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Comparative Study Between Mixed Dyeing of Wool Fiber in One and Two Dye Baths with Natural Indigo and Madder Colorants

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Abstract: The objective of the present study was to compare between the mixed dyeing of wool fiber in one and two dye baths using natural indigo (*Indigofera tinctoria*) and madder colorants (*Rubia tinctorum*). Based on the reduction and oxidation reaction conditions under the same dyeing mechanism for the indigo and madder colorants, the exhaustion rate of the mixed dyeing in two dye baths was higher than that for mixed dyeing in one dye bath. The poor exhaustion rate that was achieved when dyeing in a single dye bath indicated the incompatibility of both dyes when mixed in one dye bath. However, both methods achieved a rigid fixation as confirmed by washing-fastness test 105C6A01.

Keywords: Wool, Indigo, Madder Colorants, Mixed Dye Bath, *Rubia tinctorum, Indigoferra tinctoria,* Washing Fastness

1. Introduction

Natural dyes produce an extraordinary diversity of rich and complex complementary colors. Natural dyes from plants may contain dozens of compounds and their proportions vary with soil type and weather. The use of natural dyes in textile coloration has been gaining an increased popularity globally, most likely because of environmental concerns, eco-safety, and pollution control. Yarn that is dyed with natural dye such as madder shows a subtle variation of color under the microscope. Yarn that is dyed with the synthetic equivalent of madder (alizarin and purpurin) does not have a wealth of color variation and appears more uniform [1]. However, larger dye quantities and longer dyeing times are required by natural dyes, and the poor fastness properties of almost all color ranges hinder and limit their development and use, particularly in industry.

In this context, various research studies have focused on natural dyes and their application in dyeing natural textiles [2, 8]. *Mohd Yusuf et al.* examined the effect of color and fastness properties of wool fibers that were dyed with *Rubia cordifolia* as a natural dye and *Acacia catechu* as an anchoring agent. The colorimetric (CIE L*a*b*) and fastness properties were improved considerably by using *A. catechu* as a biomordant and *R. cordifolia* as a natural dye, which may be a profitable option in industrial and commercial spheres [9]. *Cristae et al.* investigated the effect of some commonly used antioxidants and ultraviolet absorbers on the light fastness of madder, weld, and woad on cotton yarn. They found that vitamin C and gallic acid were most effective in improving the natural dyes [10].

However, few scientific works exist regarding the mixing of natural dyes. The aim of this study was to study the mixing of two natural dyes, indigo (*Indigofera tinctoria*) and madder colorants (*Rubia tinctorum*), in one dye bath, and to compare the efficiency of this method to that of dyeing in two dye baths. The exhaustion and fixation dye rates of the two wool fiber samples were calculated for the two methods. The dyeing process was achieved by the reduction and oxidation mechanism of fixation. The washing fastness of both dyeing procedures (mixed dyeing in one and two dye baths) was tested and compared.

2. Experimental

2.1. Materials

2.1.1. Wool Fiber Features

Wool fiber was from Boujaâd in Morocco. White fleece was compacted and homogenized into a medium-weight fleece of 1.5–3 kg and the fiber fineness was 50–60 using the Bradford scale [7, 8].

2.1.2. Natural Dye

Alizarine and other anthraquinone dyes were extracted from the *R. tinctorum* plant, which grows in southeast Morocco [11]. The extraction method was based on the enzymatic hydrolysis (endogenous conversion) of the dried and powdered root of the plant. The indigo dye was extracted from *Indigoferra tinctoria*, and purchased from the Association Couleur Garance, Lauris-France.

2.1.3. Chemicals

Alkali reagents, sodium hydroxide (NaOH), sodium carbonate (Na_2CO_3), sodium bicarbonate ($NaHCO_3$), and the acidic reagent, acetic acid (CH_3COOH), were of analytical grade and were from Lobachemie Company (Mumbai, India), and VWR Prolabo Chemicals Company (Fontenay-Sous Bois, France), respectively. A Marseille soap type was prepared from vegetable oil, and was from a mini supermarket. Sodium chloride (NaCl) was of analytical grade and was from Solvachim (Casablanca–Morocco).

2.1.4. Argan's Pulp

The reducing agent, Argan's pulp, was collected from around an Argan tree from Essaouira in South Morocco. This natural product was composed of 20% reducer sugar, 13% cellulose, 6% protein, 2% fat, and 4% latex (comprised of 86% *cis*-polyisoprene: rubber) [12, 13].

2.1.5. Spectrophotometry

An ultraviolet–visible spectrophotometer (Thermo, Helios Epsilon) was used at 325–1100 nm with a spectral bandwidth of 1 nm.

2.1.6. pH Meter

A Henne AD1000 pH meter was used. This multimeter can measure pH, oxidation–reduction potential, and temperature.

2.1.7. Bath

A 250-mL flask was used. Heating was by a thermostat hotplate (Scilogex MS-H280-Pro).

2.1.8. Filter

The filter that was used in this study is a metallic sieve (1–5-mm diameter).

2.2. Dyeing Process with Indigo and Madder in Two Baths

2.2.1. Preparation of the Reducer

Argan's pulp (30 g) was added into 100 mL of distilled water and heated at 95°C for 30 min. The extract was filtered by using a metallic sieve.

2.2.2. Preparation of Indigo Vat

Indigo (0.1 g) was placed in 100 mL of distilled water that contained 1.2 g of sodium carbonate and 1.2 g of sodium bicarbonate, at pH 10.86 and 45°C for 30 min and at a ratio of 1/100. Then 4 g of fructose (100 mL of prepared extract from 30 g of Argan's pulp) and 1 g of NaCl were added.

2.2.3. Dyeing in the Indigo Vat

Wool yarn (1 g) was soaked and wrung before being placed in the indigo vat. The dyeing conditions were 100 mL at 45°C for 30 min and a ratio of 1/100.

2.2.4. Preparation of Madder Extract Dyes

Plant sample (5 g) was stirred in 100 mL of water at 45°C for 90 min. The solution was filtered using a metallic sieve and well-preserved to the dyeing phase. The residue was dried and weighed to calculate the approximate amount of dye that exists in the initial madder plant.

2.2.5. Preparation of Madder Dye Bath

The dyeing conditions for the madder dye bath were 100 mL of dye (extract prepared from 5 g *R. tinctorum* in 100 mL water), 1.2 g sodium carbonate, and 1.2 g sodium bicarbonate, pH 9.65, at 60°C for 30 min and with a ratio of 1/100.

2.2.6. Reduction of Madder Dye Bath

The reducing conditions of the madder dye bath were 100 mL of prepared madder dye bath, 3 g of fructose (or 30 g of Argan's pulp extract), 10 g/L NaCl at 45°C for 30 min and with a liquid ratio of 1/100.

2.2.7. Dyeing in the Reduced Madder Dye Bath

Wool yarn (1 g) that had been dyed with indigo was placed in the reduced madder dye bath, at 45°C for 30 min with a liquid ratio 1/100.

2.2.8. Spectral Analysis

Spectrophotometer calibration

Spectrophotometer calibration was achieved by using a standard solution that was prepared according to the mass of wool yarn, the concentration of reducer (fructose or Argan's pulp), NaCl, and alkali (NaOH, sodium carbonate, and sodium bicarbonate) that was added to the dye baths.

Measurement of dye exhaustion and fixation rate

1 mL was removed of solution from each dye bath for measurement. Each sample was diluted to 10 mL using the prepared standard solutions. The absorbance measurements are shown in Tables 1 and 2. The absorbances were measured at 380 nm.

2.3. Dyeing Process with Indigo and Madder in One Bath

2.3.1. Preparation of Indigo Vat

Indigo (0.05g) was stirred in 50 mL distilled water at 45°C for 10 min to reduce the indigo grain size.

2.3.2. Preparation of Madder Colorants Bath

Madder plant (5g) was macerated in 100 mL distilled water at 40°C for 2 h, then filtered by using a metallic sieve.

Only 50 mL was taken from the prepared extract for use in the mixture dyeing with indigo.

2.3.3. Preparation of Reduced Mixed Dye Bath

The reducing conditions of the mixed dye bath were 100 mL prepared indigo (50 mL) and madder colorants (50 mL) in the dye bath, 3 g fructose (or 30 g of Argan's pulp extract), 1 g NaCl, 1.2 g sodium carbonate, and 1.2 g sodium bicarbonate, pH 10.73, at 45°C for 30 min and a liquid ratio of 1/100.

2.3.4. Dyeing in the Mixed Dye Bath

Wool yarn (1 g) was soaked and wrung before being placed in the mixed dye bath. The dyeing conditions were 100 mL at 45°C for 30 min and a liquid ratio of 1/100.

2.3.5. Cold Rinse

Rinsing of the samples (for mixed dyeing in one and two baths) was conducted at the end of the dyeing process to remove dyes from the fiber and inter-surfaces and to neutralize the alkaline medium.

2.3.6. Oxidation and Rinse

Oxidation was achieved for samples in open air for 15 min for the rinsing phases. At the end of the dyeing process, two successive rinses were conducted in cold water.

2.3.7. Acidification (Neutralization)

Samples were treated in acidic solution at pH 6.5 with CH_3COOH (30%).

2.3.8. Drying

Samples were dried in a sterile environment at 60°C and 80°C.

2.3.9. Soaping

Samples were treated with 0.6 g/L Marseille soap at 60°C for 15 min, with a liquid ratio of 1/100. The soaping step was used to test the washing fastness of the samples that were dyed in the mixed dye bath.

2.3.10. Washing Fastness

The washing fastness was determined at 40°C over 30 min according to ISO 105-C6:A1S [14].

3. Results and Discussion

3.1. Exhaustion Rate of Indigo in Dye Bath

The absorbances and concentrations of the initial and final dye bath are presented in Table 1.

Table 1. *Exhaustion rate of indigo in dye bath.*

	Initial dye bath	Final dye bath	Exhaustion rate ((Ci-Cf)/Ci)*100 (%)
Indigo absorbance	0,213	0.125	66.24
Indigo concentration (g/L)	0.631	0.213	

The calculated concentration of indigo (0.631 g/L) was lower than the experimental concentration that was introduced into the dye bath (1 g/L). This can be explained by the incomplete solubility of indigo at pH 10.86.

3.2. Exhaustion Rate of Madder Colorants in Dye Bath

The absorbances and concentrations of the initial and final dye baths are presented in Table 2.

Table 2. *Exhaustion rate of madder colorants in dye bath.*

	Initial dye bath	Final dye bath	Exhaustion rate ((Ci-Cf)/Ci)*100 (%)
Madder colorants absorbance	0.526	0.409	22.14
Madder colorants concentration (g/L)	45.34	35.30	

The exhaustion rate of the madder colorants was lower because of the high alkaline medium. The alizarine and other anthraquinones in the madder plant are soluble in the high alkaline medium.

3.3. Exhaustion Rate of Indigo and Madder Colorants in Mixed Dye Bath

The concentrations of the initial and final dye bath are presented in Table 3.

Table 3. *Exhaustion rate of indigo and madder colorants in mixed dye bath.*

	Initial dye bath	Final dye bath	Exhaustion rate ((Ci-Cf)/Ci)*100
Concentration of indigo and madder colorants in one dye bath (g/L)	18.84	16.98	10.0%

The exhaustion rate was considerably lower. This may result from the low absorbance of the madder colorants towards the fiber in the high alkaline medium.

3.4. Exhaustion Rate Comparison of Indigo and Madder Colorants in One and Two Dye Baths

The summary of measurements of the exhaustion rate of indigo and madder colorants in one and two dye baths are given in Table 4.

Table 4. *Comparative exhaustion rates of indigo and madder colorants for dyeing in one and two dye baths.*

Samples	Exhaustion rate of indigo in the first dye bath ((Ci-Cf)/Ci)*100	Exhaustion rate of madder colorants in the second dye bath ((Ci-Cf)/Ci)*100	Exhaustion rate of indigo and madder colorants in one mixed dye bath ((Ci-Cf)/Ci)*100
Dyeing in two dye baths	66.2%	22.1%	-
Dyeing in one dye bath	-	-	10.0%

It was concluded that the mixed dyeing in two dye baths with indigo and madder colorants achieved a more intense and uniform coloration. The results were attributed to the difference in optimal dyeing conditions between the indigo and madder colorants. The pH of the medium was the most important factor that caused this difference. Therefore, indigo required a high alkaline medium to achieve complete solubility in a dye bath. In contrast, the madder colorants had a lower substantivity towards wool fiber in this high alkaline medium.

3.5. Fixation of Indigo and Madder Colorants in Two Dye Baths

Dyes that were removed from the sample that were dyed in two baths (indigo and madder colorants) during the soaping phase were very little in the dyeing process, and therefore, they were not taken into account in the calculation of the fixation dye rate. Complete fixation was achieved.

3.6. Fixation of Indigo and Madder Colorants in One Dye Bath

Dyes that were removed from the sample that was dyed in one dye bath during the soaping phase were very little during the dyeing process, therefore, it was believed that complete fixation was achieved. For both methods, the high dye fixation was attributed to the insoluble form of dyes inside the fiber, which was formed after oxidation and achieved important physicochemical interactions (hydrogen bonds) and physical interactions (Van der Waals interactions) with the fiber.

3.7. Washing Fastness

Measurements that were obtained for the two soaping samples are presented in Table 5.

Table 5. *Washing fastness of indigo and madder colorants for dyeing in one and two baths.*

Samples	Washing fastness (assessing staining)						Washing fastness (assessing change in color)
	WO	PAC	PES	PA	CO	CA	
Dyeing in two baths	5	5	5	5	5	5	4–5
Dyeing in one bath	5	5	5	5	5	5	4–5

The values from these experiments confirm the solid fixation of dyes as described previously.

An excellent washing fastness was achieved for the combined method. In fact, the reduction method (vat system) of the indigo and madder colorants in the combined method improved the diffusion and fixation on the fiber and converted the dye into a non-soluble molecule inside the fiber. Both methods achieved a similar fastness. This was explained by the seam fixation mechanism (reduction and oxidation reactions).

4. Conclusions

This study confirmed the incompatibility of mixing indigo and madder colorants in one dye bath. However, dyeing in two dye baths yielded a higher dye exhaustion rate. This result was attributed to the different optimal dyeing conditions of the indigo and madder colorants. The indigo required a high alkaline medium to achieve complete solubility in the dye bath. In contrast, the madder colorants had a lower substantivity towards wool fiber in such a high alkaline medium. Both dyeing methods (mixed dyeing in one and two dye baths) achieved a rigid fixation of dyes and an excellent degree of washing fastness. This behavior may result from the fixation mechanism of indigo and madder colorants, which is based on the reduction and oxidation reactions. After oxidation, the dyes were converted to an insoluble form inside the fiber by the formation of stable interactions (hydrogen bonds and Van der Waals interactions) with different amino acids of the wool fiber.

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