Microwave Induced Sustainable Isolation of Laccaic Acid from Lac Insect for Nylon Dyeing

Rehman, Fazal-ur
Department of Applied Chemistry, Government College University, Faisalabad 38000, PAKISTAN

Adeel, Shahid*
Department of Chemistry, Government College University, Faisalabad 38000, PAKISTAN

Pervaiz, Memoona
Department of Applied Chemistry, Government College University, Faisalabad 38000, PAKISTAN

Haji, Aminoddin*[+]
Department of Textile Engineering, Yazd University, Yazd, I.R. IRAN

Haddar, Wafa*
Department of Early Childhood, University College of Turabah, Taif University, Taif, 21944, SAUDI ARABIA

Hussaan, Muhammad
Department of Botany, Government College University, Faisalabad 38000, PAKISTAN

Amin, Nimra
Department of Applied Chemistry, Government College University, Faisalabad 38000, PAKISTAN

Guesmi, Ahlem
Chemistry Department, College of Science, IMSIU (Imam Mohammad Ibn Saud Islamic University), Riyadh, SAUDI ARABIA

ABSTRACT: *Green technologies in isolation and extraction of natural products have always been welcomed due to awareness about environmental standards for global health. The current research was performed to use microwave energy to extract natural dyes from lac insects and their application onto nylon fabric. For isolation of natural dye from lac insect in acidic and acidified methanolic media Mw irradiation for 3, 5, and 7 min. has been given and used to dye nylon fabric. Bio-mordants from herbal-based sources such as Acacia and Turmeric were also employed to develop new shades and to improve fastness properties in comparison with metallic salts of Al (Alum) and Fe (ferrous sulfate). The evidence from this study suggests an increase in color yield (K/S) with the use of 5 min. of microwave energy when acid solubilized extract of lac insect was used to dye onto nylon fabric. It has been demonstrated also that in the case of the pre-mordanting method, 5% of Acacia and 1% of Turmeric give the best fastness properties and the highest color yields. It is concluded that Microwave energy has an excellent efficacy to isolate the colorant, whereas the addition of bio-mordants has made the process more sustainable and greener.*

KEYWORDS: *Acacia, Alum; Iron sulfate; Lac dye; Microwave; Nylon; Sustainability; Turmeric.*

* *To whom correspondence should be addressed.*
+ *E-mail: shahidadeel@gcuf.edu.pk ; ahaji@yazd.ac.ir ; haddar_wafa@hotmail.com*
1021-9986/2021/6/1849-1859 11/$/6.01

INTRODUCTION

In the current scenario, due to serious environmental threats and health risks associated with the use of synthetic colorants, the global community is looking towards natural dyes as an alternative almost in every field of life [1-4]. Synthetic dyes are normally used in a lot of commercial products such as food, cosmetics, textiles, paper, etc. However, after observation by environmental and dermatologist associations, these have been found fatal towards the environment, agriculture land, and global health [5-7]. Studies also demonstrated that many synthetic dyes enter into the human body *via* the perspiration process from fabrics and cause cancer, skin allergy, and DNA damage [8,9]. So, it is obvious that loads of synthetic colorants are toxic, obnoxious and cause serious threats towards the aquatic system and global warming [10,11]. Due to all these negative side effects of synthetic dyes (carcinogenic, mutagenic and genotoxic), the revival of natural dyes in all fields have been welcomed and it is considered as an excellent alternative to replace the use of chemical dyes [12-14]. These dyes have excellent deodorant, antibacterial, antiviral, antioxidant, anti-inflammatory properties due to which mostly these are essential parts of the herbal medication system [15-18].

Indeed, the natural color is obtained from renewable and eco-friendly natural sources such as fauna, flora [19], microbes, and insects [20-22]. Among these sources, insects [23] based natural colorants are also considered due to their biological and therapeutical values for coloration of textiles, cosmetics, foods, flavors, etc. [24,25]. These insect sources include Cochineal [23] Kermes [26] and Lac (Kerria lacca) [4]. Lac dye is one of the oldest naturally occurring sources of anthraquinone dye [27,28]. It is found in the sticky resins containing several anthraquinone derivatives from laccaic acid A, B, C, D, and E, but laccaic acids A and B are found more abundantly [29]. Commercially, being known as Natural Red 25 (CI 75450) and this dye exhibits excellent anti-fungal and anti-bacterial properties [29,30]. Being sparingly soluble in water, its color is orange in acidic dyebath and purple to reddish-violet in alkaline dyebath. It yields orange to purplish red color on matrixes such as textiles, cosmetics, food additives, flavors, etc. [31, 32].

A lot of traditional extraction methods such as soaking, stirring, heating, refluxing, etc. have been used for the extraction of natural products from natural sources. But, due to the high energy-consuming, expensive costs, as well as the consumption of more solvent, the use of these conventional methods, are facing a decline [33]. In the current era, many new technologies have been introduced for effective extraction and potential isolation from natural sources [34] but according to several studies, Microwave-Assisted Extraction (MAE) seems to be more compatible [35]. Indeed, this method of extraction gives better yields by enhancing the vibration and separation of materials in the solvent [36,37]. Moreover, it saves extra time, material, energy, and solvent consumption [38-40]. This treatment is currently used as a commercial tool employed from laboratory scale to industrial level. With non-ionization, clean, and uniform heating nature, it affects directly a particular target (animal cell or plant cell) to interact with solvent *via* ionic conditions and dipole rotation. Thus, through this unique mode of action not only excellent yield is obtained, but also via rapid mass transfer kinetics solvent consumption, energy and time are saved [38, 41]. Physically, it can tune fabrics surface to make it excellently substantive for bio-active molecules without harming its chemical nature [42]. Hence, such advantages have now encouraged researchers to utilize this technique in the extraction and dyeing process with natural colorants.

Because of the advantages of such radiation, and natural dyes of animal origin, the current study has been aimed to make effective isolation of laccaic acid from lac powder to dye nylon and to improve the rating of fastness properties using ecofriendly chemical and bio-mordants.

EXPERIMENTAL SECTION

Chemical products and textile materials

Lac powder was purchased from Nutri Herb, China, and Nylon fabric was provided by Esprit Maille Industry, Tunisia (GSM= 150g/m^2). Commercial-based chemicals such as alum and ferrous sulfate were used as mordants without further purification. Acacia (*Acacia nilotica*) bark and Turmeric rhizomes (*Curcuma longa*) after initial pretreatment were sieved up to 20 mesh and were used for a bio-mordanting purpose.

Isolation process

Isolation of colorant was carried out by boiling a particular amount of powder (1g) with an acidic solution (2% HCl) for 60 min. by keeping the powder to solvent ratio at 1:25. After boiling, the crude material was filtered and used for the irradiation and dyeing process. In another set of experiments, isolation of colorant was carried out by refluxing1 g of powder with acidified methanolic medium for 60 min. by keeping the powder to solvent ratio of 1:25. After refluxing, the crude mixture was filtered and the clear filtrate obtained was used for further experiments.

Irradiation and dyeing Process

To stimulate the colorant, the acidic extract and acidified methanolic extract were exposed to microwave treatment for three different durations (3, 5 and 7 min.) using a domestic microwave device. Nylon fabric was also irradiated with the microwave device for 3, 5, and 7 min. for the modification of the surface to enhance its uptake ability. A comparison study was conducted between irradiated (RE) and un-irradiated extract (NRE) which were used to color irradiated fabric (RF) and un-irradiated fabric (NRF) at 80 °C for 45 min., keeping extract of pH 3 to fabric ratio of 1:25.

Mordanting process

Bio-mordanting, a newly employed process for the development of shade and rating of fastness properties has been carried out. For this purpose, the sieved plant powder of Acacia and Turmeric was boiled for 60 min. by keeping powder to water ratio of 1:25. Electrolytes of Al and Fe were employed for the mordanting of nylon fabrics to get new shades for making comparative studies. For this purpose, concentrations of 1, 3, 5, 7, and 9% of these mordants were employed [42-43].

Analysis of colorfastness characteristics

Assessment of Color yield (K/S) using Kubelka-Munk equation (equation 1) and colorimetric parameters (*L*, a* and b**) of all dyed and mordanted fabric were carried out by Spectraflash SF600 spectrophotometer (Data Color, USA). The rating of fastness properties to light, washing and rubbing was carried out according to ISO standards by with Greyscale at Q.A and Q.C Lab of Noor Fatima Fabrics, Faisalabad, Pakistan).

$$K/S = (1 - R)^2 / 2R \quad (1)$$

RESULTS AND DISCUSSION

Microwave treatment is going to revolutionize the world of natural products for utilization in various fields. Microwave-assisted isolation in natural dyeing has been welcomed by several researchers because it has solved many problems such as isolation yield, reduction of solvent consumption, etc. [42-43]. In the current study, two types of solvent media (acidic and acidified methanol) have been used to isolate the colorant from lac dye. According to Fig.1, the results showed that the best color yield (K/S) was obtained in the case of un-irradiated fabrics (NRF) dyed with irradiated acidified extract (RE) exposed at 5 min.

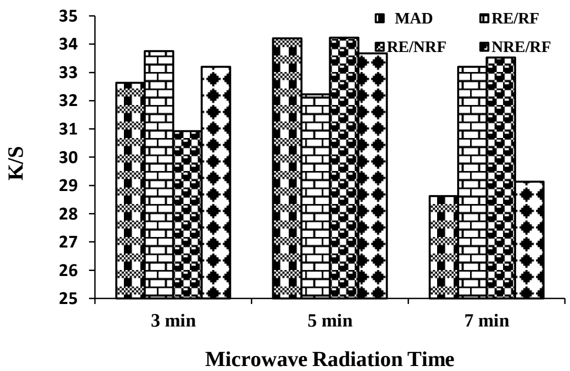


Fig. 1: Effect of microwave radiation duration on the obtained color yield(K/S) of irradiated and un-irradiated Nylon fabrics dyed with an acidified solubilized extract from lac dye. RF= irradiated fabric; NRF= un-irradiated fabric; RE= irradiated extract; NRE= un-irradiated extract; MAD = Microwave-Assisted Dying.

Upon changing the medium from acid solubilized to acidic methanol solubilized medium although microwave treatment for 7 min. has given excellent results into the irradiated fabric (RF) as it is shown in Fig. 2 yet lower than that of acid solubilized extract. This good color yield is due to the excellent mode of action by microwave treatment which has caused encapsulation of laccaic acid from crude Lac powder via solid-liquid interaction and efficient mass transfer kinetics [44]. Also, Nylon favors acidic dye bath because the amide linkage of Nylon is prone to acidic medium for firm electrostatic interaction with lac dye functional sites (-COOH group). Low radiation treatment may not be able to rupture the animal cell wall, whereas high radiation may cause extraction of other molecules along with the colorant that comes into the filtrate and upon dyeing affect the shade [45-47]. Hence, radiation for 5 min., the encapsulation of colorant from the cell wall in acidic medium and effective sorption of colorant (laccaic acid) into fabric occur simultaneously when used for dyeing under Microwave-Assisted Dyeing (MAD) process.

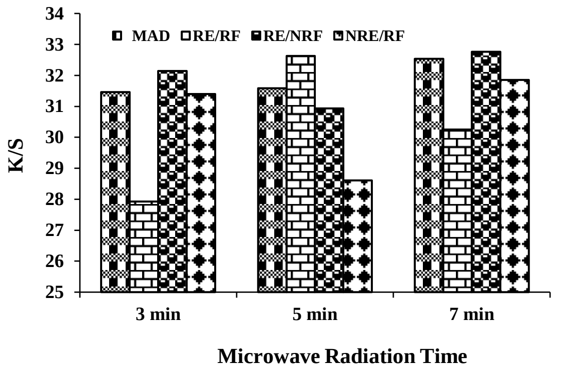
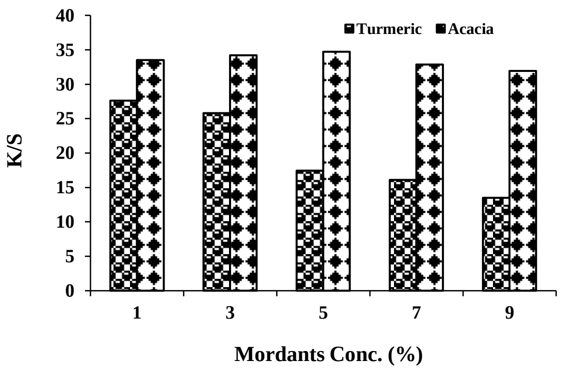


Fig. 2: Effect of microwave radiation duration on the obtained color yield (K/S) of irradiated and un-irradiated Nylon fabrics dyed with acidified methanol solubilized extracts from lac dye. RF= irradiated fabric; NRF= un-irradiated fabric; RE= irradiated extract; NRE= un-irradiated extract; MAD = Microwave-Assisted Aying.



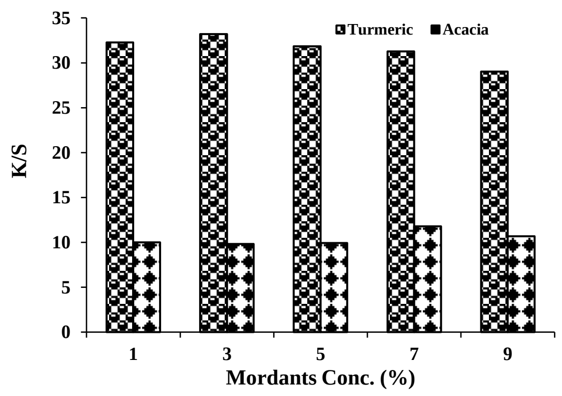


Fig. 3: (a) Effect of concentration of pre-bio-mordants on color yield (K/S) of un-irradiated Nylon fabrics dyed at optimal conditions: Case of the pre-mordanting method. (b) Effect of concentration of post-bio-mordants on color yield (K/S) of un-irradiated Nylon fabrics dyed at optimal conditions: Case of post-mordanting method

After dyeing followed by finishing not only good color yield (K/S) is obtained but also acceptable results are achieved. Thus, it is recommended that acid the solubilized extract should be used for dyeing of nylon fabric under microwave treatment for 5 min. to get acceptable results.

Mordanting is necessary for dyeing natural and synthetic fabric with natural colorants because, without its application, colors are not too fast as desired by customers. Conventionally salts of Alum and Ferrous sulfate were used for developing new tints and for getting the desired fastness. Nowadays, a ray of plant-based mordants has been spread. This is because the functional isolates of plants have excellent anti-oxidant, anti-bacterial and ayurvedic properties which make the process greener, sustainable and aesthetic [34, 48]. The results are given in Fig. 3a for pre-mordanting reveal that 1% of Turmeric extract and 5% of Acacia extract as pre-mordanting have given excellent results. Similarly, 3% of Turmeric extract and 7% of Acacia extract as post-bio-mordants have given excellent results given in Fig. 3b. This is because the Turmeric and Acacia have such functional isolates (ellagitannins) which contain OH as the binding site that interacts with amide linkage of Nylon fabric and (-COOH) and (-OH) groups of colorants via additional attractive forces to give high coloring strength and acceptable fastness rating [49,50]. The purposed bio-mordant Nylon and colorant interaction has been shown in Fig.4. Upon washing, this firmly absorbed complex shows maximum resistance to detaching thereby resulting in a darker shade with acceptable results.

In comparison, when electrolytes of Al (alum) and Fe (ferrous sulfate) were employed, it was found that mordanting with 1% Fe (ferrous sulfate) and 3% of Al (alum) as pre-mordant has produced the highest color strength (Fig. 5a) Whereas 1% of Al (alum) and 3% of Fe (ferrous sulfate) have given the highest color yield (K/S) when applied by post-mordanting method (Fig. 5b). This is due to the formation of the metal-dye complex onto fabric before and after mordanting [51], where also, the nature of metal, colorant, and fabric played their role in obtaining firm fixation of colorant [52]. The purposed mechanism for metal-dye complex formation has been shown in Fig. 6. Upon washing, this firmly absorbed complex shows maximum resistance to detaching thereby resulting in a darker shade with acceptable results.

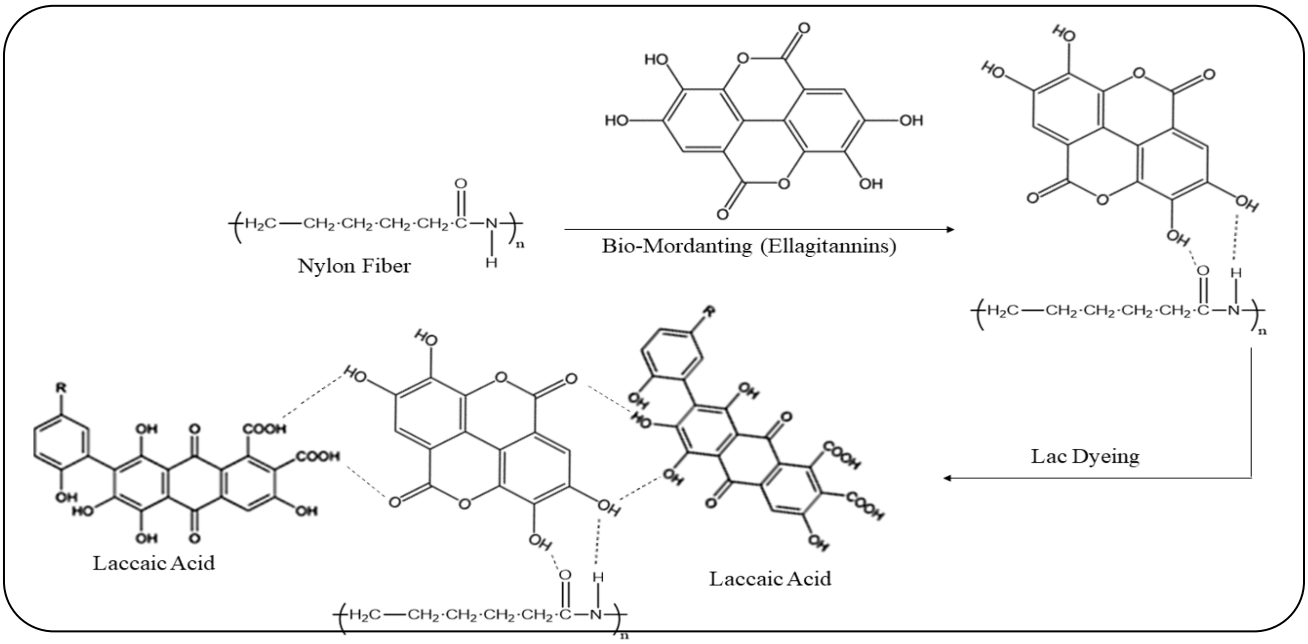


Fig.4: Proposed mechanism for interaction between bio-mordant, nylon, and colorant.

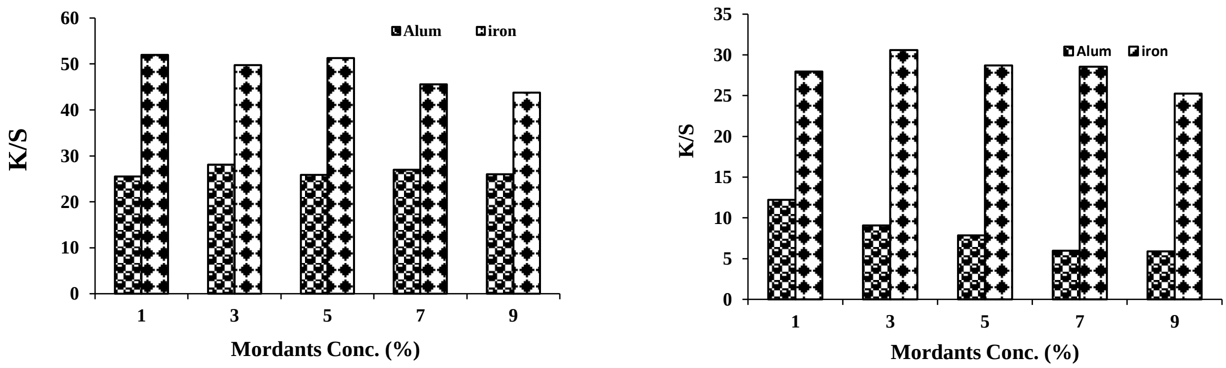


Fig. 5: (a) Effect of concentration of metallic mordants on color yield (K/S) of un-irradiated Nylon fabrics dyed at optimal conditions: Case of the pre-mordanting method. (b) Effect of concentration of metallic mordants on color yield (K/S) of un-irradiated Nylon fabrics dyed at optimal conditions: Case of the post-mordanting method.

The color coordinates given in Table 1a for bio-mordanting reveal that mostly fabric dyed are reddish yellow in tone and darker in shade upon post-mordanting and brighter in shade upon pre-mordanting with Turmeric. The utilization of Acacia has given reverse results because the utilization of Acacia has been given reddish yellow hues but brighter shade upon pre-mordanting and darker shade upon post-mordanting. Hence, overall depending upon the mode of mordanting the variety of hues that can be obtained for lac dyeing of Nylon fabric show that sustainable chemical and bio-mordant should be used for Nylon dyeing using animal-based lac dye. Similarly, the color coordinates given in Table 1b for chemical mordants reveal that mostly fabric dyed before or after mordanting is more reddish yellow in hue but darker in shade after post-mordanting and brighter after pre-mordanting. But, the utilization of Ferrous sulfate has given reverse tonal variation because of the low reduction process; it has given reddish yellow tint but darker upon pre-mordanting and brighter upon the post-mordanting process. Hence, chemical mordanting before or after dyeing of Nylon with Laccaic acid isolated from Lac powder has given a variety of shades.

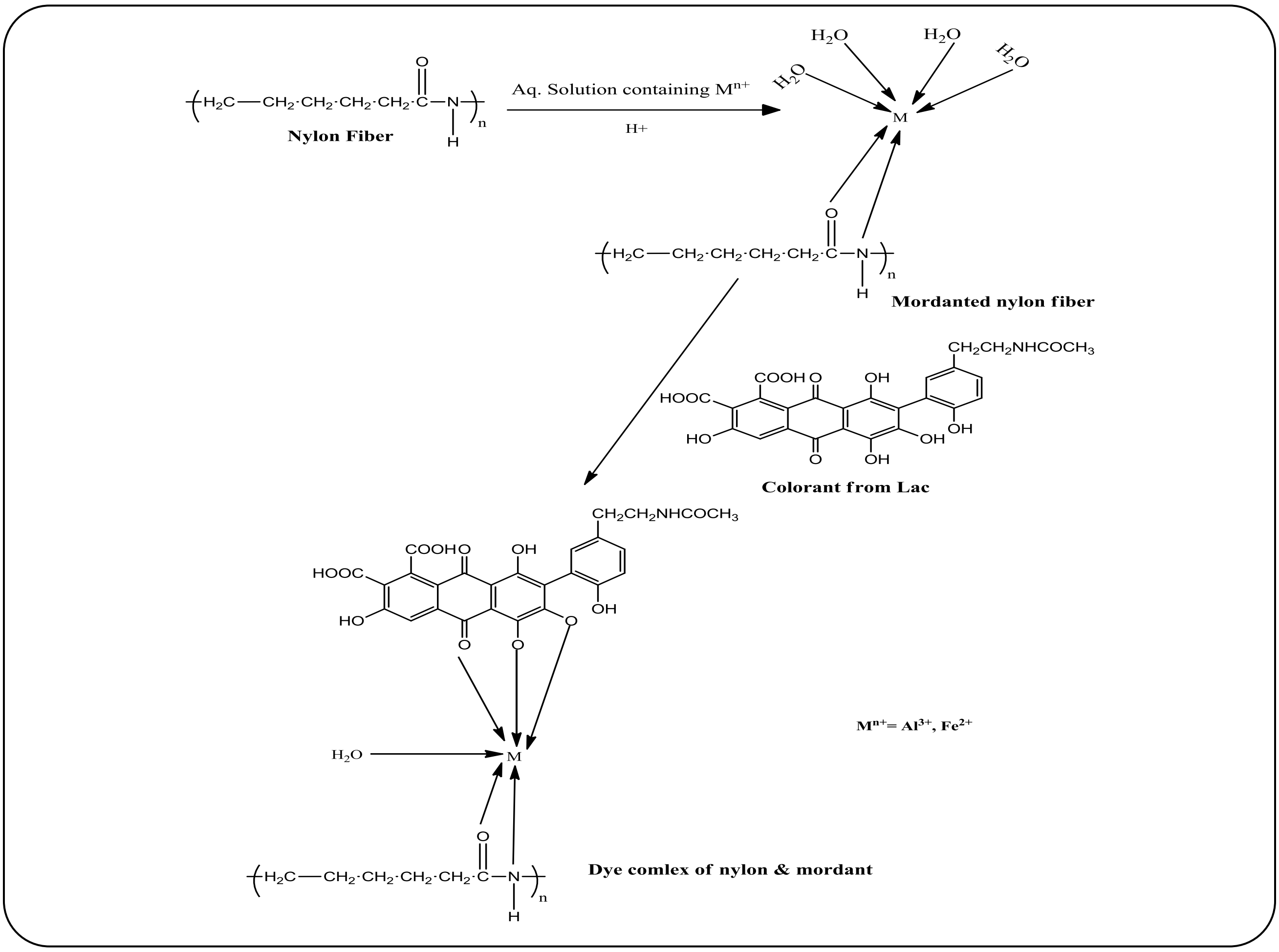


Fig.6: Proposed mechanism for metallic mordant, nylon, and colorant interaction.

Fastness ratings in the natural dyeing process are always very important because the sustainable mordants improve the tint strength via either formation of the metal-dye complex onto fabric or via extra H-bonding with fabric functional sites (Amide linkage) and (-COOH) and (-OH) groups of colorants (laccaic acid) [50-54]. The results given in Table 2a reveal that chemical mordants have improved the fastness rating from good to excellent. Similarly, the utilization of bio-mordants also has enhanced the rating of fastness from good to excellent, as shown in Table 2b. Efficiently, the conjugation system of colorant nature of fabric and isolation yield through proper dyeing methods upon interaction hinders the fastness agents like heat, detergents, crocking, etc. [34]. Thus, microwave treatment used for isolation of colorant from lac powder followed by dyeing of nylon fabric before and after chemical or bio-mordanting has given acceptable color characteristics.

CONCLUSIONS

The current need for sustainable eco-friendly products in textiles is the due need of today's global community. Of these products, animal-derived colorant having ayurvedic and therapeutic nature is being widely encouraged in the applied field. Lac dye (Laccaic acid) colorant for Nylon dyeing has been explored in acid solubilized medium followed by microwave treatment for 5 min. The highest color yield (K/S) was obtained when the extract was irradiated for 5 min. and applied on un-irradiated fabric and bio-mordanted using herbal-based mordants. Acceptable fastness ratings were obtained when a low amount of eco-friendly chemical and bio-mordants were employed before or after dyeing. Thus, the addition of bio-mordants has not only made the process of dying of nylon greener, ayurvedic but also the application of microwave radiation has made process energy and labor effective.

Table 1a: Effect of concentrations of bio-mordants on color yield (K/S) and colorimetric parameters of un-irradiated Nylon fabrics dyed with irradiated acid solubilized extracts at optimal condition.

Types of Mordanting	Mordant conc. (%)	Turmeric			Acacia		
		L*	a*	b*	L*	a*	b*
Pre mordanting	1	29.12	34.83	21.51	25.91	31.89	22.97
	3	31.54	37.28	24.07	28.17	38.48	27.20
	5	36.31	38.28	22.86	23.71	30.29	19.92
	7	36.32	36.91	21.54	28.33	35.26	27.14
	9	33.81	33.67	13.86	24.56	30.45	20.16
Post mordanting	1	14.07	23.39	10.87	32.11	33.45	8.30
	3	19.32	23.97	11.82	33.01	32.08	9.83
	5	20.26	24.58	12.61	34.14	31.30	10.60
	7	20.73	25.47	13.41	31.66	28.88	12.41
	9	22.08	24.60	14.02	32.98	27.79	13.19

*L** = lighter/darker shades; *a** = redder/greener tone; *b** = yellower/bluer tone

Table 1b: Effect of concentrations of metallic mordants on color yield (K/S) and colorimetric parameters of un-irradiated Nylon fabrics dyed with irradiated acid solubilized extracts at optimal condition.

Types of Mordanting	Mordant conc. (%)	Alum			Ferrous sulfate		
		L*	a*	b*	L*	a*	b*
Pre mordanting	1	24.89	32.19	2.9608	23.64	13.59	30.01
	3	29.42	38.93	3.7097	24.22	12.96	21.45
	5	24.58	31.80	4.675	22.37	19.51	13.96
	7	28.49	36.94	5.5524	26.07	16.00	19.37
	9	26.15	34.64	6.1322	28.85	16.02	14.86
Post mordanting	1	32.26	32.75	6.38	18.15	11.36	4.36
	3	36.01	31.25	7.69	15.87	8.99	2.26
	5	37.35	31.08	5.57	17.44	10.09	3.39
	7	40.86	29.87	5.57	18.51	12.88	5.77
	9	40.86	30.35	5.00	17.91	8.01	3.71

*L** = lighter/darker shades; *a** = redder/greener tone; *b** = yellower/bluer tone

Table 2a: Effect of concentrations of metallic mordants on fastness rating of un-irradiated Nylon fabrics dyed with irradiated acid solubilized extracts at optimal condition.

		Alum				Ferrous sulfate			
	Mordant conc.	LF	WF	DRF	WRF	LF	WF	DRF	WRF
Pre Mordanting	1	4/5	4	4	4	4/5	4	4	4
	3	4/5	4	4	4	4/5	3	4	5
	5	5	4	4	4	4/5	5	4	5
	7	4/5	4	4	5	4//5	4	4	4
	9	5	5	4	5	5	4	4	5
Post mordanting	1	5	4	4	4	4/5	4	4	4
	3	5	4	4	4	5	4	4	4/5
	5	5	4	4	4	5	4	4	5
	7	5	4	4	4	4/5	4	4/5	5
	9	4/5	4	4-5	5	4/5	4	4	5

LF = light fastness, WF = wash fastness, DRF = dry rubbing fastness, WRF = wet rubbing fastness

Table 2b: Effect of concentrations of bio-mordants on fastness rating of un-irradiated Nylon fabrics dyed with irradiated acid solubilized extracts at optimal condition.

		Turmeric				Acacia			
	Mordant conc.	LF	WF	DRF	WRF	LF	WF	DRF	WRF
Pre Mordanting	1	5	4	¾	5	5	4	5	5
	3	5	5	4	4	4/5	3	5	4
	5	5	5	5	4	5	4	5	3/4
	7	4/5	4	5	4	4/5	3	5	4
	9	4/5	3	5	4	4/5	4	5	4
Post mordanting	1	4/5	3	4	5	4/5	4	3/4	5
	3	4/5	4	4	5	5	4	4	5
	5	4/5	4	4	5/4	5	4	4	5
	7	4/5	4	5	4/5	4/5	4	4	5
	9	5	4	5	5	5	4	4	5

LF = light fastness, WF = wash fastness, DRF = dry rubbing fastness, WRF = wet rubbing fastness

Acknowledgments

We are thankful to Mr. Zafar Iqbal, Noor Fatima Fabrics (Pvt.) Faisalabad, Pakistan, and to Mr. Muhammad Abbas Harris Dyes and Chemicals Faisalabad Pakistan for providing us the technical assistance during this study.

Received: Feb. 20, 2020; Accepted: June 29, 2020

REFERENCES

[1] Haddar W., Ticha M.B., Guesmi A., Khoffi F., Durand B., A Novel Approach for A Natural Dyeing Process of Cotton Fabric with *Hibiscus Mutabilis* (Gulzuba): Process Development and Optimization Using Statistical Analysis, *J. Clean. Prod.*, **68** :114-120 (2014).

[2] Souissi M., Guesmi A., Moussa A., Valorization of Natural Dye Extracted from Date Palm Pits (*Phoenix Dactylifera*) For Dyeing of Cotton Fabric. Part 1: Optimization of Extraction Process Using Taguchi Design, *J. Clean. Prod.*,**202**:1045-1055(2018).

[3] Wizi J., Wang L., Hou X., Tao Y., Ma B., Yang Y., Ultrasound-Microwave Assisted Extraction of Natural Colorants from Sorghum Husk with Different Solvents, *Ind. Crop. Prod.*, **120**: 203-213 (2018).

[4] Adeel S., Rehman F., Khosa M.K., Anum T., Shahid M., Zia K.M., Zuber M., Microwave Assisted Appraisal of Neem Bark Based Tannin Natural Dye and its Application onto Bio-Mordanted Cotton Fabric, *Iran. J. Chem. Chem. Eng.(IJCCE)*, **39**(2): 157-168 (2020).

[5] Haddar W., Baaka N., Meksi N., Ticha M.B., Guesmi A., Mhenni M.F., Use of Ultrasonic Energy for Enhancing the Dyeing Performances of Polyamide Fibers With Olive Vegetable Water, *Fiber Polym*., **16**: 1506-1511 (2015).

[6] Haddar W., Ticha B.M., Meksi N., Guesmi A., Application of Anthocyanins as Natural Dye Extracted from *Brassica Oleracea* L. Var. Capitata F. Rubra: Dyeing Studies of Wool and Silk Fibres, *Nat. Prod. Res*., **32**:141-148 (2018).

[7] Adeel S., Kiran S., Ahmad T., Habib N., Tariq K., Hussaan M., "Bio- Mordants in Conjunction with Sustainable Radiation Tools for Modification of Dyeing of Natural Fibers", *Frontiers of Textile Materials: Polymers, Nanomaterials, Enzymes, and Advanced Modification Techniques*, 355-367 (2020).

[8] Comlekcioglu N., Aygan A., Kutlu M., Kocabas Y.Z., Antimicrobial Activities of Some Natural Dyes and Dyed Wool Yarn, *Iran. J. Chem. Chem. Eng.* (IJCCE), **36**(4):137-144 (2017).

[9] Choudhury A.K.R., Eco-Friendly Dyes and Dyeing, *Adv. Mater. Technol. Environ. Appl*.,**2**:145-176 (2018).

[10] Ashraf M., Safdar M.E., Shahzad S.M., Aziz A., Piracaha M.A., Suleman M., Ahmad M.B., Challenges and Opportunities for Using Wastewater in Agriculture: A Revie, *J. Appl. Agri. Biotech*., **2(2)**:1-20 (2018).

[11] Echeverria C.A., Handoko W., Pahlevani F., Sahajwalla V., Cascading Use of Textile Waste for the Advancement of Fibre Reinforced Composites for Building Applications, *J. Clean. Prod.*, **208**: 1524-1536 (2019).

[12] Kumbhar S., Hankare P., Sabale S., Kumbhar R., Eco-Friendly Dyeing of Cotton with Brown Natural Dye Extracted from *Ficus Amplissima* Smith Leaves, *Environ. Chem. Lett*., **17(2)**: 1161-1166 (2019).

[13] Hernández VA., Galleguillos F., Thibaut R., Müller A., Fungal Dyes for Textile Applications: Testing of Industrial Conditions for Wool Fabrics Dyeing, *J. Text. I.*, **110**:61-66 (2019).

[14] Kaynar H., Ucar E., Alternative Plants to be Used in Natural Dyeing on Wool Yarn Fibers, *J. Nat. Fibers*, **6**: 379-387 (2019).

[15] Islam SU., Wani S.A., Mohammad F., Imparting Functionality Viz Color, Antioxidant and Anti-Bacterial Properties to Develop Multifunctional Wool with *Tectona Grandis* Leaves Extract Using Reflectance Spectroscopy, *Int. J. Biol.Macromol*., **109**: 907-913 (2018).

[16] Pal A., Kumar R., Upadhyay L., Tripathi Y., Antifungal Activity of Natural Dye from Aerial Biomass of Barleria *prionitis* L. and Dyed Fabrics, *Iran. J. Chem. Chem. Eng*., **37(1)**: 213-221 (2018).

[17] Li L., Thakur K., Liao B.Y., Zhang J.G., Wei Z.J., Antioxidant and Antimicrobial Potential of Polysaccharides Sequentially Extracted from *Polygonatum Cyrtonema* Hua, *Int. J. Biol. Macromol*., **114**: 317-323 (2018).

[18] Zhou Y., Yang Z.Y., Tang R.C., Facile and Green Preparation of Bioactive and UV Protective Silk Materials Using the Extract from Red Radish (*Raphanus Sativus* L.) Through Adsorption Technique, *Arab. J. Chem*.,**13(1)**: 3276-3285 (2018).

[19] Kaynar H., Ucar E., Alternative Plants to be Used in Natural Dyeing on Wool Yarn Fibers, *J. Nat. Fibers*, **6**: 379-387 (2019).

[20] Haji A., Improved Natural Dyeing of Cotton by Plasma Treatment and Chitosan Coating. Optimization by Response Surface Methodology, *Cellul. Chem. Technol*.,**51(9-10)**: 975-982 (2017).

[21] Agnhage T., Perwuelz A., Behary N., Towards Sustainable *Rubia Tinctorum* L. Dyeing of Woven Fabric: How Life Cycle Assessment Can Contribute, *J. Clean. Prod*., **141**: 1221-1230 (2017).

[21] Haji A., Qavamnia S.S., Nasiriboroumand M., The Use of D-Optimal Design in Optimization of Wool Dyeing with *Juglans Regia* Bark, *Ind. Textila*, **69**:104-110 (2018).

[22] Cooksey C.J., The Red Insect Dyes: Carminic, Kermesic and Laccaic Acids and Their Derivatives, *Biotech. Histochem.*, **94**: 100-107 (2019).

[23] Backes E., Pereira C., Barros L., Prieto M.A., Genena A.K., Barreiro M.F., Ferreira I.C., Recovery of Bioactive Anthocyanin Pigments from *Ficus Carica* L. Peel by Heat, Microwave, and Ultrasound-Based Extraction Techniques, *Food Res. Int.*,**113**:197-209 (2018).

[24] Shabbir M., Rather L.J., Mohammad F., Economically Viable UV-Protective and Antioxidant Finishing of Wool Fabric Dyed with *Tagetes Erecta* Flower Extract: Valorization of Marigold, *Ind. Crop. Prod.*,**119**: 277-282 (2018).

[25] El-Boraei N.F., Halim S.A., Ibrahim M.A., Effective Corrosion Inhibition of Mild Steel in Acidic Medium using Inexpensive Kermes Natural Dye: Experimental and Quantum Chemical Study, *Anti-Corros Method M.*, **65**:626-636 (2018).

[26] Naik S.D., Effect of Lac Dyeing on Colour Fastness of Silk Yarn, *Karnataka J. Agri. Sci.,* **27**: 60-62 (2014).

[27] Santos R., Hallett J., Oliveira M.C., Sousa M.M., Sarraguça J., Simmonds M.S.J., Nesbitt M., HPLC-DAD-MS Analysis of Colorant and Resinous Components of Lac-Dye: A Comparison between Kerria and *Paratachardina Genera*, *Dyes Pigm.*, **118**: 129-136 (2015).

[28] Srivastava S., Chowdhury A.R., Maurya S., Antimicrobial Efficacy of Methylated Lac Dye, An Anthraquinone Derivative, *Indian. J. Microbiol.*, **57**: 470-476 (2017).

[29] Mongkholrattanasit R., Ariyakuare K., Limtrakool T., Saiwan C., Rungruangkitkrai N., Punrattanasin N., Nakpathom M., An Evaluation of Silk Fabric Dyed with Lac Dye by Using Pad-Dry Technique. A Research on Effect of Mordant Concentration, *Adv. Mater. Res.*,**118**: 569-572 (2013).

[30] Liu L., Zhang J., Tang R.C., Adsorption and Functional Properties of Natural Lac Dye on Chitosan Fiber, *React. Funct. Polym.*,**73**: 1559-1566 (2013).

[31] Sharma K.K., "Lac Insects and Host Plants". In Industrial Entomology, Springer, Singapore,157-180 (2017).

[32] Kazemi M., Khodaiyan F., Labbafi M., Hosseini S.S., Hojjati M., Pistachio Green Hull Pectin: Optimization of Microwave-Assisted Extraction and Evaluation of Its Physicochemical, Structural and Functional Properties, *Food Chem.,* **271**:663-672 (2019).

[33] Zia K.M., Adeel S., Khosa M.K., Aslam H., Zuber M., Influence of Ultrasonic Radiation on Extraction and Green Dyeing of Mordanted Cotton Using Neem Bark Extract, *J. Ind. Eng. Chem.*,**77(25)**: 317-322 (2019).

[34] Duval J., Pecher V., Poujol M., Lesellier E., Research Advances for the Extraction, Analysis and Uses of Anthraquinones: A Review, *Ind. Crop. Prod.*, **94**: 812-833 (2016).

[35] Park S., Choi H.M., Microwave-Mmediated Rapid Oxidation and Ccationization of Cotton Cellulose, *Cell. Chem. Technol*., **52**:311-322 (2018).

[36] Hosseini S.S., Khodaiyan F., Yarmand M.S., Optimization of Microwave Assisted Extraction of Pectin from Sour Orange Peel and its Physicochemical Properties, *Carbohyd. Polym*., **140**:59-65 (2016).

[37] Vega A., Ramírez-Corona N., Palou E., López-Malo A., Estimation of Mass Transfer Coefficients of the Extraction Process of Essential Oil from Orange Peel Using Microwave-Assisted Extraction, *J. Food Eng.*, **170**:136-143 (2016).

[38] Naccarato A., Tassone A., Moretti S., Elliani R., Sprovieri F., Pirrone N., Tagarelli A., A Green Approach for Organophosphate Ester Determination in Airborne particulate Matter: Microwave-Assisted Extraction Using Hydroalcoholic Mixture Coupled with Solid-Phase Micro Extraction Gas Chromatography-Tandem Mass Spectrometry, *Talanta*, **189**: 657-665 (2018).

[39] Kutlu N., Bicak C., Ekinci D., Kilic E., Erdem N., Isci A., Sakiyan O., Application of Response Surface Methodology to Optimize Microwave-Assisted Extraction of Total Phenolic Compounds from Caucasian Whortleberry, *Gida J. Food.*,**43**:264-272 (2018).

[40] Adeel S., Hussaan M., Rehman F., Habib N., Salman M., Naz S., Amin N., Akhtar N., Microwave-Assisted Sustainable Dyeing of Wool Fabric Using Cochineal-Based Carminic Acid as Natural Colorant, *J. Nat. Fibers*,**16(7)**:1026-1034 (2019).

[41] Kapoore R., Butler T., Pandhal J., Vaidyanathan S., Microwave-Assisted Extraction for Microalgae: From Biofuels to Biorefinery, *Biology*,**7**:1-125 (2018).

[42] Amin N., Rehamn F., Adeel S., Ahmad T., Muneer M., Haji A., Sustainable Application of Cochineal Based Anthraquinone Dye for the Coloration of Bio-Mordanted Silk Fabric, *Environ. Sci. Pollut. Res*., **27**: 6851-6860 (2020).

[43] Adeel S., Zuber M., Rehman F., Zia K.M., Microwave-Assisted Extraction and Dyeing of Chemical and Bio-Mordanted Cotton Fabric Using Harmal Seeds as a Source of Natural Dye, *Environ. Sci.Pollut. Res*., **25(11)**:11100-11110 (2018).

[44] Gala S., Sumarno S., Mahfud M., Microwave-Assisted Extraction of Natural Dyes from *Coleus Atropurpureus* Leaves: The Effect of Solvent, *In MATEC Web of Conferences*, **156**:06011 (2018).

[45] Rodríguez-González I., Ortega-Toro R., Díaz C., Influence of Microwave-and Ultrasound-Assisted Extraction on Bioactive Compounds from Pollen, *Contem. Eng. Sci*.,**11**:1669-1676(2018).

[46] Thangabai T., Kalaiarasi K., Microwave-assisted Natural Dye Extraction from *Pterocarpus soyauxii*, *Int. J. Eng. Technol. Sci. Res*., **5**: 2394-3386(2018).

[47] 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 Polym*.,**19**: 2359-2364 (2018).

[49] Ismal O.E, Greener Natural Dyeing Pathway Using a By-Product of Olive Oil; Prina and Biomordants, *Fibers Polym*., **18(4)**: 773-785 (2017).

[50] Yusuf M., Mohammad F., Shabbir M., Khan M.A., Eco-Dyeing of Wool with *Rubia Cordifolia* Root Extract: Assessment of the Effect of *Acacia Catechu* as Bio-mordant on Color and Fastness Properties, *Text. Cloth. Sustain*., **2**:1-9 (2017).

[51] Adeel S., Naseer K., Javed S., Mahmmod S., Tang R.C., Amin N., Naz S., Microwave-Assisted Improvement in Dyeing Behavior of Chemical and Bio-Mordanted Silk Fabric Using Safflower (*Carthamus Tinctorius* L) Extract, *J. Nat. Fibers*, **17(1)**: 55-65 (2020).

[52] Shabbir M., Islam S.U., Bukhari M.N., Rather L.J., Khan M.A., Mohammad F., Application of *Terminalia Chebula* Natural Dye on Wool Fiber-Evaluation of Color and Fastness Properties, *Text. Cloth. Sustain*., **2**: 1-9 (2017).

[53] Baaka N., Haddar W., Ticha M.B., Amorim M.T.P., Henni M.M.F., Sustainability Issues of Ultrasonic Wool Dyeing with Grape Pomace Colourant, *Nat. Prod. Res*., **31**: 1655-1662 (2017).

[54] Hayat T., Adeel S., Rehman F., Batool F., Amin N., Ahamd T., Ozomay M., ., Waste Black Tea Leaves (*Camelia sinensis*) as Sustainable Source of Tannin Natural Colorant for Bio-Treated Silk Dyeing, *Env. Sci Pollut Res.*, 29(16):24035-24048 (2022).

[55] Batool F., Adeel S., Azeem M., Iqbal N., Natural Dye Yielding Potential and Compounds of Selected Vegetable Residues Belonging to Brassicaceae: An Approach towards Sustainability, *Pak. J. Bot.,* 54(1): 329-336 (2022).