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DYEING ANALYSIS WITH BIO-MORDANT SOURCE IN ORGANIC COTTON DYEING WITH MADDER (*Rubia tinctorum* L.)

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ABSTRACT

This study aimed at investigating organic cotton fabrics dyed with five different percentages (5 %, 10 %, 20 %, 40 % and 80 %) of the roots of madder (*Rubia tinctorum* L.) via the shellacs of gall oak (*Quercus infectoria* Olivier; 1 %, 2%, 4%, 8 %, 16 %) mordant. In this case, the natural dyeing was completely performed without a chemical mordant. For this reason, this dyeing can be called environmentally friendly. A reversed - phase high performance liquid chromatography with diode array detection method (RP-HPLC-DAD) was utilized for the identification of natural dyes (anthraquinones and tannins) present in the natural - dyed organic cotton extracts. The naturally dyed cotton samples were studied calorimetrically and their colour coordinates L^*, a^*, b^*, C^*, h, K/S and ΔE^* values were determined. At the same time, the view related to the organic cotton fabric was taken by a scanning electron microscope (SEM).

KEYWORDS: Madder, Gall oak, HPLC, Colour, Organic cotton, Tannin.

1. INTRODUCTION

Cotton is an attractive substance to use in natural dyeing. For natural dyeing, tannin - containing plants can be used as a mordant before dyeing of the fabrics [1]. In terms of dyeing, the natural red colour from plants are mostly anthraquinone compounds. These anthraquinone compounds have been used to dye textiles for a very long time [2]. Madder (*Rubia tinctorum* L.) dye plant is a red colour source that contains a lot of anthraquinones compounds (such as alizarin, purpurin, *etc.*). This plant has been used in the natural dyeing (the dyeing of wool, silk, and cotton) since ancient times [3]. Their root parts contain about 1.9 % of dye compounds [4]. The dyeing analyses relating to the dyeings of silk samples with madder plants have been reported[5]. In another work, madder and gall oak plants were used to dye silk fabrics in the past. The dyed silk fabrics were analyzed to identify the dyes using HPLC-DAD. In addition to this, their fastness and colour values were also investigated [6]. At the same time, this plant was also used for making lake pigments until the nineteenth century. Thus, it is an old dye plant [7]. The madder lakes from these plant compounds were also obtained, previously [8]. The extract including the dyes of madder is very popular due to being durable and requested. For this reason, the madder plant was mostly used in works relating to natural dyeing [9]. The shellacs of gall oak (*Quercus infectoria* Olivier) plant contain tannin compounds [10]. The shellacs can include tannin structures up to 35 % [1]. The gall oaks including tannin compounds have antibacterial and antifungal activities [11]. Their extracts include principally ellagic acid. This compound has an affinity in terms of the dyeing of materials because of the presence of -OH groups in its structure [12]. The tannin compounds obtained from these plant shellacs have no negative effects on the environment. They can be used for both the mordanting and dyeing of the fibres. In addition, this plant was also used to produce the lake pigments including the compounds such as gallic acid and ellagic acid [10]. The dyeing analyses of silk samples of madder and gall oak plants were recently realised. The tannin and anthraquinone compounds in this natural dyeing were used to colour silk samples [11].

In recent years, organic cotton fabrics have been dyed with gall oak (*Quercus infectoria* Olivier) and barberry (*Berberis vulgaris*) plants using microwave irradiation and conventional methods. Then, the colour

measurements and the analyses of the dyed fabrics have been realised [13]. Alternatively, the gall oak plant has been used to dye organic cotton fabrics in another study. Also, different analytical and technical methods have been performed [14]. In another study, cotton dyeing with weld dye plants in the textile industry was evaluated in terms of sustainability and mass production [15].

In this work, fifteen organic cotton fabrics were coloured by the dried gall oak (*Quercus infectoria* Olivier) and madder (*Rubia tinctorum* L.) plant. The cotton samples used madder (*Rubia tinctorum* L.) plant (5 %, 10 %, 20 %, 40, and 80 %) and the gall oak (*Quercus infectoria* Olivier) (1%, 2%, 4%, 8% and 16 %) plant in this situation.

The dyes present in the mordanted and the natural dyed–cotton fabric extracts were determined by HPLC-DAD. At the same time, the colour measurements (L*, a*, b*, and K/S, *etc.*) relating to the vegetable-dyed – cotton samples were made. The SEM photograph of the organic cotton fabric sample was taken.

2. MATERIALS AND METHODS

Cotton, Plants, and Chemicals

Organic cotton knitting fabric, gall oak (*Quercus infectoria* Olivier), and madder (*Rubia tinctorum* L.) plants were provided by the Turkish Cultural Foundation, Cultural Heritage Preservation, and Natural Dyes Laboratory (Istanbul, Turkey). 100 % organic knitting interlock cotton fabric was used in this study and the weight of the fabric was 230 g/m^2. Hydrochloric acid (HCl) and methanol (CH_3OH), acetonitrile (CH_3CN), and trifluoroacetic acid (TFA) were obtained from Merck (Germany). High-purity water was obtained by passing water through a Milli-Q treatment system (Millipore, USA), and the HPLC mobile phase was prepared using Milli-Q water.

Apparatus

A water bath (Nüve NB 20), a heater (WiseStir M S-H-20D), and weighing instruments (Radwag AS 220/C/2; Ohaus, Pioenner Item: PA4102C) were used in this study.

Treatment with soap of organic knitting cotton fabrics

The organic cotton fabrics (ten pieces) were made ready for the study. The ten pieces of cotton fabrics were weighed (nearly 70 g). One piece of the cotton fabric was approximately 7.0 g and at the same piece was measured nearly 15 cm (width) x 22 cm (height). Then, the cotton fabrics (70 g) were added to water (cotton fabric quantity/water quantity: 1/10). The cotton fabrics were added to a bath containing 5 g soap/L. They were kept at 100°C (±2) for 30 min. After this time, the treated cotton fabrics were removed from the beaker. Then, they were washed with distilled water. After that, they were dried at room temperature [16].

Mordanting procedure

All treated organic cotton samples were separately mordanted with gall oak (1 %, 2 %, 4 %, 8 %, and 16 %) except the direct madder – dyed samples which were without the mordant. The mordant baths were made in the ratio 20:1 for all mordanting. The wet soaped cotton fabrics were added at 30ºC to the bath. For example, for 7 g of organic cotton fabric, the water quantity was 140 ml. The mordanting time for this work was designated as 30 min. In this time, the temperature of the mordant baths was taken to 70°C. Each mordanted cotton fabric was taken from each beaker after the mordanting and they were then washed with distilled water. Then, the mordanted - cotton samples were squeezed and dried in the open air at room temperature. The gall oak–mordanted organic cotton fabrics (1 %, 2 %, 4 %, 8 %, and 16 %; five pieces) were used for both the mordanting without the dyeing and before the dyeing with madder.

Dyeing procedure

In this study, for each one dyed (madder) organic cotton sample, the dye plant % was taken as 5 %, 10 %, 20 %, 40 %, and 80 %. In short, the mordanted or/and dyed organic cotton samples were fifteen samples. Nevertheless, the only madder-dyed cotton samples were five samples (5 %, 10 %, 20 %, 40 %, and 80 %) without mordanting. The liquor ratios related to the dye baths were the same with the mordant baths (20:1). The percentage of the dye plants was taken via the fabric quantity (for example; for 1 g organic cotton fabric sample, it was 0.05 g madder plant (5 %)). In addition, the dyeing time was 30 min at 70°C. After this time, the natural dyed–organic

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cotton fabrics taken from the dye baths were washed together with distilled water and squeezed and dried in the open air at room temperature.

In Table 1, in an applied dyeing procedure, it can be seen that mordant (%) and madder (%) plant percentages are given. This table shows dyeing properties for cotton fabrics dyed with the plants. Besides, it indicates us that madder values were more according to the gall oak plant as a percentage. For a procedure in this table, it can be said that after plants were mixed, the dyeing was made ready for the study. For example, 1 % gall oak and 5 % madder plants were mixed to dye cotton fabric sample number 11 in the same bath. For 11 to 15 sample numbers, other mixing procedures were followed in the same way. The liquor ratios were the same in the baths (20:1). The wet soaped cotton fabrics were added at 30ºC in the dye bath. The dyeing time was 30 min at 70°C. After this time, the natural dyed–organic cotton fabrics taken from the dye baths were washed together with distilled water and squeezed and dried in the open air at room temperature.

Table 1. *Dyeing properties for cotton fabrics dyed with the plants*

Sample no.	Mordant (Gall oak) (%)	Dye (Madder) (%)	Liquor ratio	Temperature (°C)	Time (min)
1	1	-	20:1	70	30
2	2	-	20:1	70	30
3	4	-	20:1	70	30
4	8	-	20:1	70	30
5	16	-	20:1	70	30
6	-	5	20:1	70	30
7	-	10	20:1	70	30
8	-	20	20:1	70	30
9	-	40	20:1	70	30
10	-	80	20:1	70	30
11	1	5	20:1	70	30
12	2	10	20:1	70	30
13	4	20	20:1	70	30
14	8	40	20:1	70	30
15	16	80	20:1	70	30

Color measurements

The colour measurements relating to the natural dyed organic cotton fabrics were carried out using a Konica Minolta CM-2300d Software Spectra Magic NX. Table 2 shows dyeing parameters and K/S values, according to the percentages of gall oak and madder. According to the CIEL*a*b* (1976), the colour measurements were realised. At this point, the measurements relating L*, a*, b*, C*, h, and ΔE* were carried out with using a portable spectrophotometer coupled to a PC under D65 illuminate/10° observer. At the same time, K/S values of the natural dyed organic cotton fabrics were also measured [1]. Colorimetric data obtained for the gall oak or/and madder dyed cotton fabrics are presented in Table 3.

Table 2*. Dyeing parameters and K/S values, according to the percentages of gall oak and madder*

Sample No	Gall oak (%)	Madder (%)	K/S
1	1	-	0.290205
2	2	-	0.380835
3	4	-	0.79615
4	8	-	1.94566
5	16	-	3.283635
6	-	5	0.51106
7	-	10	0.958325
8	-	20	1.220805
9	-	40	1.99228
10	-	80	2.2242

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11	1	5	0.702255
12	2	10	1.251405
13	4	20	1.70026
14	8	40	2.462385
15	16	80	3.01098
Untreated (soaped) cotton			0.192895

K/S values were taken at 360 nm.

Table 3 . *Colorimetric data obtaining for the madder or / and gall oak - dyed organic cotton fabrics*

Sample No	L*	a*	b*	C*	h	ΔL*	Δa*	Δb*	ΔE*ab
1	89.35	0.545	10.935	10.95	87.165	-1.43	0.434	0.40	1.545
2	88.97	0.775	11.55	11.575	86.16	-1.805	0.67	0.21	1.935
3	86.695	1.13	12.97	13.015	85.03	-4.08	1.02	1.63	4.51
4	83.68	1.42	15.885	15.945	84.895	-7.095	1.315	4.545	8.53
5	81.715	1.92	17.8	17.905	83.85	-9.06	1.815	6.465	11.275
6	80.86	12.015	15.905	19.93	52.915	-9.92	11.915	4.565	16.16
7	74.765	17.765	19.55	26.415	47.74	-16.01	17.655	8.21	25.21
8	71.615	17.46	20.89	27.225	50.11	-19.16	17.355	9.55	27.555
9	65.97	21.87	24.01	32.48	47.67	-24.805	21.76	12.675	35.35
10	64.07	22.435	24.965	33.565	48.05	-26.705	22.325	13.62	37.38
11	80.05	12.055	18.09	21.745	56.32	-10.73	11.95	6.755	17.42
12	75.205	15.16	22.085	26.785	55.535	-15.57	15.05	10.745	24.17
13	72.0	16.645	24.985	30.02	56.325	-18.775	16.54	13.645	28.5
14	68.145	18.47	26.82	32.565	55.445	-22.63	18.365	15.48	33.0
15	66.37	19.175	28.2	34.1	55.785	-24.41	19.065	16.88	35.265
Undyed	90.775	0.105	11.335	11.335	89.475	-	-	-	-

HPLC Instrumentation

The instrumentation and the HPLC elution program were as described in an earlier report [1].

Extraction procedure for HPLC analysis

The extraction procedure for HPLC analysis was as described in an earlier report [17]. Dyed cotton fabrics were weighed at an average of 10 mg.

Scan Electron Microscopy Analysis

Before the analysis, the 1x1 cm sample, piece which is attached with a carbon conductive band to the stub, was coated with Au-Pd alloy for 120 seconds under high vacuum with a Leica Ace 200 model device by a directional sputtering method. The prepared coated sample was scanned with a Carl Zeiss Sigma 300 VP model field emission scanning electron microscope at 15 kV acceleration voltage. Zeiss In-Lens secondary electron detector was used for imaging under high vacuum conditions with Polaroid 545 magnification reference. Figure 1 shows magnification of 3.00 KX (A), 500 X (B), 250 X (C), and 500 X (D) of organic cotton fabric.

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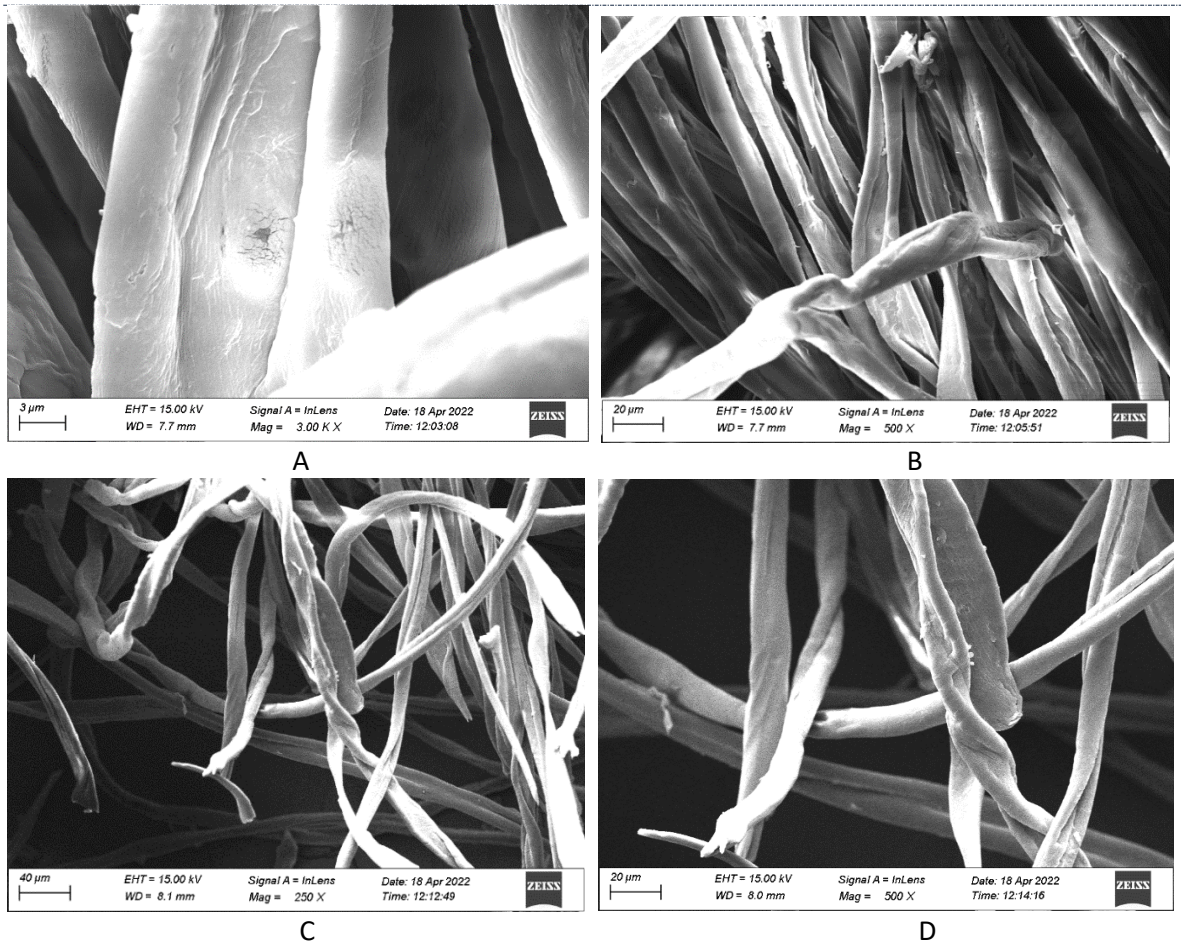

Figure 1. Magnification of 3.00 KX (A), 500 X (B), 250 X (C) and 500 X (D) of organic cotton fabric

3. RESULT AND DISCUSSION

Colour analysis

As can be seen in Table 3, the highest lightness value belongs to the undyed cotton fabric (undyed sample), excluding the samples. The highest lightness (L*) value among the samples belongs to the sample number 1. This value was 89.35. In this point, 1 % gall oak plant was used to dye cotton fabric. Conversely, the lowest lightness (L^*) value belongs to the sample number 10. The value concerning it was 64.07. At this point, cotton fabric was dyed with 80 % madder plant. Complete contrast between the lightness values was detected between sample 1 (89.35) and sample number 10 (64.07). The lowest red and green (a^*) value was given in sample number 1. This lowest value was 0.545. Here, 1 % gall oak plant was used to dye cotton fabric. The highest value was also obtained with sample number 10. The obtained value was 22.435. Complete contrast between the red and green values was detected between sample number 1 (0.545) and sample number 10 (22.435). The lowest yellow and blue value (b^*) were determined as 10.935. The dyeing (1 % gall oak, sample number one) here was realised. The highest yellow and blue value (b^*) was 28.2. The dyeing here was with together 16 % gall oak and 80 % madder plant (sample number fifteen). Complete contrast between the yellow and blue values was detected between sample number 1 (10.935) and sample number 15 (28.2). The lowest chroma (C*) value was given for sample number 1. This value was 10.95. The highest chroma value was also obtained with sample number 15 (16 % gall oak + 80 % madder). The value was 34.1. Among all samples, the highest value of ΔE^*_{ab} belongs to sample 10. The lowest one belongs to the number 1 sample. The wavelength of maximum absorption for the work was taken as λ_{max} = 360 nm. Table 2 shows three types of dyeings groups. The highest K/S values were obtained from the sample numbers 5, 14, and 15. The lowest K/S value was 0.290205. Dyeing for this value was performed with a 1 % gall oak plant. In the table, the K/S values of the dyeing procedure in three groups are given. The K/S values

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in each of these groups increased themselves from beginning to end. K/S values obtained from curves with gall oak (*Quercus infectoria* Olivier) and madder (*Rubia tinctorum* L.) plants are presented in Figure 2 and Figure 3. K/S values obtaining mixing of gall oak and madder plants are given in Figure 4. h values obtained with using of gall oak (%) plant are also given in Figure 5. Although the percentages of plants used in mordanting obtained with gall oak plants were low, the h values were determined to be quite high.

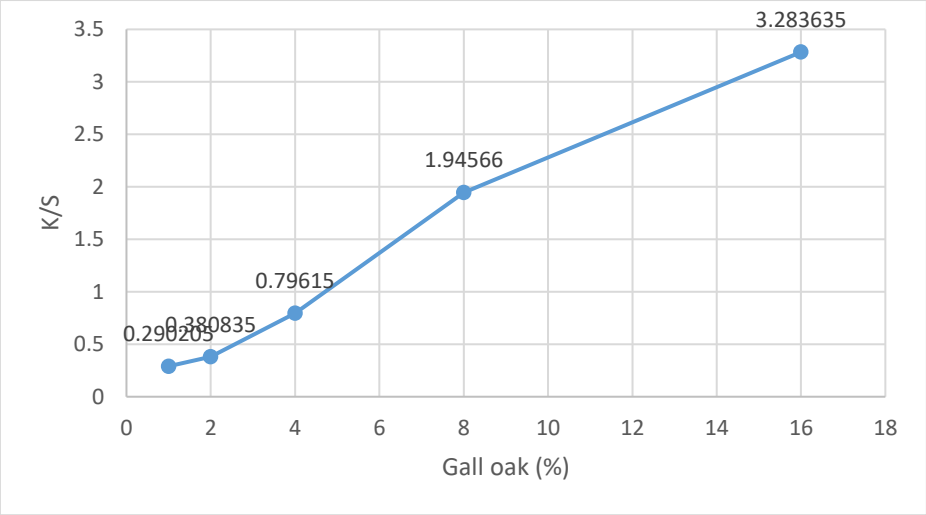

Figure 2. K/S values with gall oak (Quercus infectoria Olivier) plant

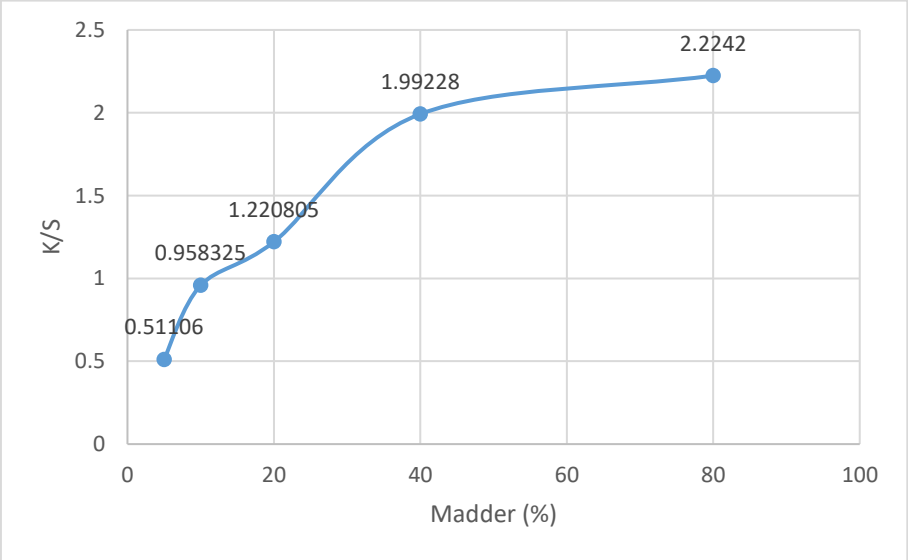

Figure 3. K/S values with madder (Rubia tinctorum L.) plant

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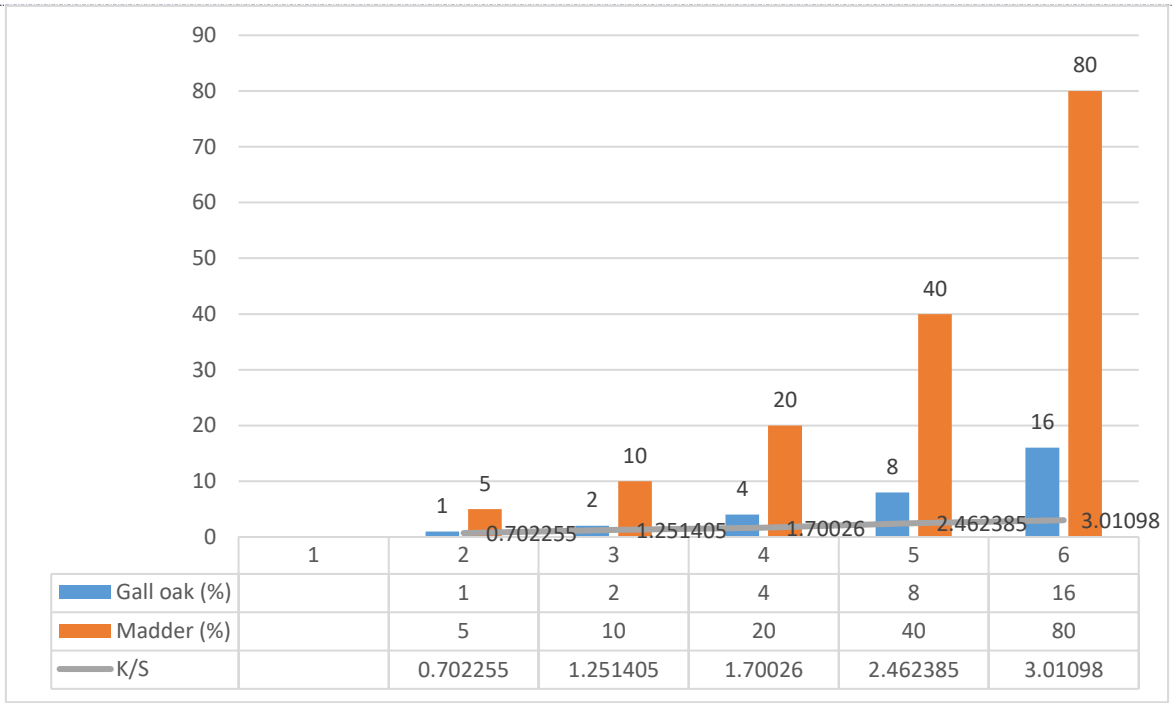

Figure 4. K/S values obtained by mixing of gall oak and madder plants

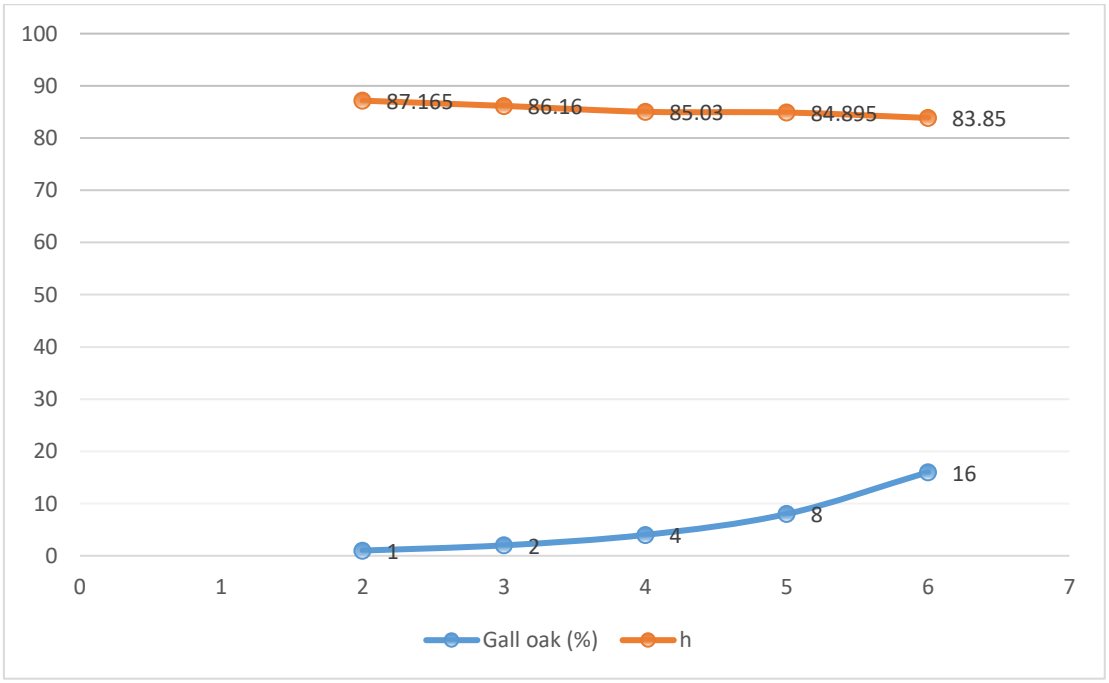

Figure 5. h values obtained by using gall oak (%) plant

HPLC Analysis

The extracts obtained from cotton samples dyed with gall oak (*Quercus infectoria* Olivier) and madder (*Rubia tinctorum* L.) plants were analysed by using RP-HPLC with a DAD detector. At the same time, the natural dyes in the acid hydrolysed natural dyed-organic cotton fabrics were qualitatively identified. Tannins (gallic acid or/and

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ellagic acid) and anthraquinones (alizarin, purpurin, munjistin, *etc*.) compounds in the natural dyed-organic cotton fabric extracts were investigated in this work. HPLC-DAD chromatogram of 80 % madder dyed organic cotton fabric is given in Figure 6. Photodiode array spectra of identified natural dyes in Figure 6 are given in Figure 7. In addition to these, the HPLC-DAD chromatogram of 16% gall oak + 80 % madder dyed organic cotton fabric is shown in Figure 8. Photodiode array spectra of identified natural dyes in Figure 8 are given in Figure 9. Besides, the HPLC-DAD chromatogram of 16 % gall oak dyed organic cotton fabric is shown in Figure 10. Photodiode array spectra of identified natural dyes in Figure 10 are given in Figure 11. Chromatograms were selected according to their peak intensities. The values of peak heights to identified dyes in cotton samples dyed with dye plants are given in Table 4.

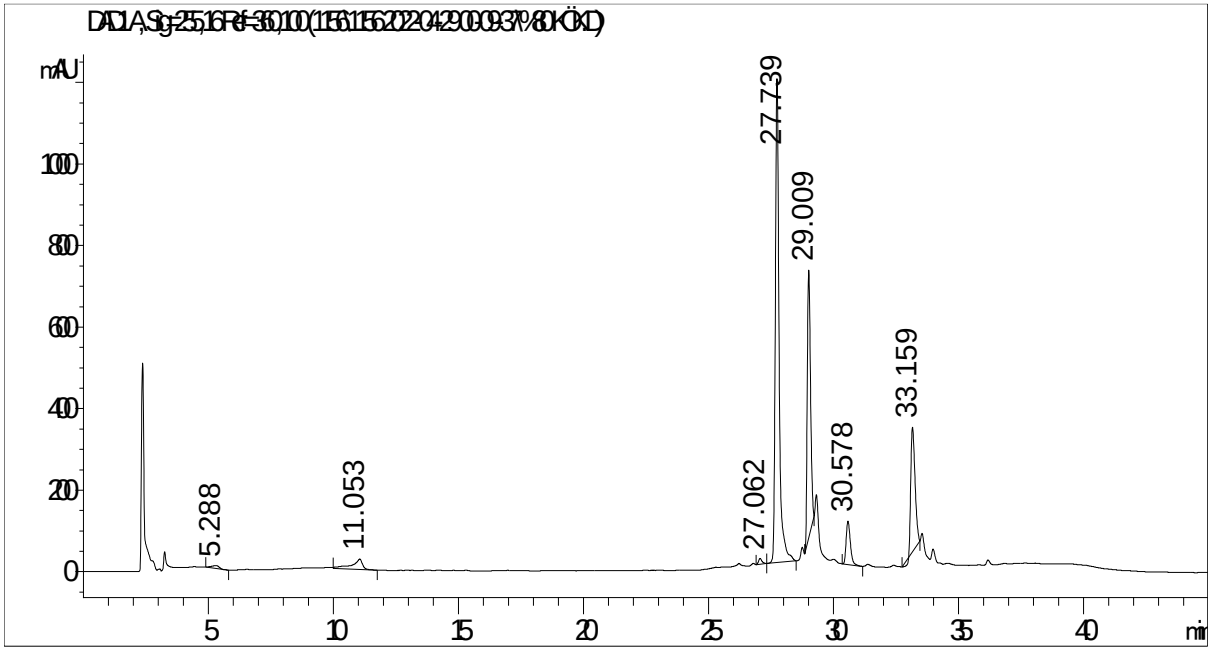
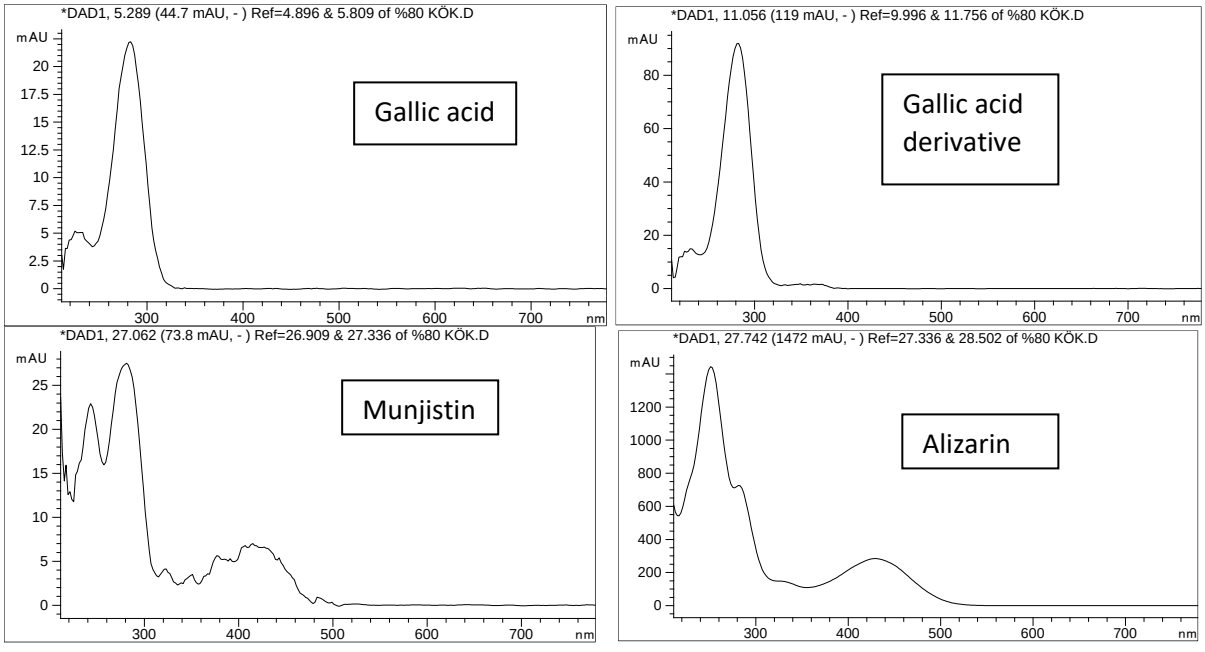


Figure 6. HPLC-DAD chromatogram of 80 % madder dyed organic cotton fabric



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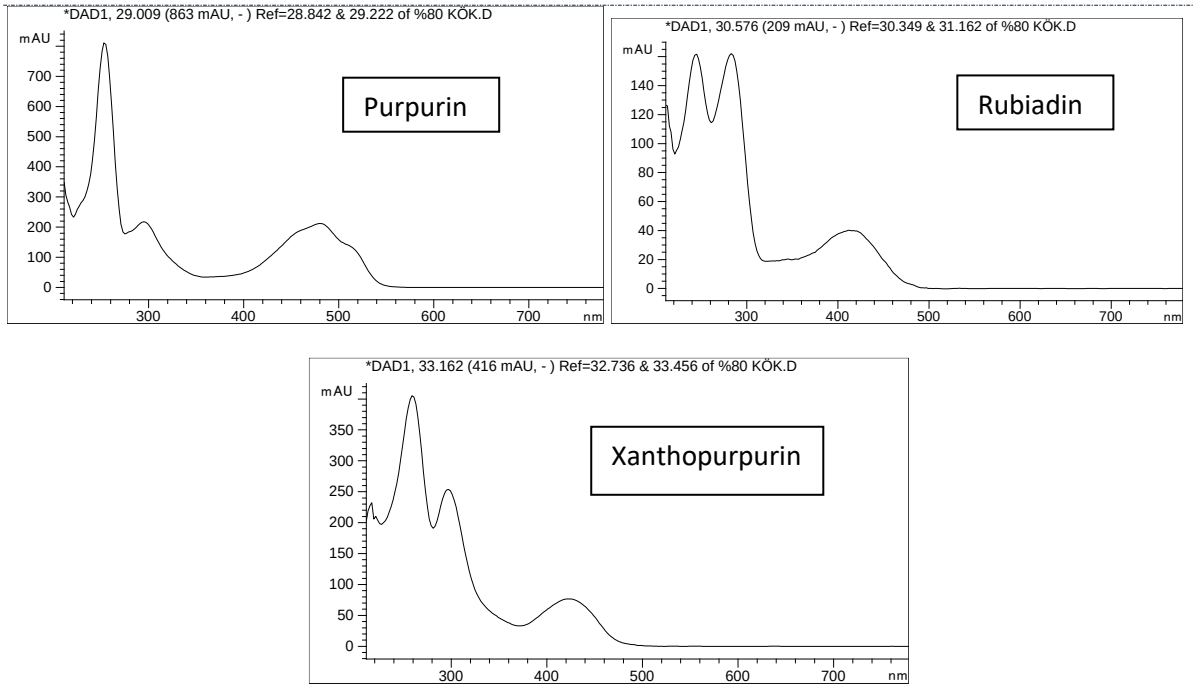

Figure 7. Photodiode array spectra of identified natural dyes in Figure 6

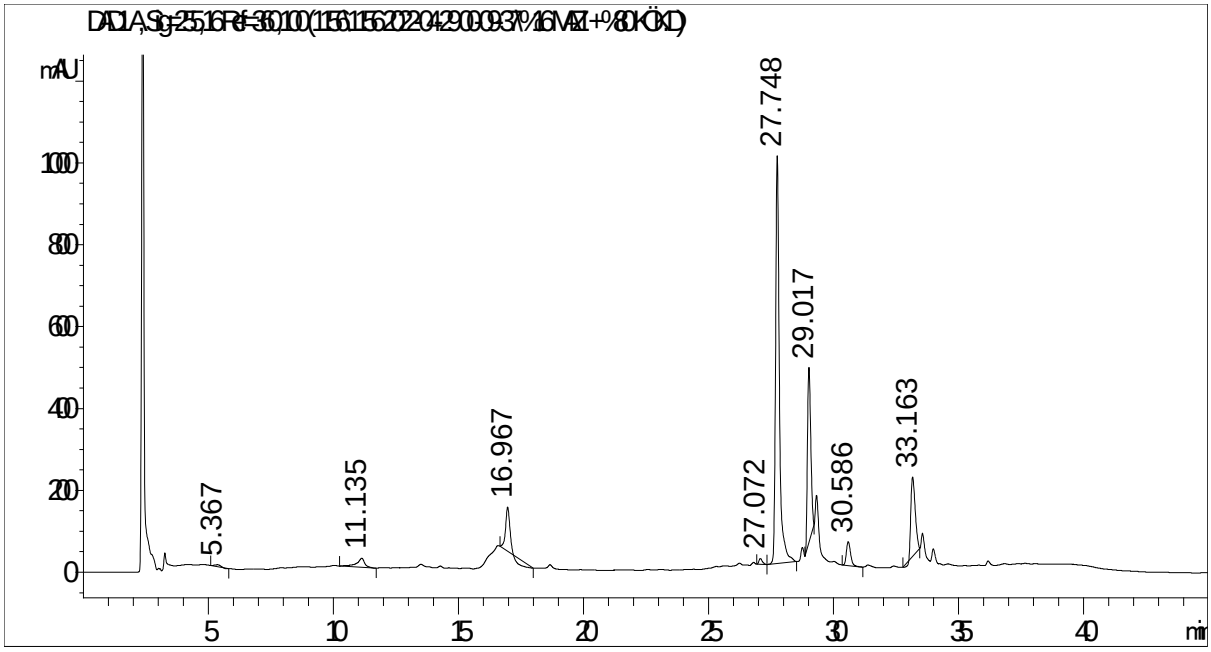

Figure 8. HPLC-DAD chromatogram of 16 % gall oak + 80 % madder dyed organic cotton fabric

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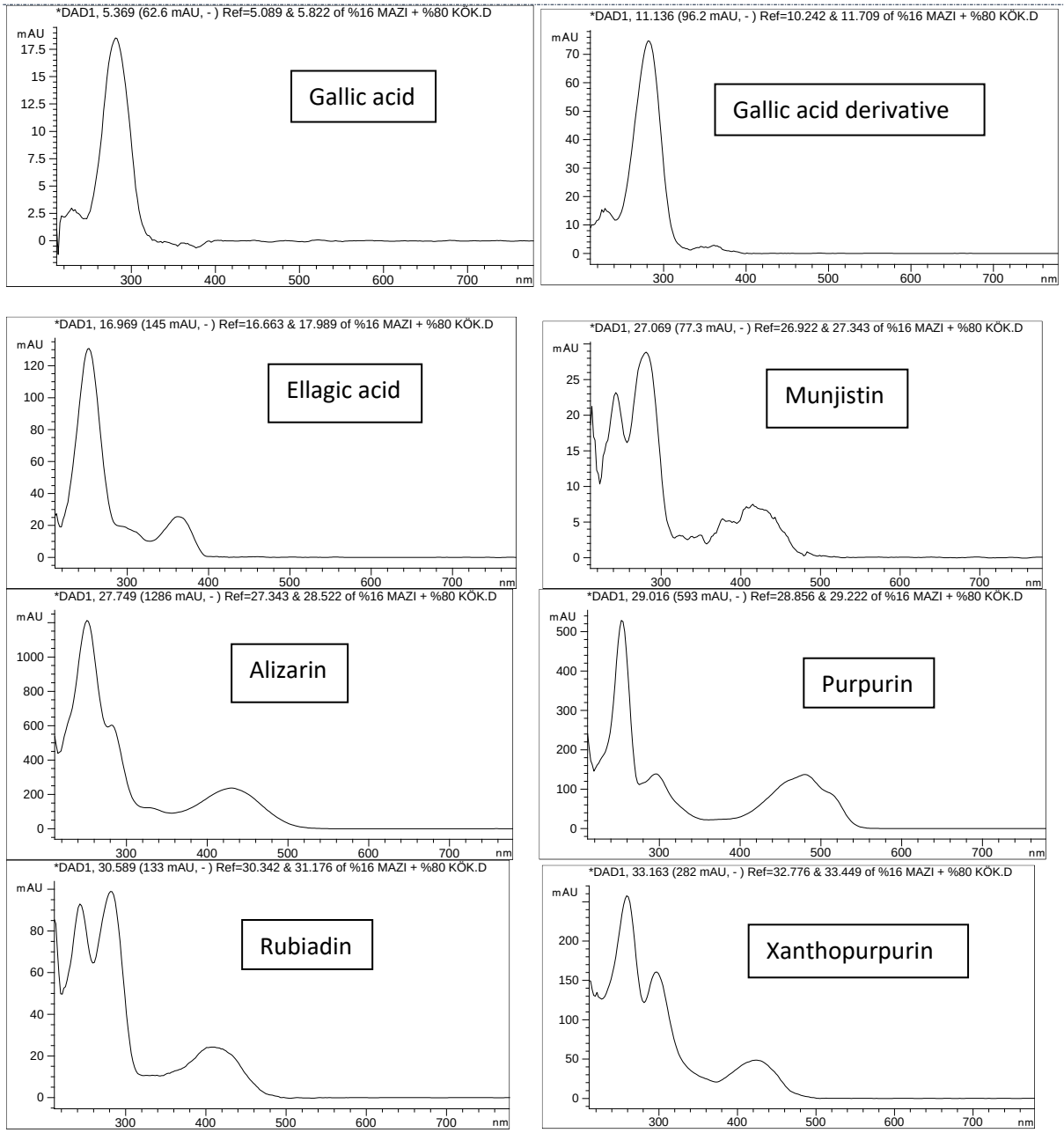

Figure 9. Photodiode array spectra of identified natural dyes in Figure 8 [11,18]

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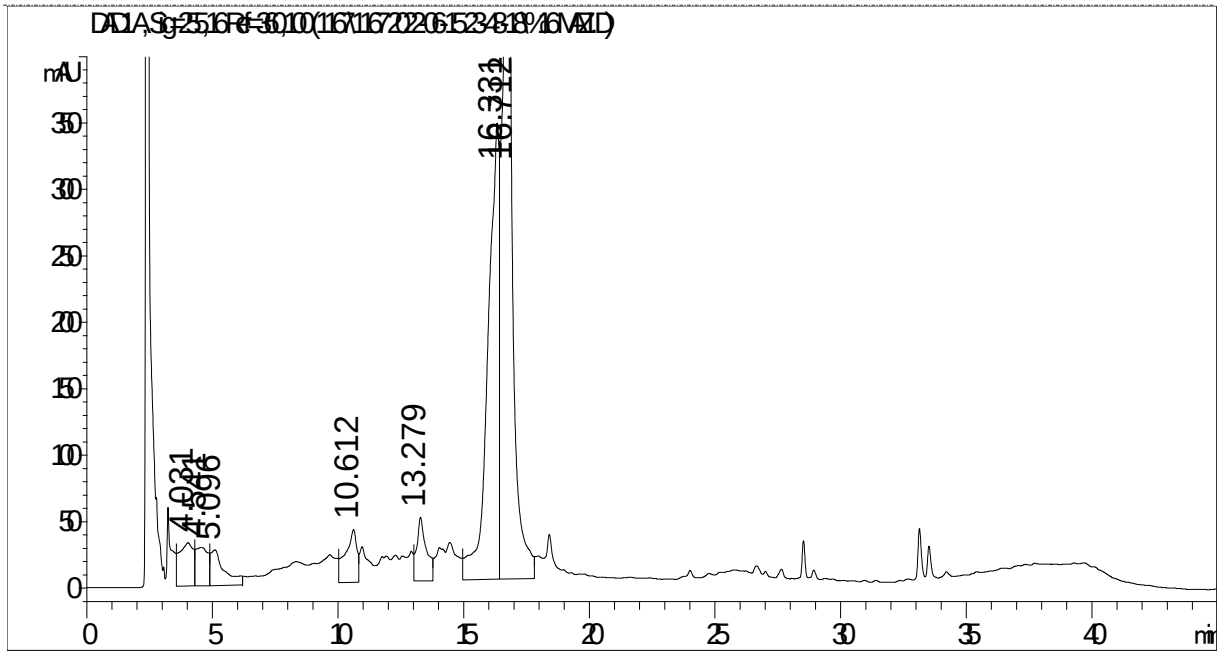
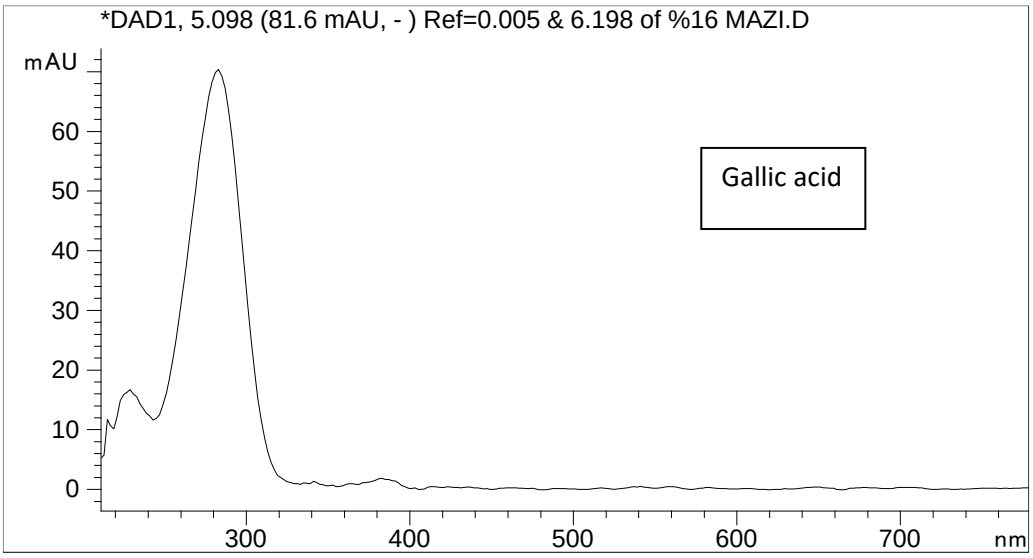


Figure 10. HPLC-DAD chromatogram of 16 % gall oak dyed organic cotton fabric



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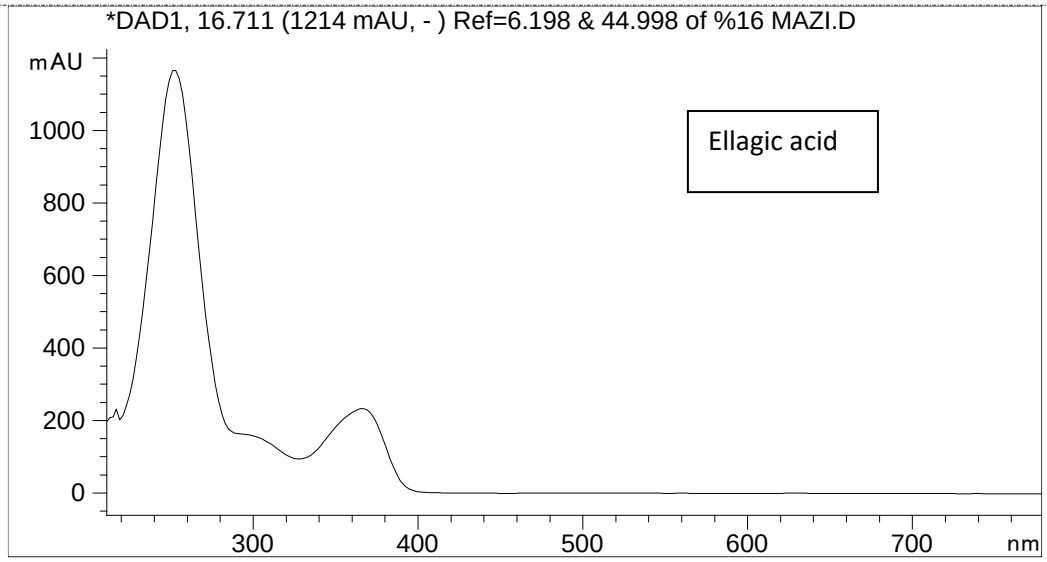

Figure 11. Photodiode array spectra of identified natural dyes in Figure 10

Table 4. *The values of peak heights to identified dyes in cotton samples dyed with dye plants*

	Alizarin	Munjistin	Purpurin	Rubiadin	Xanthopurpurin	Gallic acid	Gallic acid derivative	Ellagic acid	Ellagic acid derivative
5% madder	1801,5	2,3	70,9	28,7	29,9	0	0	0	0
10% madder	618	5,5	312,3	45,5	188,2	0	0	0	0
20% madder	752,4	6,2	384	51,4	208	0	0	0	0
40% madder	912,4	6,5	420,2	66,2	286	12,5	20,2	0	0
80% madder	1187,2	14,3	656,6	106,5	304,8	6,6	25,5	0	0
1% gall oak +5% madder	182,5	2,1	72,4	29,4	30,7	3,1	5	18,9	0
2% gall oak + 10% madder	301	3,8	123,8	49,3	51,9	4,2	8,1	29,8	0
4% gall oak + 20% madder	502,1	5,1	208,6	80,2	80,6	6,4	15,4	54,5	0
8% gall oak + 40 % madder	898,4	8,5	365,8	154,7	145,4	10,2	26,3	92,6	0
16 % gall oak + 80% madder	996,4	14,2	427,5	58,5	194	4,7	22,6	107,4	0
1% gall oak	0	0	0	0	0	29.6	18.9	0	4.9 / 5.8
2% gall oak	0	0	0	0	0	43,2	30,4	0	7,6 / 8
4% gall oak	0	0	0	0	0	79,2	53,7	0	12,6 / 15,3
8 % gall oak	0	0	0	0	0	15,2	17,3 / 24,5 / 26,4	711,4	38,6 / 302,8
16% gall oak	0	0	0	0	0	26,9	28,8 / 32,6 / 39,8	950,1	47,9 /342,5

4. CONCLUSION

Consequently, organic cotton fabrics can be dyed with natural dyes (gall oak or/and madder) by using three dyeing procedures. After all, I believe that colouring organic cotton fabrics with natural dyes can provide an important advantage for environmental processes. In this study, the effects of tannin and tannin derivatives on h values were observed. In this context, very high h values were obtained only in gall oak dyeing. This will be a guide for future studies. As a result of these dyeings, natural wastes were formed and environmentally friendly dyeing was realized

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