Textile & Leather Review

ISSN 2623-6281 | www.tlr-journal.com | 10.31881/TLR

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How to cite: Buyukakinci BY, Karadag R. Optimization of the natural dyes extraction (Madder and Wallonia oak) and Cotton Dyeing Using Microwave Irradiation. Textile & Leather Review. 2022; 5:451-462. https://doi.org/10.31881/TLR.2022.42

How to link: https://doi.org/10.31881/TLR.2022.42

Published: 21 September 2022

Optimization of the natural dyes extraction (Madder and Wallonia oak) and Cotton Dyeing Using Microwave Irradiation

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Article
https://doi.org/10.31881/TLR.2022.42
Received 23 June 2022; Accepted 11 September 2022; Published 21 September 2022

ABSTRACT
*In this work, microwave irradiation was used to extract two dye plants (Quercus ithaburansis Decaisne and Rubia tinctorum L.) and to dye natural cotton. Using microwave (MW) energy, the extraction time for these plants and the dyeing time for organic cotton fabric were optimized. The colouring compounds were analysed in the extractions and dyed fabrics by reversed-phase high-performance liquid chromatographic method coupled with a photodiode array detector (RP-HPLC PAD). CIEL*a*b* spectrophotometer measured the colour characteristics of all the dyed fabrics. Scanning electron microscopy with energy dispersion spectroscopy (SEM-EDX) was used to image the dyed organic cotton fabrics. The colouristic and colourfastness properties of the dyed fabrics were investigated. According to the test results, it has been determined that the 5 minutes of extraction and dyeing time using with microwave irradiation method is very eligible. During these processes, energy and timesavings were achieved by eco-friendly MW irradiation and minimized cotton damage.*

KEYWORDS
extraction, dyeing, microwave, natural dyes, cotton

INTRODUCTION

Madder (*Rubiatinctorumb L.*) and Walloon oak (*Quercus ithaburansis*) are two important dye plants for natural dyeing. Madder has been used for red colour since ancient times. It has been used in many archaeology and historical textile. The main colouring compounds of madder are alizarin, purpurin, rubiadin, munjistin, etc. The colouring compounds were identified in the historical textile. Acorn caps of Walloon oak were used for dyeing. Because of its high tannin content, this oak has been important for dyeing and tanning for ages [1]. The main colouring compounds of acorn cups are gallic acid, ellagic acid, and their derivatives. In recent years, madder and acorn caps of Walloon oak have been used for sustainable natural dyeing [2]. Antifungal activity and durability of madder and gall nuts were investigated in silk fabrics [3]. The colour changes and fastness properties of Walloon oak and Anatolian buckthorn were evaluated in the dyed silk fabrics [4]. Many studies were made about madder and Wallonia oak by the conventional dyeing method for silk fabrics [5]. The microwave irradiation method has recently been used in dyeing and natural dye extraction [6,7].

The infrared radiation between the 1-100 cm wavelength and 300-30000 MHz radio frequencies refers to Microwaves [8]. These high-frequency waves are capable of penetrating materials and causing heat to have occurred in the process. The electric field energy is converted into heat through the dielectric losses of the dipolar material. Microwaves (MW) can be absorbed, diffracted, and reflected. After microwaves are absorbed by the materials, very low energy units or photons remain. This energy is not enough to break down the molecular and atomic structures. Thus the MW energy is not harmful to the textile materials and doesn't defeat their structure [9,10]. Thanks to microwave heating, it is possible to dye textile materials homogeneously in an immediate time, because molecular diffusion is more effective during the MW irradiation process [8-11]. Many studies have been reported on MW energy used for textile dyeing. MW irradiation can penetrate all the particles of material instantly and in uniform. It is cost-effective and environmentally friendly [6,7,9,12].

In the textile sector, many uses for microwave energy have been investigated, such as heating, drying, dyeing and finishing processes, dye fixation, and printing of fabrics; but limited work is found in the literature, especially on the extraction of natural plants and dyeing natural cotton fabric, in which microwaves are used as a heat source [6,7,13,14]. The conventional extraction and dyeing process requires long and continuous operation times at high temperatures.

In the textile sector, natural production and sustainability are important for a cleaner world. So, many researchers are studying the natural dyeing of textiles. In the natural dyeing proses, Ultrasonic energy was also used to increase colour strength and sustainability [15]. Rehman et al. used MW energy in plant-based dyeing of silk fabric and extraction of natural plants. The high colour strength and colour fastness were provided [16].

Adeel et al. also conducted a similar study using MW irradiation for the dyeing of nylon fabric and achieved positive results [17].

In the present study, two plant species, *Quercus ithaburansis Decaisne* and *Rubia tinctorum L*. were extracted and dyed natural cotton fabrics using MW irradiation. Unlike classical natural dyeing processes [15-17], no mordant was used in this study. Mordant is a dye fixative that binds dyes on fabrics by forming a coordination complex between dyes and textile molecules. In this study, surface modification and dye fixation on the cotton fabric were strengthened using MW irradiation.

The dye extracts and dyed samples were analytically and technically examined. MW energy was preferred not only to achieve energy and time savings but also to minimize cotton damage during the process. In addition, this heating source enables dye molecules to diffuse into the textile fabrics more strongly [12].

EXPERIMENTAL

Materials and Methods

In this study, 100% organic knitting cotton fabric (230 g/m^2) was obtained from Cultural Heritage Preservation and Natural Dye Laboratory, Turkey. Dried dye plants (madder and Wallonia oak) were purchased from Raw Materials of Natural Dyes Co., Turkey. Nonionic detergent (nonylphenol ethoxylates) was supplied by ZAG Kimya, Turkey, and used as a scouring agent for natural cotton fabric. A Samsung model MS23K3515AW microwave oven (max. 800 watts; 2450 MHz) was used for extraction and dyeing.

Extraction and dyeing process

Quercus ithaburansis Decaisne and *Rubia tinctorum L*. were crushed to the powder form and then were extracted (10 g of the powder in 100 mL water) at 90 °C using microwave irradiation (100 Watt). Various extraction periods (5, 10 and 15 minutes) were studied to find the optimal value. In the end, the solution was filtered off and left to cool down. After scouring cotton fabrics with nonionic detergent (0.5%) in the bath with Fabric to Liquor ratio (F:L) 1:20 (w:v), they were dyed without using any metal mordant. Dye solutions extracted from plants (*Quercus ithaburansis Decaisne* and *Rubia tinctorum L*.) were used for dyeing organic cotton samples. The dyeings were conducted in a 250 mL glass beaker in atmospheric conditions using microwave irradiation. The cotton fabric to dye-bath ratio was 1:20 (w:v) and the plants' percentage in the dye solution was 10%. The energy power was 100 W. The samples remained at 60°C for five minutes in the dye bath, and then they cooled. After dyed samples were flushed using tap water and allowed to dry in the open air. Samples were evaluated for colour strength and fastness properties. The colourfastness tests were conducted by washing and rubbing [18,19].

RP-HPLC-PAD analysis

Chromatographic measurements were carried out using an Agilent 1200 series system (Agilent Technologies, Hewlett-Packard, Germany), including G1322A Degasser, G1311A Quat pump, G1329A autosampler, G13166 TCC, and G1315D Diode Array Detector. DAD detection is performed by scanning from 191 to 799 nm with a resolution of 2 nm, and the chromatographic peaks were monitored at 255, 268, 276, 350, 491, 520, 580 and 620 nm. A Nova Pak C18 analytical column (39 × 150 mm, 4 µm, Part NoWAT086344, Waters) was used. Analytical and guard columns were maintained at 30°C and the data station was the Agilent Chemstation [2]. A Gradient elution program [13] was utilized for chromatographic separations of the hydrolysed samples [17].

Colour measurement and colourfastness

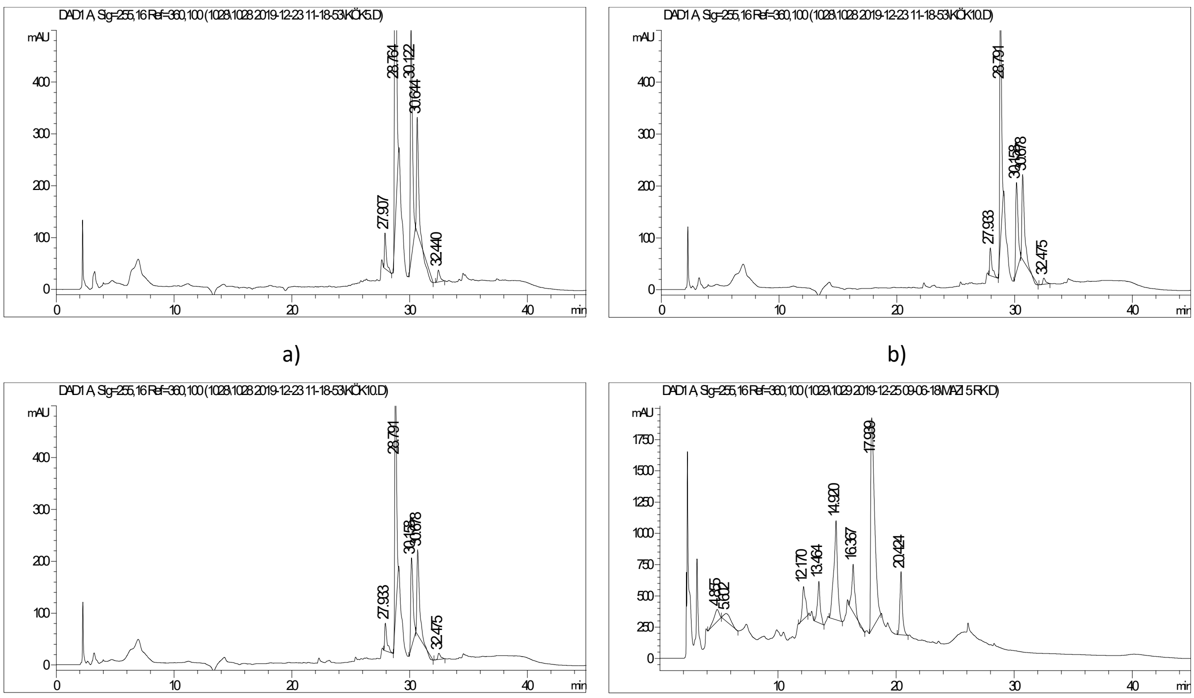
Colour measurements of the dyed samples were carried out using DATA COLOR Spectrophotometer. In CIEL*a*b* system, L* corresponds to brightness, a* to the red-green coordinate, b* to the yellow-blue coordinate, and C to chroma/relative saturations. It was equipped with an illuminant of D 65 10° observer [11]. The colourfastness properties of washing and rubbing were rated according to ISO standard methods ISO 105-C06 (A1S) and ISO 105-X12, respectively.

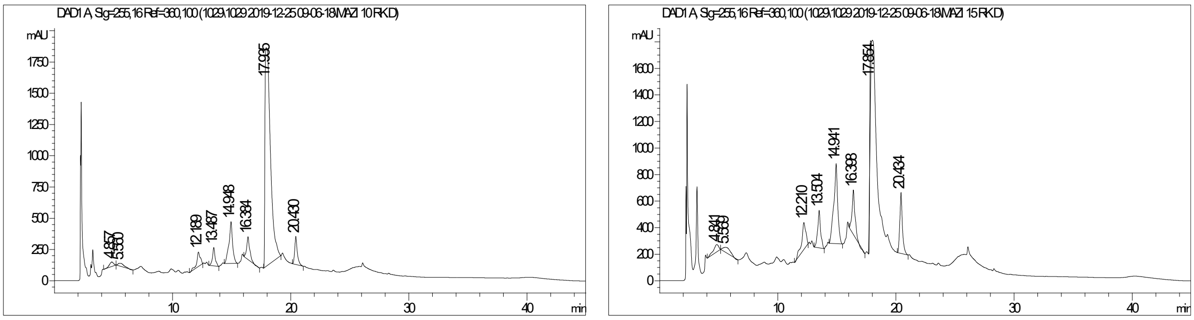
SEM-EDX analysis

The undyed and dyed cotton fabrics were analysed by Scanning Electron Microscope equipped with Energy Dispersive X-ray Spectrometer (SEM-EDX) for surface imaging and elemental analysis. TESCAN VEGA3 SEM (Brno, Czech Republic) and Bruker EDX detector (Massachusetts, ABD) were used. Analytical protocols of SEM-EDX are given in previous studies [6,7,20-22].

RESULTS AND DISCUSSION

In this study, two different plants were extracted using MW energy at three different periods. The results of Chromatograms for the dye extracts are given in Figure 1.





e) f)

Figure 1. Chromatography of dyeplantextracts (a)*Rubiatinctorum*extract (5 minutes); b) *Rubiatinctorum*extract (10 minutes); c) *Rubiatinctorum*extract (15 minutes); d) *Quercus ithaburansis*extract (5 minutes); e) *Quercus ithaburansis*extract (10 minutes); f) *Quercus ithaburansis*extract (15 minutes)

According to the extraction periods and identified colouring compounds, the percentages of their dye extracts (in the dye bath) are shown in Figures 2 and 3. Gallic acid, its derivatives, ellagic acid, and its derivatives were detected in the *Quercus ithaburansis* extracts (Figure 1). Antragallol, alizarin, purpurin, munjistin, and rubiadin were detected in the *Rubia tinctorum* extracts (Figure 2). Alizarin and purpurinare are very important colouring compounds for red colours dyeings with madder roots. The amounts of alizarin and purpurin are higher than munjistin and rubiadin in the madder roots. Colouring compounds of alizarin and purpurin were maximally dissolved in the 5 minutes of extraction time. The amount of munjistin has increased as the extraction time has been increased. However, the amounts of alizarin and purpurin decreased accordingly during the 10 and 15 minute extraction periods. There has not been a significant change in the amounts of antragallol and rubiadin with the increase in extraction periods (Figure 2). The optimal extraction time is 5 minutes for *Rubia tinctorum.* Therefore, dyeings were performed for the 5 and 10 minute extracts.

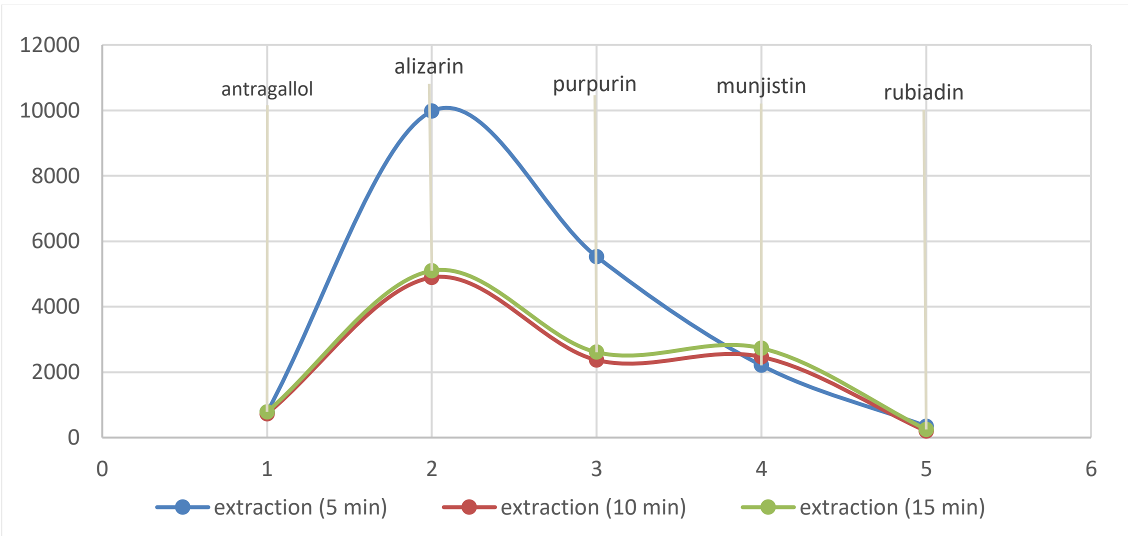


Figure 2. Identified dyestuffs and their peak (heights) in the extractions

Quantities of gallic acid, ellagic acid, and their derivatives have not significantly differed between the 5, 10 and 15 minute extraction periods. However, ellagic acid is maximum at extraction period of 5 and 10 minutes and minimum at 15 minutes (Figure 3). Ellagic acid transformed into ellagic acid derivatives in the 15 minute extraction time. Optima extraction time is 5 or 10 minutes for *Quercus infectoria.*

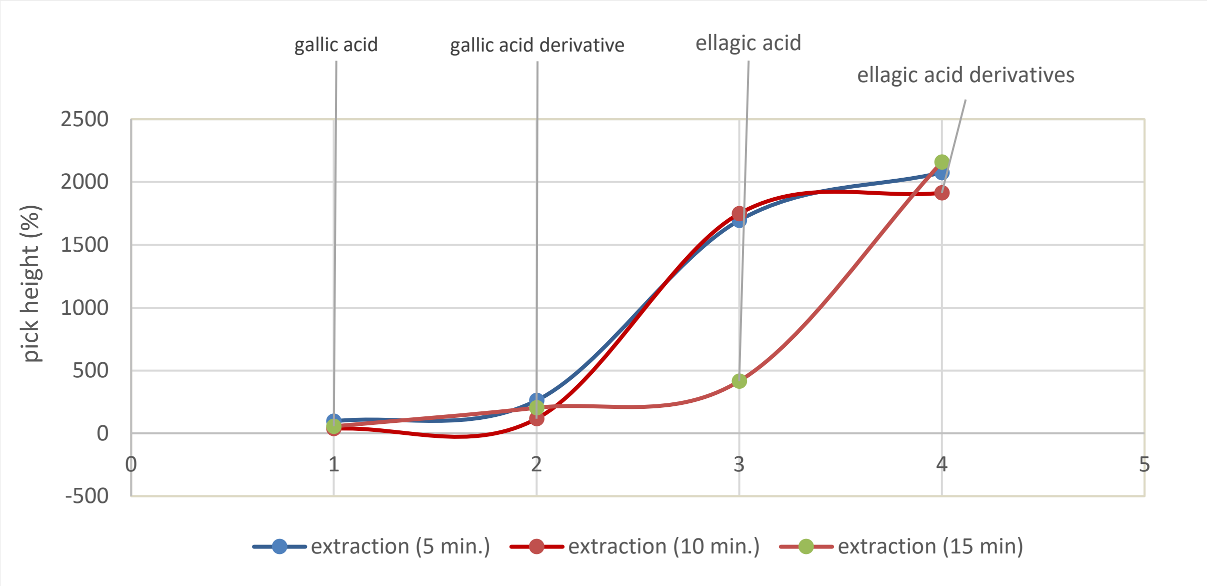


Figure 3. Identified dyestuffs and their peak (heights) in the extractions

The L*, a*, b*, C* and h values are similar in the 5 and 10 minute extraction periods for *Quercus infectoria* (Table 1).

Table 1. Colour coordinates of the undyed and dyed fabrics in the 5 and 10 minute extracts

Dye	L*	a*	b*	C*	h°
Undyed fabric	91.81	0.66	10.06	10.09	86.25
Quercus ithaburansis Decaisne (ext. time-5minutes)	76.27	2.58	25.65	25.78	84.25
Quercus ithaburansis Decaisne (ext. time-10minutes)	74.68	3.08	29.83	29.99	84.10
Rubia tinctorum L (ext. time-5minutes)	68.32	21.14	13.26	24.95	32.09
Rubia tinctorum L (ext. time-10minutes)	70.54	18.95	10.09	21.47	28.02

The colour fastness tests were conducted by washing and rubbing [19,20]. The average values of all experiments were reported (Figure 4). Colour fastness values are excellent in the 5 and 10 minute extraction periods for *Quercus ithaburansis* dyeing (Figure 4).

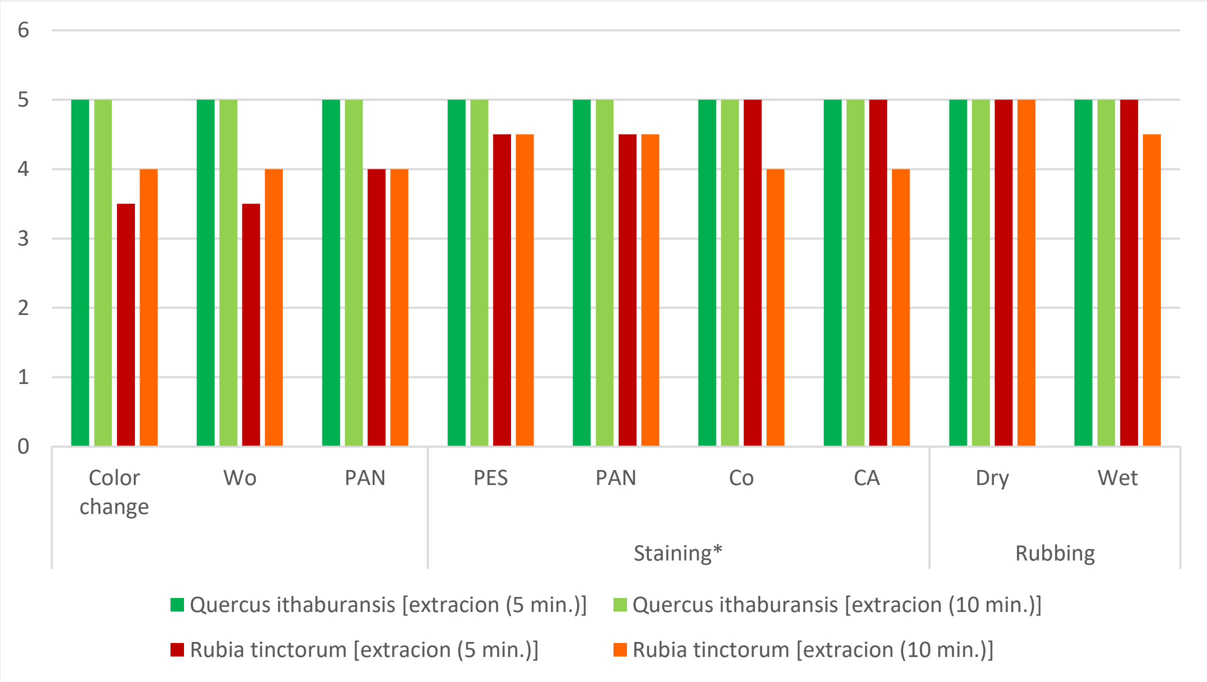
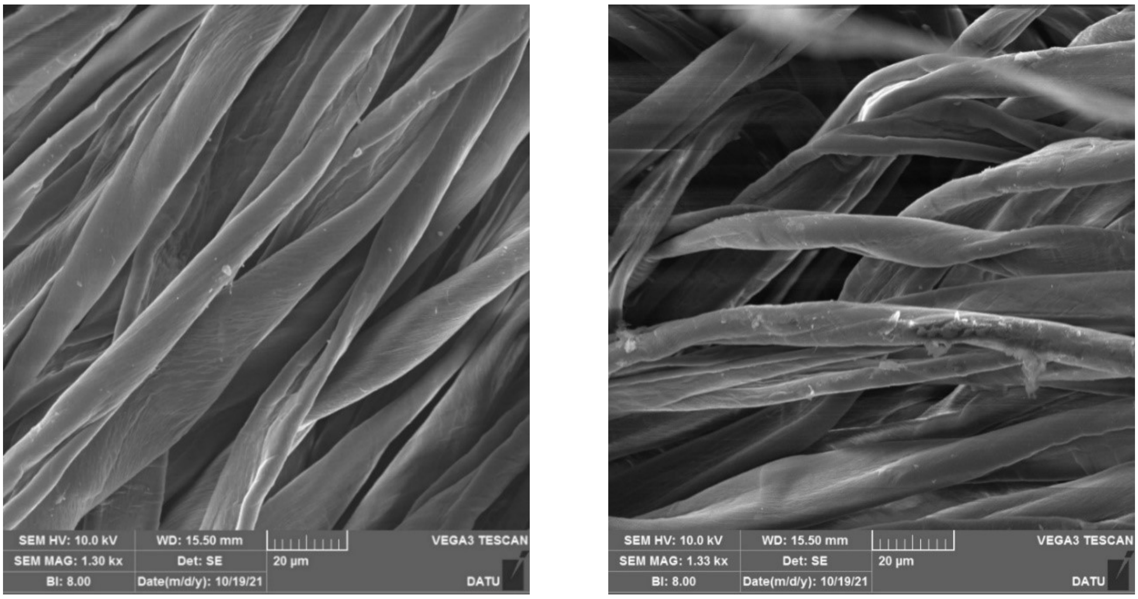


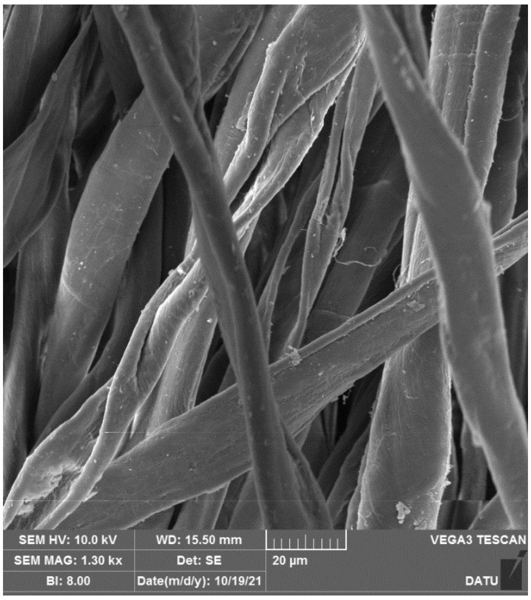
Figure 4. Colourfastness according to washing and rubbing

**CA: Cellulose acetate; Co: Cotton; PA: Polyamide; PES: Polyester; PAN: Acrylic; Wo: Wool*

The SEM images of the undyed and dyed fabrics are shown in Figure 5 and the elemental analysis results are also shown in Figure 6. It was observed that no damage occurred in microwave irradiation dyeing. It was evaluated that the Silicon (Si) and Sodium (Na) elements come from the tap water during the rinsing process or from the plants used in dyeing.



a) b)



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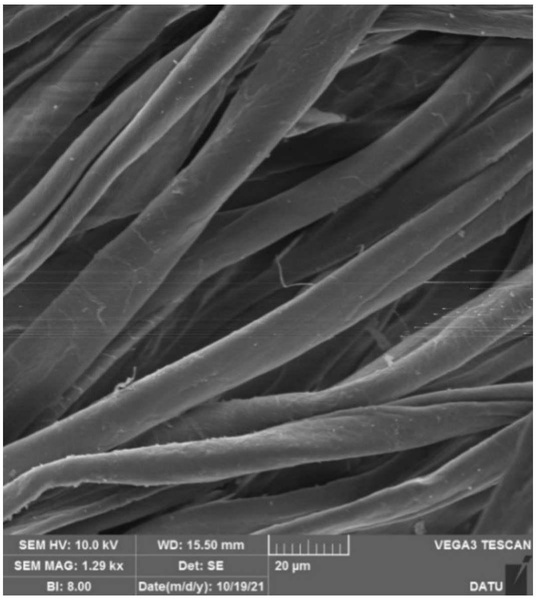
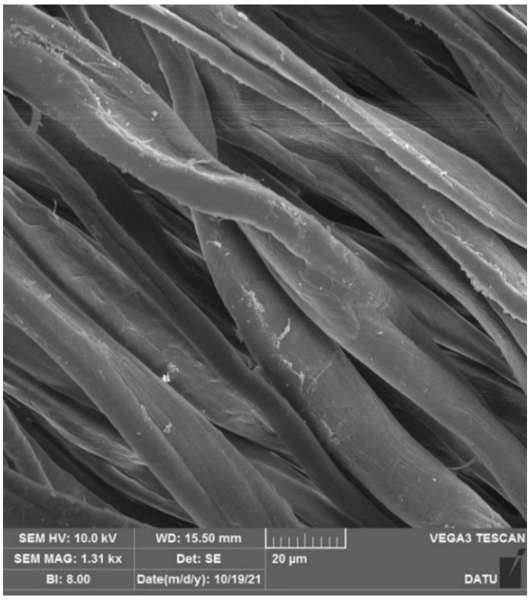


d)



e)

Figure 5. SEM imagine: (a) undyed cotton; (b) dyed cotton with *Rubia tinctorum* in 5 minute extract; (c) dyed cotton with *Rubia tinctorum* in 10 minute extract; (d) dyed cotton with *Quercus ithaburansis* in 5 minute extract; (e) dyed cotton with *Quercus ithaburansis* in 10 minute extract

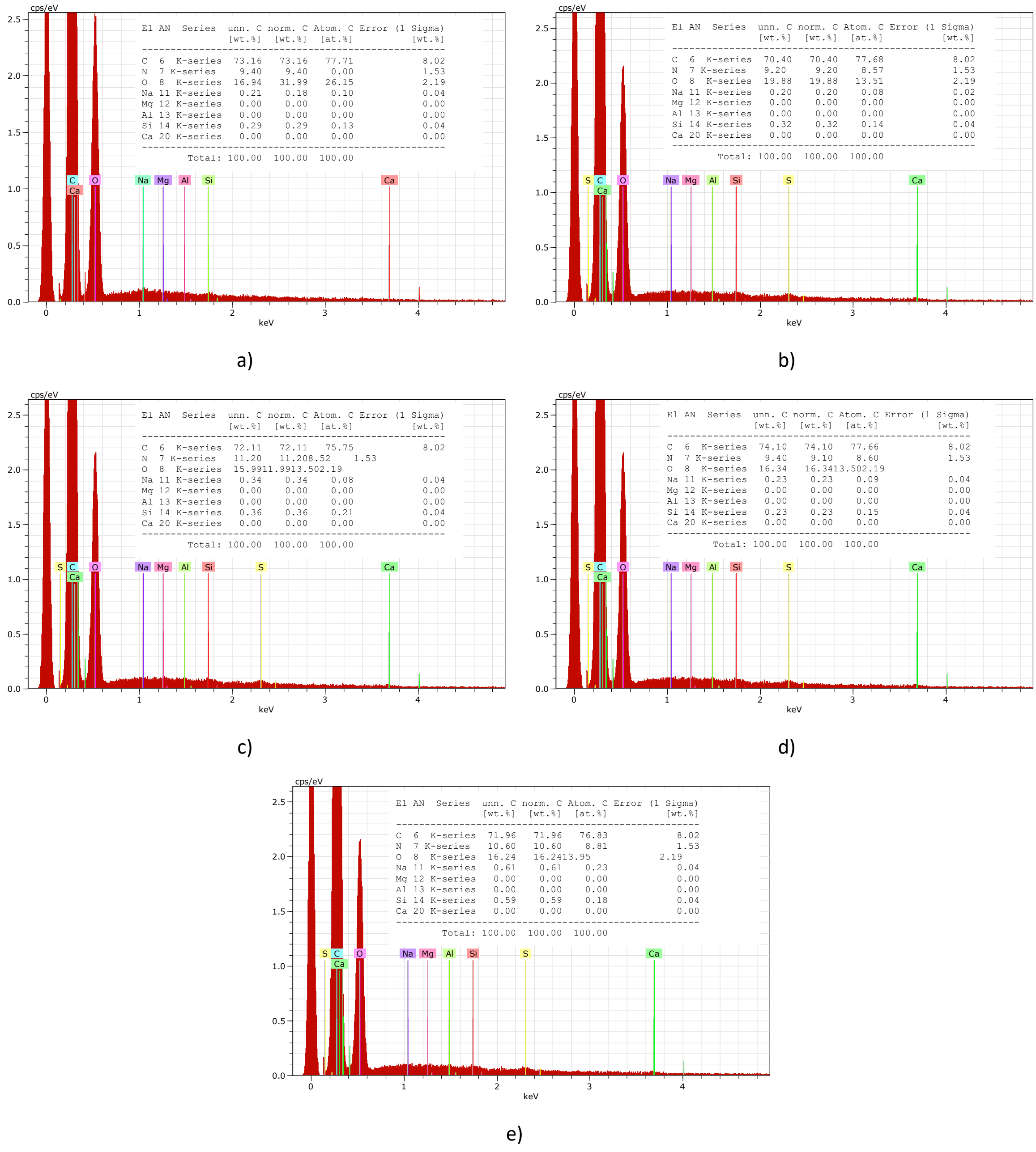


Figure 6. SEM elemental analysis: (a) undyed cotton; b) dyed cotton with *Rubia tinctorum* in 5 minute extract; c) dyed cotton with *Rubia tinctorum* in 10 minute extract; d) dyed cotton with *Quercus ithaburansis* in 5 minute extract; e) dyed cotton with *Quercus ithaburansis* in 10 minute extract

At first, gallic acid, gallic acid derivatives, ellagic acid, and ellagic acid derivatives form covalent bonds with OH groups of cellulose; then these compounds bond with antragallol, alizarin, purpurin, munjistin and rubiadin.

As a result of the analysis, only 5 and 10 minutes of extract solutions were used for dyeing cotton fabric with MW irradiation at 60°C for 5 minutes. Then, the dyed fabrics were compared in colour measurement and fastness. Looking at CIEL*a*b* and colourfastness values, similar successful results are seen in dyeing under MW irradiation for 5 minutes. The same results were also found according to RP-HPLC-PAD analyses.

CONCLUSION

In this study, two different plants were extracted using MW energy at 3 different periods. According to the results of the RP-HPLC-PAD analysis for the dye extract, the quantity of the total colouring compounds has the best values for 5 minutes compared to the 10 and 15 minute MW irradiation period. The extraction process over 10 minutes has also no significant effect. It was seen that MW energy does not have any damage to the dyed fabric.

This dyeing process is green because of using natural organic cotton fabric and natural dyes. In addition, the dyeing time and, consequently, energy consumption were reduced by using microwave irradiation.

Author Contributions

Conceptualization – Büyükakıncı BY and Karadag R; methodology – Büyükakıncı BY and Karadag R; formal analysis – Büyükakıncı BY and Karadag R; investigation – Büyükakıncı BY and Karadag R; resources – Büyükakıncı BY and Karadag R; writing-original draft preparation – Büyükakıncı BY and Karadag R; writing-review and editing – Büyükakıncı BY and Karadag R; visualization – Büyükakıncı BY and Karadag R; supervision – Büyükakıncı BY and Karadag R. Both authors contributed to the article equally. The authors have read and agreed to the published version of the manuscript.

Conflicts of Interest

The authors declare no conflict of interest.

Funding

This research received no external funding.

This article was published under the competence of Guest Editor Shahid Adeel.

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