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Environmentally Dyeing of Wool Yarns Using of Combination of Myrobalan and Walnut Husk as Bio-mordants

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ABSTRACT

Today, it is important to pay attention to the reduction of toxic effluents and pollution caused by industrial effluents. The textile industry and dyeing procedure is one of the most important industries in the world, and its effluent causes environmental pollution. Reducing the use of chemical compounds in the dyeing process can reduce these adverse effects. In this article, the aim is to investigate the use of the natural colorant of Madder in the presence of a mixture of two natural tannins-rich mordant (Myrobalan and Walnut husk). For the application of mordant, the pre-mordanting method is used and their performance is compared with Alum. A mixture of two natural mordant with different percentages, the total of which will be 10 %, was used. The changes in the application of the mordant and the natural dye of the yarns are evaluated using the FTIR test. The removal of the peak related to C-N bonding indicates the proper interaction between the mordant, the dye and the yarns. The results illustrated that the use of mordant and increasing the dye concentration increases the color strength. The results showed that the best percentage of mordant mixing is WH:YM=7.5 %:2.5 %. ISO standards were used to check the fastness properties of dyed yarns and moderate to good results were obtained. Prog. Color Colorants Coat. 16 (2023), 197-205© Institute for Color Science and Technology.

1. Introduction

Natural extracted from flowers, leaves, roots, seeds, and bark were long used for dyeing of fibers. These natural dyes can be a good alternative to chemical and synthetic compounds. The use of natural dye in the dyeing process can reduce pollution caused by the use of chemical dyes. Many chemical dyes have toxic or allergic effects [1-3]. The limitation of the use of natural dyes is the low fastness and low color strength of these compounds in dyeing procedure. This defect is solved by using chemical compounds, some of which are: Al^{3+}, Cu^{2+}, Cr^{3+}, Co^{2+}, Ni^{2+}, Fe^{2+}, etc. are used. Among them Cr^{3+}, Cu^{2+}, Ni^{2+}, Co^{2+}, etc. These compounds are environmental hazards that can be replaced with all kinds of extracts. Plants with tannins have good performance instead of these metal salts [4, 5].

Shahnoradi et al. selected metal and new bio-mordant in cotton fabric dyeing using new natural dye from Hibiscus sabdariffa L. and investigated the antibacterial, fastness and color strength of dyed

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samples. All three conventional mordanting methods were used in this research and all kinds of metal mordant including iron, copper, zinc and aluminum salts were used. Regarding natural mordant, lemon peel and pine fruit were selected. The results showed that the used natural mordant have fastness and antibacterial properties that can compete with mineral mordant [6]. For the first time, Zhang et al. used vine tea extract from the class of flavonoid dyes for cotton dyeing and improved the dyeing properties by using natural mordant. The results show that the use of tannin-rich natural mordant can improve the color properties and fastness of dyed fibers and reduce the pollution of using metal mordant [7]. Dyeing leather as clothing or shoe uppers is one of the uses of dyes, and the use of natural dyes reduces the pollution of this process. Mahdi et al [8]. used Acacia nilotica bark for this purpose. Dyeing was done with two mordanting methods pre- and post-mordanting, and to compare the results, dyeing was also done without using mordanting. The fastness properties of the dyed samples are from medium to excellent. The effect of yellow and black Myrobalan as a natural mordant in the presence of two natural dyes, Madder and Weld, was investigated in the dyeing of wool yarns. The results showed that the use of mordant improves the chemical bond between natural yarns and dyes. The fastness properties of dyed yarns in the presence of natural mordant are similar to those of yarns dyed in the presence of metal mordant. Therefore, the use of Myrobalan can replace the metal mordant of alum salt [9]. Haque et al. studied the dyeing of wool fabrics using Black rice extract. To optimize the dyeing conditions, a new method was used, and to improve the fastness properties, the mordant was used. The results showed that black rice extract creates antimicrobial and UV resistant properties on fibers [10].

This research is the use of natural mordant and dye in yarns dyeing and reducing the pollution of the dyeing process. For this purpose, natural tannin-rich mordant, Walnut husk and Myrobalan are used and they are applied on the yarns using the pre-mordanting method. Madder is used as a natural dye. Instrumental tests such as FTIR are used to evaluate the performance of mordant and dyes on yarns. Fastness properties will be checked using standard (ISO) methods. To compare the results of using natural mordant, alum sal will be used as metal mordant.

2. Experimental

2.1. Materials and instrumentation

In this research, wool yarn with 215 tex/2 fold characteristic was used as a natural fiber used in Iranian carpets. Madder having common name of Rubia argyi belongs to Rubiacease family are the main sources of natural dyes obtained from vegetable for wool dyeing. Natural dye (Madder) and Yellow Myrobalan and Walnut Husk as natural mordant were obtained in the north and in the center of Iran. The common name Yellow Myrobalan and Walnut husk also appears in the names of species in related genera, notably *Terminalia chebula* and *Juglandaceae*, respectively. The madder and natural bio-mordants innovatively extracted and fully characterized in our pervious paper [11]. Ethanol was obtained in laboratory grade from Merck Co. and was used without purification.

The equipment used in this research are: FTIR-ATR spectra as Perkin Elmer, USA) equipped with a ZnSe crystal, Color Eye 7000 A spectrophotometer from Gretag Macbeth and Xeno test 150s Hanau for light fastness as ISO 105-B02:2014(en). Wash, rub and perspiration fastness properties of the dyed yarns were determined according ISO 105-C10 2006(en), ISO 105-X12 2016 and ISO-105-E04:1994(E) standards.

2.2. Mordanting processes

Wool was washed in the process for 20 min using a soap solution. two mordants, i.e., WH:YM= 5, 2.5, 4, 6 and 7.5 % (where WH = Walnut husk and YM = Yellow Myrobalan), and the concentrations of dye of 5 - 20 percentage were utilized for dyeing. The wool yarns were mordanted by pre-mordanting method in water, M:L ratio 1:40 (Figure 1).

2.3. Dyeing of wool yarns

The dyeing process was done in L:R=40:1 and 5, 10, 20 and 40 % (o.w.f %) of natural dye conditions. Dyeing started at 25 °C and the temperature reached the boiling point (100 °C) for 30 minutes. The contents of the dyeing bath were gently stirred at boiling temperature for 60 minutes. Next, the temperature of the bath was slowly cooled and the dyed samples, removed and washed (Figure 1).

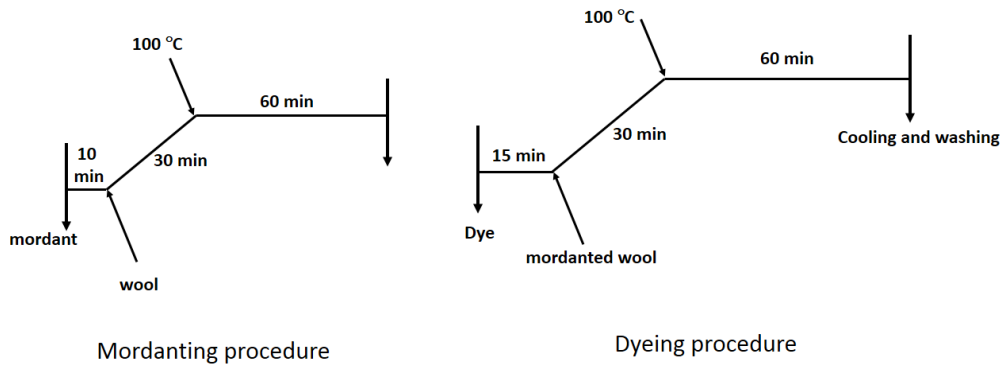


Figure 1: Mordanting and dyeing curve.

3. Results and Discussion

One of the most widely used medicinal plants is Myrobalan with the scientific name Terminalia chebula. The use of this fruit in the traditional medicine of India and Iran has a long history. This fruit has been used for the treatment of various diseases since the past and has had different names in different regions. In other words, the history of the medical use of Myrobalan is completely clear and documented. The medical use of Myrobalan is based on the temperament of people, for which no exact laboratory criteria have been defined [12, 13]. The second natural source used in this research is Walnut husk, which is widely used to remove polluted waste water, and a lot of research has been done on this issue. Walnut with the scientific name Juglans regia L is native to Iran, Afghanistan and western China. Walnut is the second largest product of the nut class in 2019, with an estimated production of 3.7 million tons [14]. 70 % of all plant and wood products are related to Lingocellulose. The main components of Walnut husk include lignin, cellulose, hemicellulose and tannins. The tannin in Walnut husk can be extracted and used in various applications. It is difficult to accurately estimate the constituents of walnut husk, as in many other natural sources. But their approximate percentages are: lignin: 50.3 %, hemicellulose: 22.4 %, cellulose: 23.9 and ash: 3.4 % [15, 16]. In this study, natural dye and mordant resource was acheived from underbrush grown and tree in the north and center of Iran.

The FTIR spectra data of washed wool fiber and mordanted dyed wool fibers are taken and represented in Table 1 and Figure 2. Infrared spectra of the wool fiber indicate absorption peaks assigned to peptide bonding, as reference structure one may expect from wool [17, 18]. 16 to 18 % of wool fibers are water, which is a significant weight ratio. On the other hand, wool is considered a hydrophilic fiber that has the ability to absorb a lot of water. Purification of wool yarns is very complicated and causes impurity in the product [19- 21]. The FTIR spectra of dyed wool fibers show that all C-N peaks are disappeared due to an interaction between fiber, mordant and dye molecules.

Table 1: FTIR data of wool, mordanted yarns.

sample	FTIR (cm^{-1})
Wool yarn	O-H str. 3378; C=O bond str. 1689; C=C str. 1602, 1447
Mordanted-yarn (natural mordant)	O-H str. 3524; C=O bond str. 1700; C=C str. 1609, 1453
Mordanted-yarn (Alum)	O-H str. 3559; C=O bond str. 1709; C=C str. 1617, 1464
Walnut husk	O-H str. 3374; C=O str. 1711; C=C str. 1606, 1455

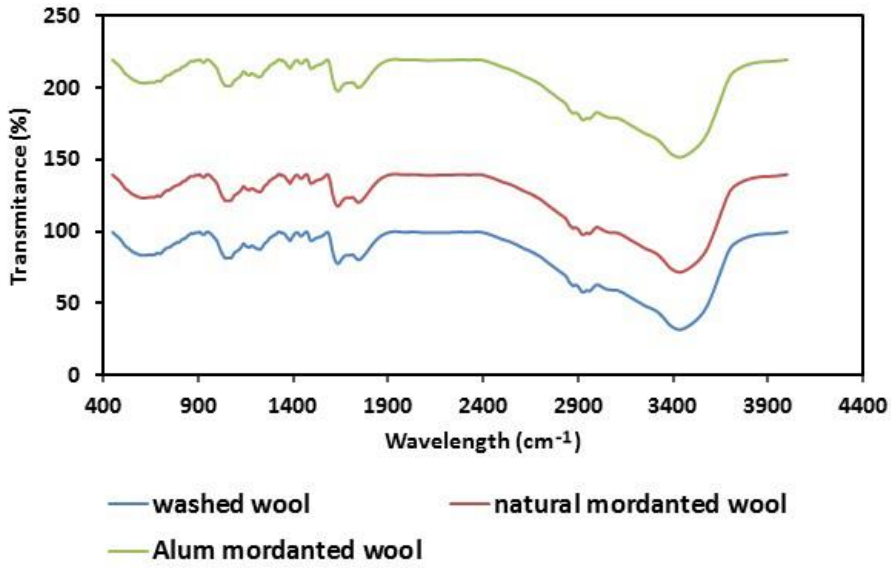


Figure 2: FTIR spectra of wool and mordanted-wool.

The use of mordant in the dyeing process strengthens the chemical bond between the fiber and the dye and has a direct effect on the amount of color strength and fastness. Of course, currently more than metal salts are used as mordant, some of which, such as chromium, are environmental hazards [18, 20]. In this research, an attempt has been made to propose a way to replace metal mordant, and the use of natural tannin-rich extracts are one of these ways [20]. For this purpose, Myrobalan and Walnut husk extract have been used, which have a high percentage of tannin. In other words, the use of a mordant significantly increase the K/S value as shown in Figure 3. This result is due to the simultaneous effect of two mordant and increasing access to tannin. The mordant bind the dyes to the fibers through chemical bonds, and this is due to their affinity for the fibers and dyes. Therefore, natural dyes that are not capable of dyeing the fibers are attached to the fibers through the process of mordanting. The type of chemical bond is different in tannin-rich mordant and mineral-rich mordant. In tannin-rich mordant, chemical bonds are created due hydroxy groups in tannins as hydrogen bonding type. Binding to the colorant component is done through other hydroxy groups of tannin [21].

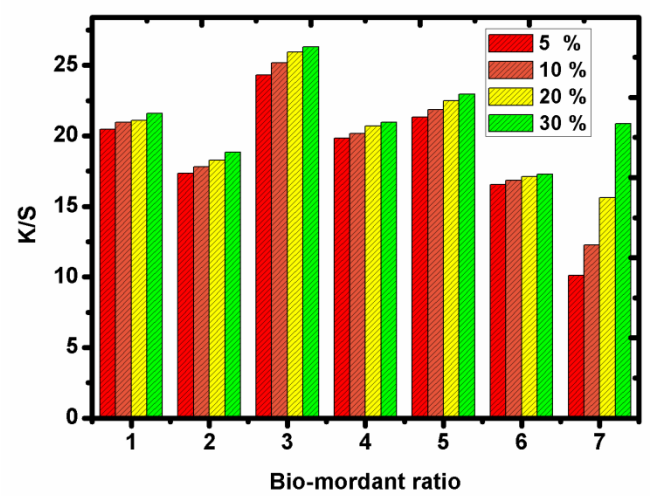


Figure 3: K/S value of dyed yarns with madder.

Table 2: Colorimetric results of samples.

Mordant		WH:YM=5:5 %	WH:YM=2.5:7.5%	WH:YM=7.5:2.5%	WH:YM=4:6%	WH:YM=6:4 %	Alum
Dye 5 %	L*	58.23	57.40	58.12	58.72	58.84	59.21
	a*	23.17	23.45	24.15	23.69	24.19	24.24
	b*	20.10	20.05	20.17	20.09	20.11	21.09
	C*	31.24	31.48	31.17	31.28	31.47	32.07
Dye 10 %	L*	54.68	54.12	55.09	54.39	54.28	55.67
	a*	27.14	26.84	27.89	27.09	27.54	28.11
	b*	20.19	20.16	20.23	20.21	20.24	20.60
	C*	35.14	35.21	35.38	35.68	35.14	35.68
Dye 20 %	L*	51.67	50.91	51.23	50.76	51.49	51.69
	a*	30.41	30.14	30.82	30.35	30.65	31.31
	b*	20.22	20.27	20.31	20.30	20.41	21.17
	C*	38.20	38.28	38.51	38.47	38.52	39.98
Dye 40 %	L*	46.52	46.17	46.57	46.23	45.86	47.28
	a*	30.49	30.23	30.98	30.47	30.74	31.50
	b*	20.48	20.51	20.64	20.66	20.74	21.21
	C*	39.14	39.19	39.31	39.94	39.27	40.19

The hues created on the fibers depend on the type of mordant. By the way, in the same conditions, by changing the mordant, different hues can be created on the fibers [22, 23]. Determining the optimal concentration of natural mordant is necessary to preserve the environment and achieve appropriate dyeing characteristics [24, 25]. In a research, it was found that in green dyeing using natural cochineal dye, the suitable pH for dyeing is around one [26]. The effect of the combination of black and yellow Myrobalan fruits and their combination as a mordant in the dyeing of wool yarns was investigated. The results showed that 4:6 = YM:BM percent is suitable for this purpose. It seems that the tannin percentage of both fruits is similar and their combination does not have a significant effect on the dyeing results [5].

Table 2 illustrated the colorimetric results of dyed wool fibers. Dyed yarns have water content in the first quarter of the color zone. Therefore, the fibers dyed with natural and mineral mordant (alum) are red in shade and only have different color depth. As expected, the increase in the concentration of the dye in the dyeing process has decreased the amount of brightness (L^*), which is due to the increase in the percentage of dye available on the yarns. Visually, darker yarns are seen. The calorimetric results of dyed yarns in the presence of mineral and natural mordant have an acceptable agreement, which indicates the possibility of replacing mineral mordant with a mixture of Myrobalan and Walnut husk extract.

The fastness properties of dyed yarns were evaluated using ISO standards and the results were reported in Table 3. As it is known, the use of mordant improves the fastness properties of dyed yarns. The washing fastness of the dyed yarns in the presence of natural mordants was about 4-5 and their light fastness was acceptable. The results are in good agreement with previous reports. In another study by the authors, the combination of two types of black and yellow Myrobalan fruit was evaluated. Although the yellow and black Myrobalan are tannin-rich component, but because they have similar chemical properties, their combination did not have a significant effect on the results, and their individual use provided similar results. The best result was obtained in the combination ratio of YM:BM=4:6 % [27]. The use of biological mordant has also been investigated and empolyed for dyeing wool. The results show that the combination of biological mordants can be effective in producing new shade with good fastness properties, and they are very

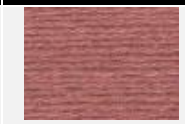
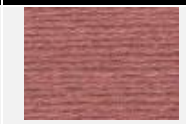
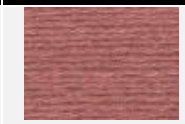
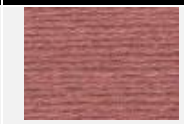
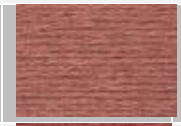
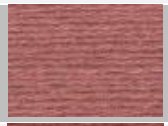
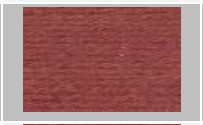
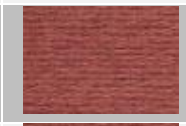
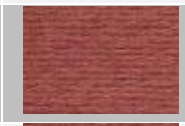
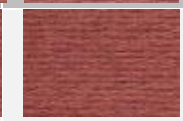
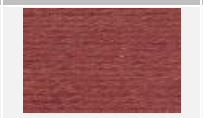
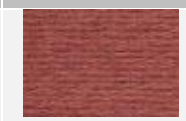
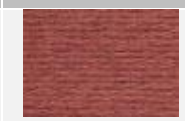
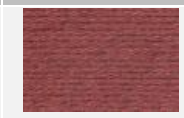
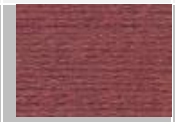
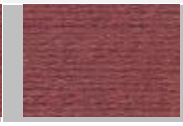
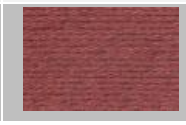
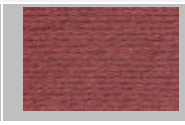
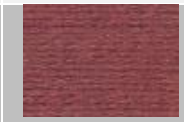
suitable for replacing mineral mordant such as iron salts [28, 29]. One of the new natural dye used by Ismail and et al. for wool dyeing is green almond skin. To check the dyeing properties, the conventional and ultrasound method were chosen in the presence of natural mordant, including pomegranate, rosemary and cedar. The results showed that the fastness properties of all the dyed fibers are good, but the shade produced by the ultrasound method is deeper than the traditional method [30]. Singh and Sheikh used the natural dye obtained from Kigelia Africana for dyeing of wool fibers and evaluated the production effluent. The results showed that there are no toxic substances in the effluent and the dyed fibers have a color perception higher than 18. On the other hand, all fastness properties of dyed was reported as medium to good [31]. The real photo of dyed yarns is shown in Table 4.

Table 3: Fastness properties of samples.

mordant			WH: YM= 5:5 %	WH: YM=2.5:7.5 %	WH: YM=7.5:2.5 %	WH: YM=4:6 %	WH: YM=6:4 %	Alum
Dye 5 %	WF[1]	Ch[4]	4	4	4	4	4	4-5
		St[5]	4	4	4	4	4	4-5
	LF[2]		4	4	4	4	4	4-5
	RF[3]	Ch[4]	4	4	4	4	4	4-5
		St[5]	4	4	4-5	4	4	4-5
	CF [6]	Ch[4]	4	4	4	4	4	4-5
		St[5]	4	4	4-5	4	4	4-5
Dye 10 %	WF[1]	Ch[4]	4	4	4	4	4	4-5
		St[5]	4-5	4-5	4-5	4-5	4-5	4-5
	LF[2]		4	4	4	4	4	4-5
	RF[3]	Ch[4]	4	4	4	4	4	4-5
		St[5]	4-5	4-5	4-5	4-5	4-5	4-5
	CF [6]	Ch[4]	4	4	4	4	4	4-5
		St[5]	4-5	4-5	4-5	4-5	4-5	4-5
Dye 20 %	WF[1]	Ch[4]	4-5	4-5	4-5	4-5	4-5	4-5
		St[5]	4-5	4-5	4-5	4-5	4-5	4-5
	LF[2]		4	4	4	4	4	4-5
	RF[3]	Ch[4]	4	4	4	4	4	4-5
		St[5]	4-5	4-5	4-5	4-5	4-5	4-5
	CF [6]	Ch[4]	4	4	4	4	4	4-5
		St[5]	4-5	4-5	4-5	4-5	4-5	4-5
Dye 40 %	WF[1]	Ch[4]	4-5	4-5	4-5	4-5	4-5	4-5
		St[5]	4-5	4-5	4-5	4-5	4-5	4-5
	LF[2]		4	4	4	4	4	4-5
	RF[3]	Ch[4]	4-5	4-5	4-5	4-5	4-5	4-5
		St[5]	4-5	4-5	4-5	4-5	4-5	4-5
	CF [6]	Ch[4]	4-5	4-5	4-5	4-5	4-5	4-5
		St[5]	4-5	4-5	4-5	4-5	4-5	4-5

1; WF; wash fastness, 2; LF: light fastness, 3; RF: rubbing fastness, 4; Ch: Change, 5; St: Staining.

Table 4: The real photo of dyed yarns.

mordant	WH:YM=5:5 %	WH:YM=2.5: 7.5 %	WH:YM=7.5: 2.5 %	WH:YM=4:6 %	WH:YM=6:4 %	Alum
Dye 5 %						
Dye 10 %						
Dye 20 %						
Dye 40 %						

Tannins are the most important types of natural mordant that can be obtained from various sources [30-36]. In this research, two natural tannin-rich sources, Myrobalan and Walnut husk, have been used. The improvement of the fastness properties and color strength in the presence of the mordant indicates the importance of using the mordant in the dyeing process and the effectiveness of the mixture of Myrobalan and Walnut husk. Yin et al. evaluated the effect of the presence of mordant in the dyeing of silk fibers with a dye obtained from wild mulberry. The results showed that the color strength is at least doubled in the presence of mordant and the fastness properties will increase significantly [37]. Black carrot extract was used for silk dyeing by Shukla and Vankar. In this study, the pre-mordanting method was used for dyeing and the results showed that the presence of the mordant improves the color strength. The best result was obtained for iron salt mordant [38].

4. Conclusion

One of the industries polluting the environment is the textile industry, which introduces a large part of the chemical dyes used in the dyeing process into the environment. The use of natural dyes can reduce the volume of this pollution, but natural dyes often have low color strength. Mordants are used to solve this limitation. In this research, the Madder as natural color was used for wool dyeing, and the effect of the presence of the mixture of Myrobalan and Walnut husk as bio-mordant was evaluated. The percentage of mordant used was 10 % and the pre-mordanting method was used. The effect of using mordant on the fibers was evaluated by the FTIR method, and the removal of cyano peak indicates the effectiveness of the mordanting process.

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