinctorum L., root natural dye. Int. J. Environ. Sci. Technol.Int. J. Environ. Sci. Technol., **13**, 2569–2578, (2016).

(22) Ahmadi, Z.;Houjeghan, F. G.A cleaner and eco-benign process for wool dyeing with madder, Rubia tinctorum L., root natural dye. Int. J. Environ. Sci. Technol,**5**, 97- 102,(2017).

(23) Mehrparvar, L.;Safapour, S.; Sadeghi-Kiakhani, M.;Gharanjig, K.Chitosan-polypropylene imine dendrimer hybrid: a new ecological for cochineal dyeing of wool. Environ Chem Lett.Environ Chem Lett, **14**, 533–539, (2016).

(24) S. Mirnezhad, S. Safapour, M. Sadeghi-Kiakhani., Dual-Mode Adsorption of Cochineal Natural Dye on Wool Fibers: Kinetic, Equilibrium, and Thermodynamic Studies Fibers and Polymers, **18**, 1134-1145., (2017).