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REGULAR ARTICLE

Investigation of Dyeing Behaviors of Bio-mordanted Wool Fabric Dyed with Natural Dye

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Abstract

In this research, green dyeing treatment of wool fabrics was examined with natural dye extracted from *Beta vulgaris* (beetroot) with an ultrasonic-assisted method. Wool fabric samples were treated with ascorbic acid, sodium carbonate, and tannic acid with different concentrations and durations via the ultrasonic-assisted method before dyeing treatment. The usability of ascorbic acid, sodium carbonate, and tannic acid was investigated as a bio-mordant agent in the natural dyeing process. After the pre-treatment with different substrates, the samples were colored with the natural dye obtained from beetroot for 5 min with the ultrasonic-assisted method. The dyeing parameters' effects were investigated on the colorimetric and fastness properties. According to the results, Fourier transform infrared spectra indicate that there are no important dissimilarities in the functional groups of wool fabric samples with the pre-mordanting process. The experimental results show that ascorbic acid, sodium carbonate, and tannic acid are used as bio-mordants. Furthermore, the pre-mordanting process, mordant agent type, mordant concentration, and mordanting time had an effect on the fastness and colorimetric behaviors of the samples. Color strength results demonstrated that ascorbic acid mordant improved the color strength of the samples (*K/S* increased from 3.51 to 4.63), attributing darker shades (lower lightness, *L*) to the wool fabric. The light fastness of samples improved from 1 to 2 with the use of ascorbic acid for 15 min mordanting time. Furthermore, the best results for color change, washing, and rubbing fastness were obtained by using tannic acid as a mordant and increasing the mordanting time. In addition, the following dyeing characteristics of wool fabrics are estimated using an artificial neural network (ANN) model. In accordance with the experimental outcomes, the suggested approach obtains regression values of more than 0.97 for all dyeing characteristics. As can be shown, the suggested approach is accomplished and can be utilized effectively for predicting colorimetric properties of wool fabric. It has been concluded that the ultrasonic-assisted method is an environmentally sustainable dyeing process of textile fibers, and bio-mordants have rendered the dyeing treatment greener and more sustainable.

Keywords Beetroot · Natural dyeing · Artificial neural network · Ultrasonic-assisted method wool

1 Introduction

In recent years, the textile industry has focused on eco-friendly and sustainable processes, so researchers have investigated natural dye usage. Natural and synthetic fiber-based textile substrates are generally dyed with synthetic dyes extensively, resulting in dyeing of textile materials being accepted to be one of the essential pollutions. Furthermore, synthetic dyes are reported as allergic, toxic, harmful, and carcinogenic responses in literature after contact with human skin. Compared with synthetic dyes, it is reported that natural dyes are environmentally friendly, biodegradable, green, and non-toxic. For this reason, natural dyes can be thought of as a significant alternative to synthetic dyes, and the demand for natural dyes in textile manufacturing is

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accruing attention owing to the increase in environmental awareness [1, 2].

Nature involves several different sources and gives researchers opportunities for research in general. Natural fauna presents fascinating and attractive color sources to humanity in a wide portfolio. A variety of animal and plant resources have been described for obtaining color and using it in the textile dyeing process [3], ultraviolet protection [4], food coloration [5], textile finishing [6], insect repellent [55], anti-microbial activity [55], and cosmetics [7]. Natural dyeing of textile materials gains great interest in research fields because of sustainability, biodegradability, and eco-friendly properties. Research about natural dyeing revolves around dyeing technique, mordants, mordanting technique, extraction methods, antimicrobial properties, ultraviolet production, development of fastness properties, and dyeing optimization methods [8–11]. The cotton dyeing mechanism with natural dye extracted from watermelon rind and mango seed kernel was investigated. Each natural dye includes different coloring chromophore groups such as betacyanin, flavonoid, quercetin, and β-carotene [49]. In another study, nylon 6 fabric was dyed with beetroot natural dye with different dyeing parameters, which are dyeing temperature, dyeing time, pH of the dye bath, and dye concentration. The optimum dyeing conditions were determined at the boiling temperature for 60 min under acidic conditions using 100% on the fabric weight [50]. Batool et al. focused on the investigation of the natural colorant of acidic and aqueous extracts of the *Melia azedarach* L. bark for dyeing cotton and silk fabric samples. The results show that aqueous extract gave maximum colorant solubility, and the samples have higher color strength compared with the acidic medium [51]. Another study reported that nylon fabrics underwent dyeing with neem leaf extract under neutral, acidic, and alkaline conditions with lemon juice mordant. Color strength and fastness values of dyed samples increase with lemon juice mordanting in an acidic medium [52]. Toki et al. dyed viscose fabric with red rose flowers with different mordanting ratios using lemon peels and oranges, tannic acid, ferrous sulfate, and copper sulfate. In addition to these studies, some researchers have examined new natural dye sources, which are *Leishmania Infantum* [35], chamomile [36], pomegranate peel [36], apple tree branches bark [36], *Opuntia Ficus-Indica* L. [37], lac [38], Vitis vinifera L. leaves [39], neem leaf [54], watermelon rind, and mango seed kernel bio-wastes [56].

In natural dyeing, mordant agents are used to both improve the fastness behaviors and the color spectrum. Generally, metal salts are preferred in the natural dyeing process, but metallic ions damaging ecosystems are released as wastewater into the environment. Many researchers deemed that natural mordants are not harmful to the environment. Within the scope of this acceptance, many natural resources are investigated as bio-mordants such as sumach, gallnut, tannin, oak galls, myrobalan, pomegranate rinds, gelatin, green tea, chitosan, black tea, kenduka, urine, milk, casein, lemon juice, tartaric acid, banana leaves, rosemary, thuja, and bentonite. Strong biomolecules called “bio-mordants” enhance a plant’s functional qualities when they interact with materials like wool, silk, and cotton. By adding an additional H-bond, these molecules can be treated with the material either after or before dyeing, producing solid and stable tones. Furthermore, by creating a unique binding with the functional regions of the material (–NHCO=silk or wool; –OH=cellulose) and the colorant functional sites (−OH or –OH and –C=O) through the transmission of ions in the system, the conjugation in the molecule contributes to strong, unfading bonds [34].

Ultrasound can be basically categorized as power ultrasound (20–100 kHz) and diagnostic ultrasound (1–10 MHz). When a liquid is subjected to ultrasonic energy, microbubbles occur and grow, oscillating ultrafast. Moreover, high micromixing increases the mass and heat transfer in the liquid phase around the particles, which causes a rise in the diffusion of species within the pores of solid [3]. Ultrasonic energy allows for better penetration of the dyes into textile fibers, resulting in more homogeneous dyeing. Furthermore, it also speeds up the dyeing process, because processing with ultrasonic energy usually gives results in less time. Ultrasonic-assisted methods use less energy compared to conventional dyeing methods. In addition, this method requires fewer chemicals than traditional methods, which, as a result, reduces the environmental impact and ensures a more sustainable production process [3]. Ultrasonic-assisted method is commonly used in natural dyeing applications.

The beetroot, or *Beta vulgaris* L., is a member of the 1400 species *Chenopodiaceae* family and originates in Europe and Asia. Beetroot is recognized as an uncommon flowering perennial plant. Although it is a plant for the cool season, it can withstand high temperatures; its ideal temperature range is between 15 and 19 °C. The plant is harvested in the summer for 75–90 days and in the winter for 100–120 days. A root vegetable, beetroot contains vitamins, minerals, carotenoids, flavonoids, and nitrates [33]. Beetroot was reported as a natural dye source in literature. Al-Amir et al. extracted betalains as a natural color from beetroot wastes to dye silk, cotton, and wool fabrics. Before dyeing, the samples were immersed directly into the chitosan solution with a 1:50 liquor ratio for 1 h at room temperature. The color strength (*K/S*) values have reached from 9.7 to 15.6 for treated silk fiber [58]. Sivakumar et al. studied the extraction of natural dye from beetroot using ultrasound and compared it with static/magnetic stirring as a control process at 45 °C. The coloring ability of extracted beetroot dye has been tested on substrates such as leather and paper and obtained to be proper for dyeing [3]. Another study reported on natural dye extracted from beetroot with a Soxhlet apparatus using

ethanol, water, aqueous methanol, and ethyl acetate. Methanol gave a higher yield of dyestuff than the other solvents. The meta-mordanting and pre-mordanting processes were carried out with alum and ferrous sulfate for the dyeing of cotton fabric samples with beetroot natural dye. Ferrous sulfate as a mordant gave a better % exhaustion on dyed cotton, and the dyeing was best achieved with pre-mordanting techniques [59].

Because dyeing properties for fibers are frequently complicated and reliant on several variables, artificial neural networks (ANNs) can be utilized to forecast them. Many factors influence dyeing results, including dye chemical properties, ambient conditions, fiber structure, and process physical conditions. ANNs can interpret non-linear and multidimensional data by learning the complicated relationships between various characteristics, allowing them to predict dyeing qualities with great accuracy. Haji and Vadood dyed cotton fabric samples mordanted with different combinations of alum using weld and madder natural dyes. Color coordinates of samples were measured, and it was tried to model the relation between the concentration of dyes and each color coordinate using ANN, regression method, support vector machine, and fuzzy logic. Except for regression models, other model methods can predict all color coordinates with high accuracy [60]. Ghaheh et al. predicted optimum dyeing conditions for woolen fabric dyeing with plum-tree leaves. The dyeing parameters were determined as dye concentration, pH level, dyeing time, and temperature. Results demonstrated that ANN ($R^2 \sim 1$) achieves much better than response surface methodology (RSM) ($R^2 > 0.92$) [61]. Aftad et al. assessed the estimation capabilities of RSM and ANN modeling for dyeing cotton fabric with clove. They considered dyeing time, extract pH, dyeing temperature, and salt for exhaustion, with *K/S* as the response variable. It is concluded that the ANN model has better estimation capacity for process optimization as compared to the response surface methodology [62]. Another study reported the predicted color strength of polyester fabric dyed with madder. The experimental design was performed with 46 samples dyed under different levels of five parameters, which are dye concentration, temperature, time, dyebath pH, and liquor ratio. The ANN and fuzzy logic models were utilized to estimate the color strength values. It was found that the best obtained ANN and fuzzy models can estimate the color strength values with mean absolute percentage errors of 2.52 and 3.01, respectively [63]. In addition to these studies, dyeing properties of air plasma-treated wool fabric dyed with *Rubia tinctorum* L. natural dye [64], dyeing characteristics of mohair fiber dyed with *Candelariella reflexa* natural dye [65], and color coordinates of polyester fabric dyed with madder natural dye [66] were predicted with the ANN model.

The principal contribution and aim of this research are to develop the dyeing behaviors of wool fabrics with natural dye achieved from beetroot using bio-mordants. To the best of the authors' knowledge, there is no existing literature investigating the dyeing of pre-mordanted wool fabric with bio-mordants with natural dye extracted from beetroot via the ultrasonic-assisted method. While numerous studies have explored the dyeing behaviors of natural dye extracted from beetroot on various textile materials, none have utilized ascorbic acid, sodium carbonate, and tannic acid as bio-mordants. Additionally, no studies have predicted dyeing characteristics and fastness properties using artificial neural networks (ANNs) based on the effects of mordant type, concentration, and mordanting duration. A greener dyeing process of wool fabrics was carried out with natural dye achieved from *Beta vulgaris* (beetroot) via an ultrasonic-assisted method for 5 min. Before the dyeing, the samples were mordanted with ascorbic acid, sodium carbonate, and tannic acid with different concentrations and durations with the ultrasonic-assisted method. According to the results, the pre-mordanting process, mordant agent type, mordant concentration, and mordanting duration had an impact on the colorimetric and fastness behaviors of the samples. An ANN model is also presented to approximate the dyeing characteristics of wool fabric samples. The suggested ANN-based findings point out that the model can be efficaciously utilized to forecast the dyeing features of wool fabric samples.

2 Experimental

2.1 Materials

100% raw wool fabric was purchased from Yunsa Inc. (Tekirdağ, Türkiye) with the properties as follows: 160 g/m^2 plain knitted (24 weft/cm and 32 warp/cm) wool yarn number of 35 Nm. The mordants tannic acid ($C_{76}H_{52}O_{46}$, molecular weight = 1701.20, CAS No. 1401–55-4), ascorbic acid ($C_6H_8O_6$, molecular weight = 176.12, CAS No. 50–81-7), and sodium carbonate (Na_2CO_3, molecular weight = 105.99, CAS No. 497–19-8) were purchased from Sigma Aldrich. Beetroot was obtained from Bursa, Türkiye, in March 2021.

In this study, beetroot was utilized as a natural dye resource. A variety of beetroot consists of yellow-colored xanthine, betalains-red-colored betanins, and cyanins. Cyanin presents 75–90% of the total color in beetroot. Betalains involve two sub-groups: betacyanins (red-violet pigments) and betaxanthins (yellow-orange pigments) (Fig. 1). Betalains absorb between 476 and 600 nm radiation with similar distribution patterns for both types of betalains, supporting the functional similarity between these classes of natural pigments [30]. In the literature, betalain has antioxidant, antiviral, and antimicrobial activities [3]. Tannic acid, ascorbic acid, and sodium carbonate were also used in this study

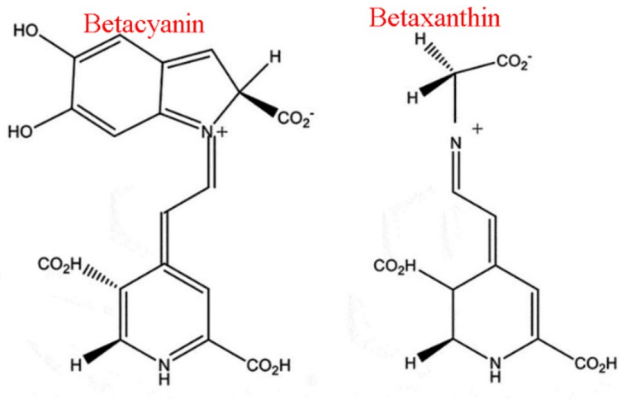


Fig. 1 Chemical structure of betacyanin and betaxanthin

as bio-mordants. The chemical structures of bio-mordants are given in Fig. 2 [12, 13].

2.2 Dye extraction

One kg of dried beetroot plant was blended with 2000 ml of distilled water for dye extraction. The mixture bath was continued at boiling temperature for 60 min and was filtered after cooling.

2.3 Dyeing

Prior to the dyeing process, wool fabric samples were treated with bio-mordants, which are tannic acid, ascorbic acid, and sodium carbonate at a 1:50 liquor ratio with 5%, 7.5%, and 10% concentration with the ultrasonic-assisted method. The pre-mordanted processes were carried out with an ultrasonic bath (ISOLAB) for 5, 10, and 15 min at 80 °C at a 40 Hz frequency. Then, the samples were removed from pre-mordanting liquor with squeezing. Then, the samples were dyed with the ultrasonic-assisted method in the ultrasonic bath (ISOLAB) for 5 min at a material-to-liquor ratio of 1:50 at pH 5.5 at 80 °C at a 40 Hz frequency. Following the dyeing process, the samples underwent a cold rinse overflow.

2.4 Colorimetric Analysis

The colorimetric features of dyed samples were characterized using Datacolor 650™ (Datacolor SPECTRUM™ USA) in regard to CIE*Lab* values (*L*, *a*, *b*) and color strength (*K/S*) [14]. The color strength (*K/S*) of samples was obtained with the Kubelka–Munk equation.

$$\frac{K}{S} = \frac{(1-R)^2}{2R} \tag{1}$$

In this equation, *K* represents the absorption coefficient, *R* represents the reflectance of the dyed samples, and *S* symbolizes the scattering coefficient [15]. The *K/S* values of samples were estimated at a wavelength of 540 nm.

2.5 Fastness Properties Analysis

In this study, light fastness analysis of samples was carried out with the Atlas Alfa 150 S instrument with the EN ISO 105-B02 standard [16]. The washing fastness of samples was analyzed with the ISO 105-C06 standard [17]. The samples were washed using the Gyrowash/James H.Heal Co.Ltd. test instrument by using 5 g/L soap at 40 °C for 30 min. Rubbing fastness of samples was determined with wet and dry standard crocking fabric for 10 cycles using the James H.Heal 255 crock meter in accordance with ISO 105-X12 [18].

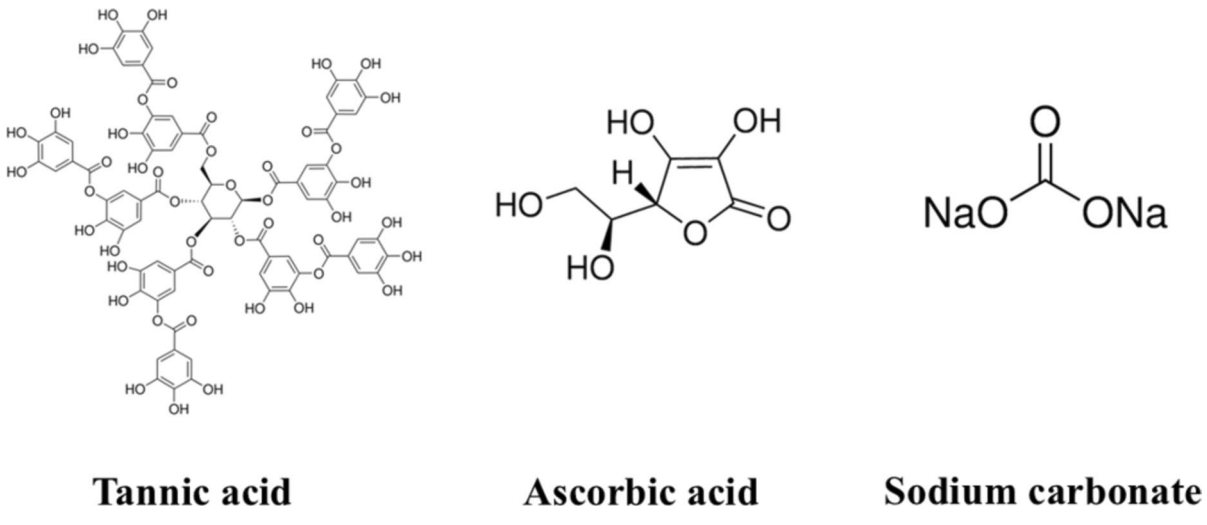


Fig. 2 Chemical structures of bio-mordants

2.6 Artificial Neural Network Modelling

A feed-forward neural network (FFNN) model was adopted in this investigation to approximate the dyeing characteristics of wool fabrics. Figure 3 indicates the design of the suggested FFNN. The network model is divided into three layers: input, hidden, and output. *L*, *a*, *b*, *K/S*, color change, dry rubbing fastness, and wet rubbing fastness are the outputs, whereas mordant type, mordant concentration, and mordanting time are the inputs. The number of neurons in the hidden layer was set at ten, as in many previous research studies [19–23]. While the input layer in FFNNs is utilized to receive data from outside and feed the network, the output layer represents the outputs and measured solubility [20, 21, 24]. The number of neurons in each input and output layer is equal to the number of input and output attributes counted. The hidden layer is utilized to optimize the network. Furthermore, the number of neurons in the hidden layer is chosen based on the problem type [20, 21, 25].

Due to its efficacy, the Levenberg–Marquardt algorithm [26] was utilized to train the suggested model, initially feeding the network with the training data. The network parameters are then adjusted based on the distinctions between the outputs and targets, and the testing dataset is ultimately utilized to assess the trained network. This process is continued until the desired level of training accuracy is reached. Throughout the training phase, the mean squared error (MSE), which is the average squared variance between outputs and aims, is employed until the least error value is obtained. The MSE is computed as shown below:

$$MSE = \frac{1}{n}\sum_{i=1}^{n}\left(Y_i - \hat{Y}_i\right)^2 \quad (2)$$

where Y denotes a vector of observed values and $\hat{Y}$ denotes a vector of n approximated values [20, 21, 23, 27].

3 Results and Discussion

3.1 Fourier Transform Infrared Analysis

Fourier transform infrared analysis (FTIR) was performed to evaluate the effect of the pre-mordanting process on wool fibers, and FTIR spectra of samples are illustrated in Fig. 4.

Wool fabric samples were qualified with FTIR spectroscopy. FTIR spectra of the wool fabric samples show functional group absorption peaks and peptide bonding, known characteristics of wool. According to the previous studies, the elemental analysis of wool was performed, and it was determined that wool involves substantial amino acid in the structure [17]. According to the FTIR results, strong differences are not observed with the pre-mordanting process using ascorbic acid, sodium carbonate, and tannic acid. The region, varying from 3300 to 2800 cm^{-1}, typifies C–H stretching, which is responsible for O–H bending. In

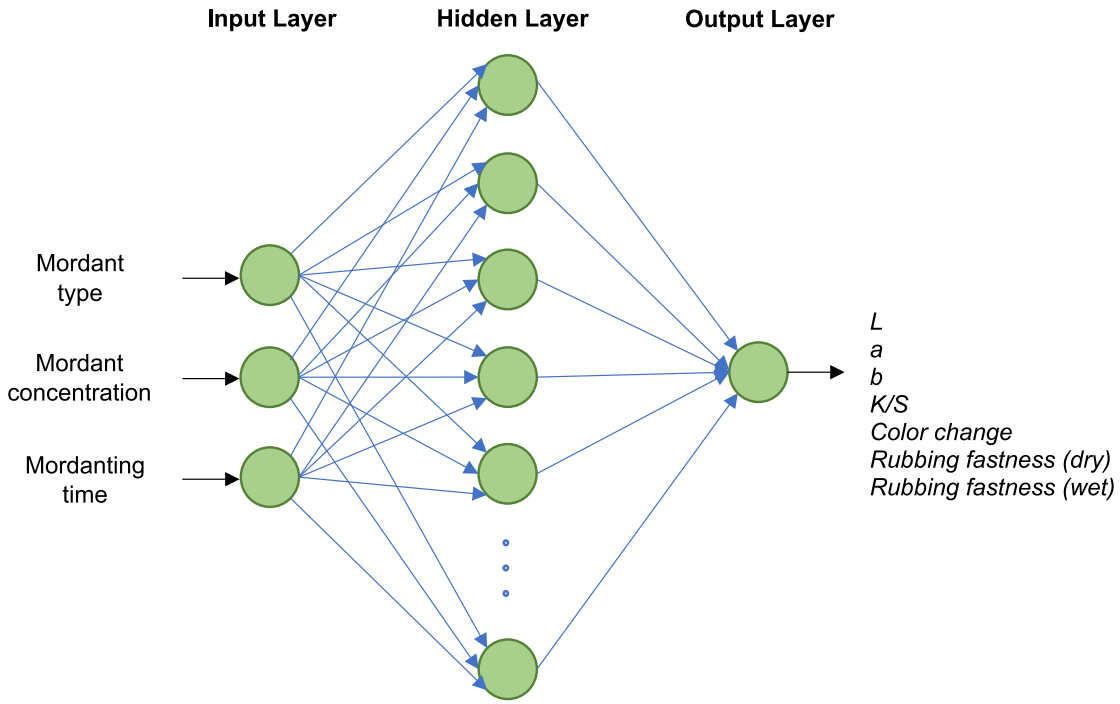


Fig. 3 The suggested feed-forward neural network model

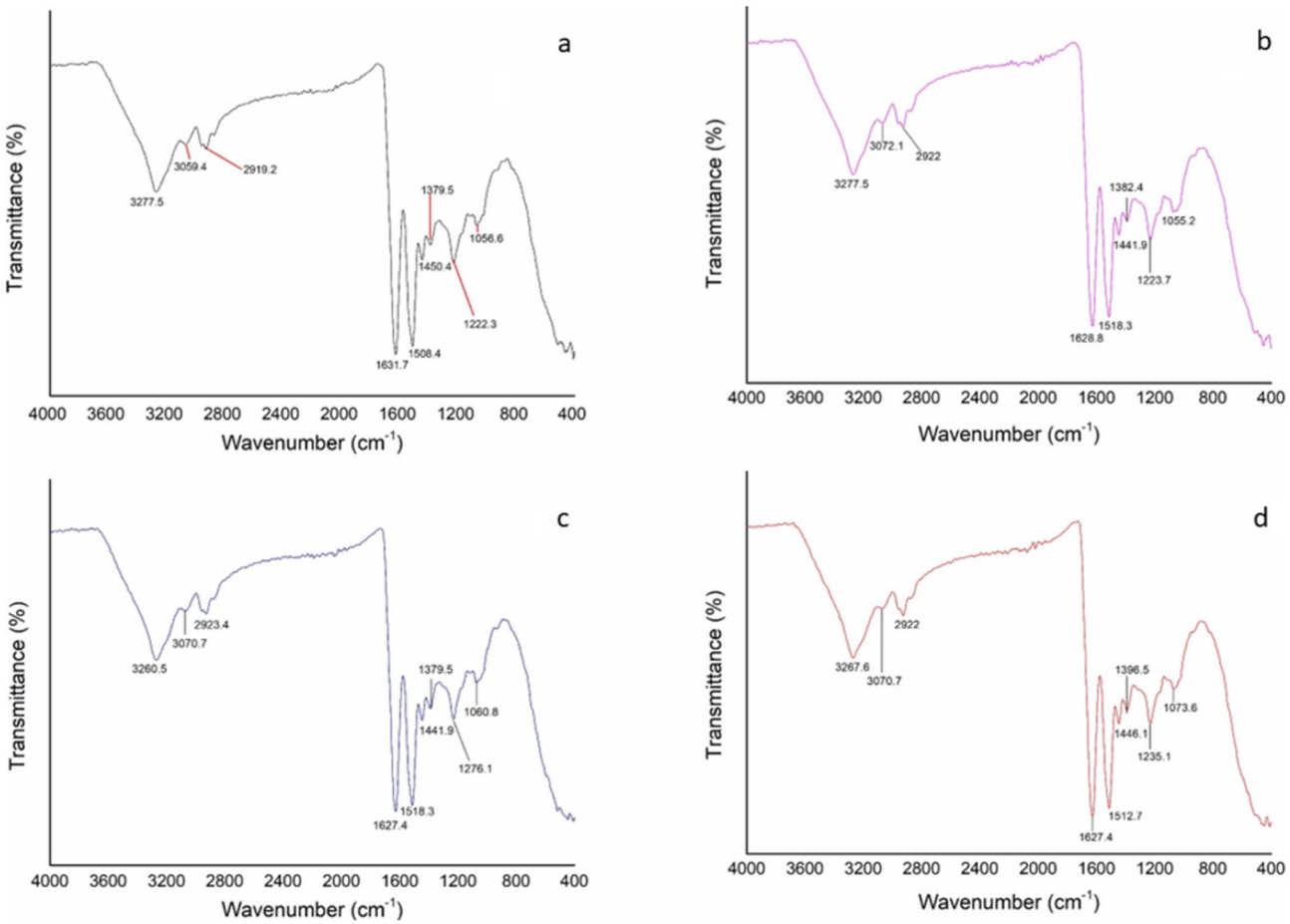


Fig. 4 The FTIR spectra of samples. (**a** Standard sample, **b** Sample mordanting with ascorbic acid, **c** Sample mordanting with sodium carbonate, **d** Sample mordanting with tannic acid)

addition, FTIR spectra illustrate that important differences are responsible for hydroxyl groups. In this region, it was not determined that there were important differences in all the samples. These broad peaks are caused by free and/or intra-/intermolecular hydroxyl bonds, which may be intricately entangled with one another and produced between wool fibers and mordanting agents. In the FTIR spectra, it is seen that significant variations are not after the pre-mordanting process with using ascorbic acid, sodium carbonate, and tannic acid at 1700–1500 did not eventuate in the amide groups (1500–1200 cm^{-1} region) with the pre-mordanting process [28]. The FTIR spectrum of wool indicates various bands, including wool keratin-related IR bands of amide III, amide II, and amide I peaks at 1200–1250, 1350–1400, 1500–1550, and 1630 cm^{-1}, respectively. The peak at 1515 cm^{-1} could be assigned to the C−N stretching and to the N − H in-plane bending vibrations (amide II). The high peak at around 3200 cm^{-1} could be attributed to the hydroxyl groups of some amino acids of wool keratin, such as serine and tyrosine [48]. The FT-IR peak allocations and positions of chemical stretching in wool fabric were tabularized in Table 1.

3.2 Color Properties

Almost all natural dyes are generally necessary chemical substances known as mordants to achieve color fastness on textile materials. Mordants, natural-based or metal salts, provide the bonding of dyes to textile materials by a chemical bridge formed between them. The existence of hydroxyl (–OH) groups in the chemistry of tannic acid, ascorbic acid, and sodium carbonate develops the color features of beetroot-dyed wool fabric. Betalain dyes, known as a natural substitution to synthetic dyes, are obtained via the extraction of beetroot. Most types of beetroot involve betalain-red-dyed betanins and cyanins and yellow-dyed xanthin. Betalains involve two sub-groups: betacyanins (red-violet pigments) and betaxanthins (yellow-orange pigments). These color agents have -OH groups in the structure, and these groups may be responsible for producing different colors on the fibers [3, 29]. Furthermore, betacyanin and betaxanthin have COO^- groups, which can cause ionic interactions between NH_3^+ groups of wool in pH 5–5.5 (Fig. 5) [40]. According to Table 2, it is determined that the color of samples is

Table 1 The FT-IR peak allocations and positions of chemical stretching in wool fabric

Peak positions (Wavenumber (cm^{-1}))	Functional groups	Respective chemical composition	References
3300–2800	C–H and O–H stretching	Serine and tyrosine	[48]
1700–1600	C=O stretching	Amide I	[57]
1600–1500	N–H stretching	Amide II	[57]
1500–1300	C–N stretching	Amide II	[57]
1300–1200	C–O stretching	Carbonyl group, amide I, amide II, amide III	[57]
1100–1000	S–O stretching	Sulfur group	[57]

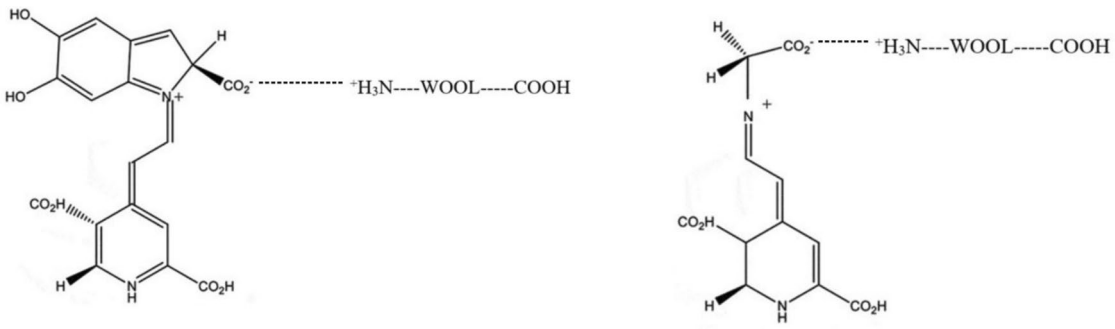


Fig. 5 Interaction between betacyanin and betaxanthin of wool

red-violet tones responsible for the betacyanin component of beetroot.

The mordanting process has a substantial effect on the fastness and color strength values of the dyed fabrics. It is known that the mordanting process develops the interaction between fibers and dye molecules via chemical complex formation. Besides providing dyestuff affinity, mordants generate different colors and increase the dye fastness. In literature, there are many natural mordants that can yield an excellent and gratifying color [28].

In the spectrophotometric measurements, *L*, *a*, and *b* are the parameters of the CIE*Lab* system, which describes the color coordinates. In these parameters, *L*, *a*, and *b* are described as the black-white scale (lightness), the red-green scale, and the yellow-blue scale. CIE*Lab* color characteristics of samples are illustrated in Table 2. According to Table 2, color coordinates and color strength of dyed samples are affected by mordant type, mordant concentration, and mordanting time. Different mordants can cause dyes to have different spectral absorption properties, which can change the color coordinates [67]. The results point out that mordant type, mordant concentration, and mordanting time are the parameters that influenced the lightness of samples. With the use of ascorbic acid, the lightness of samples decreased, while the lightness of samples increased with the use of sodium carbonate and tannic acid as mordant agents. It is thought that the *L* value differs due to the difference in the bond energy of the dyes binding with different mordants. The color of all dyed wool fabric samples appeared in the area of red–purple color. The variations of color coordinates confirmed the apparent color change by varying mordant type, mordant concentration, and mordanting time [40].

Different mordants could fabricate darker or lighter colors. Sodium carbonate and tannic acid mordants had a more darkening effect than ascorbic acid, as could be determined from the *L* values in Table 2. In addition, tannic acid penetrated the wool fabric during the dyeing process and created additional hydrogen bonds with the wool fibers, thus achieving color fixation and dispersing dye molecules. Tannic acid, which is a biomaterial, has an incomparable color. For this reason, tannic acid may deepen the color of wool fabric to increase and perform the dual effect of fixing and dyeing, which has a favorable impact in terms of developing the utilization of natural dyes [47]. Furthermore, *a* (redness) value of samples increased with the use of ascorbic acid and decreased in using sodium carbonate, and tannic acid. *b* (yellowness) values of samples increased in using bio-mordant agents. Ascorbic acid, sodium carbonate, and tannic acid, as well as beetroot natural dye, involved -OH groups with free electron pairs on the oxygen atoms, which can produce complexes of dyes. These complex forms cause the development of the color fastness and dye affinity [29].

Table 2 shows that *K/S* values were reasonably close to one another in every combination. Commonly, when the

Table 2 Color parameters and color strength values of samples

Sample code	Mordant type	Mordant concentration (%)	Mordanting time (mins)	L	a	b	Color	*K/S*
0	-	-	-	49.36	31.85	5.35		3.51
1	A. acid	5	5	48.91	32.01	6.26		3.62
2	A. acid	5	10	46.56	33.03	6.52		4.34
3	A. acid	5	15	45.70	33.40	6.95		4.63
4	A. acid	7.5	5	40.03	32.58	6.07		4.15
5	A. acid	7.5	10	47.60	32.25	5.98		3.98
6	A. acid	7.5	15	41.18	32.86	7.00		4.12
7	A. acid	10	5	47.35	32.61	5.62		4.12
8	A. acid	10	10	47.90	32.61	5.62		3.94
9	A. acid	10	15	47.02	33.23	6.64		4.19
10	S. carbonate	5	5	50.25	29.50	8.96		3.17
11	S. carbonate	5	10	51.98	26.53	10.86		2.68
12	S. carbonate	5	15	54.54	24.52	10.18		2.17
13	S. carbonate	7.5	5	49.71	29.66	8.96		2.17

outcomes were surveyed in terms of color yields (*K/S*), it could be seen that the color of samples varied with the type of mordant agent, mordant concentration, and mordanting time. The results show that *K/S* values of samples do not change in a linear way according to mordant concentration and mordanting time for sodium carbonate and tannic acid mordant agents. The *K/S* results showed that ascorbic acid mordant improved color strength (*K/S*) of the samples, attributing darker shades (lower lightness, *L*) to the wool fabric. The intense interactions between the mordant, dye, and fabric may be the basis for this improvement [29–31]. In addition, ascorbic acid is a weaker acid that can discharge protons from the COOH groups. The dissociation reactions of the wool's acid and amino acids, the wool's activation, and the double change response between the activated wool and betalains reactions are the foundations for the protonation of wool in an acidic media. Wool reacts with acid because it is an amphoteric substance; as a result, it takes protons from the acid and becomes activated or protonated wool [42]. Furthermore, Barani and Rahmani confirmed that pre-mordanting with ascorbic acid (5 wt.%) results in the thermal stability of betacyanin color in pH 5.5. In addition, increasing the ascorbic acid concentration led to an improvement in the color strength of dyed wool samples. Pre-mordanting with

Table 2 (continued)

14	S. carbonate	7.5	10	56.47	22.82	12.70		1.85
15	S. carbonate	7.5	15	54.05	24.44	10.33		2.24
16	S. carbonate	10	5	56.05	23.08	10.99		1.90
17	S. carbonate	10	10	54.20	25.19	10.64		2.25
18	S. carbonate	10	15	56.53	22.36	11.23		1.82
19	T. acid	5	5	61.23	18.57	11.29		1.23
20	T. acid	5	10	63.12	16.62	13.88		1.03
21	T. acid	5	15	64.08	12.70	14.07		0.89
22	T. acid	7.5	5	60.49	20.14	10.85		1.33
23	T. acid	7.5	10	63.11	15.48	13.18		1.01
24	T. acid	7.5	15	64.10	12.09	15.53		0.87
25	T. acid	10	5	61.52	17.99	12.40		1.18
26	T. acid	10	10	63.29	14.69	13.92		0.98
27	T. acid	10	15	64.86	12.37	14.79		0.84

A. acid Ascorbic acid, *S. carbonate* Sodium carbonate, *T. acid* Tannic acid

ascorbic acid caused slight acidification of the dye extraction, which may promote betalain stability [40].

Generally, when Table 2 is evaluated in terms of *K/S* values, it is determined that *K/S* values of samples are low. In literature, other studies that investigated dyeing behaviors of beetroot natural dye on wool fabric demonstrated that the *K/S* values of samples were low. Benli used beetroot natural color for dyeing wool fabric with metal salt mordant via the conventional dyeing method for 60 min at 100 °C with different liquor ratios. The results show that *K/S* values of samples vary from 5.75 to 12.42 at a 1:25 liquor ratio. According to the literature, it was deemed that liquor ratio and mordant type are the most effective parameters on color strength values [34]. With the increase in the liquor ratio, the number of dyestuff pigments in the liquor increases. In another study, *K/S* values of wool fabric samples differ from 2.16 to 5.71 [41]. In literature, tannins have commonly been used as mordant agents for many types of natural dyes on wool. Because of tannin polyphenolic compounds, tannic acid mordanting causes variation in the colorimetric coordinates of dyeing samples [44–46]. In addition, it is reported that tannic acid pre-mordanting showed color attributes considering *K/S* values in literature. As a result, the dyed wool fabric indicated a bright color using the natural dyes generated shades. This phenomenon can be based upon

the tendency of natural mordants to set up strong chemical interactions with dye obtained from beetroot. These interactions may include chemical complexes and coordination bonds [55]. Furthermore, bio-mordants are strong biomolecules that develop the functional behaviors of the plant when interacting with fibers such as wool, silk, and cotton. When these molecules are treated with the fibers after or before dyeing, they manufacture stable and solid tones via the extra H-bond. Moreover, the conjugation in the molecule plays a role in strong, unfading bonds by causing a special bond with the functional region of the material (-NHCO=silk or wool; –OH=cellulose) and the colorant functional points (–OH or –OH and –C=O) by the transfer of ions in the system [30].

3.3 Fastness Properties

The fastness properties of samples in terms of washing, rubbing, and light are indicated in Table 3. Washing and rubbing fastness results were examined with a gray scale varying between 1 and 5 (1 presents bad and 5 represents excellent results). In addition, light fastness values are measured with a blue scale. On the blue scale, "1" gives a bad result and "8" gives an excellent result.

The light fastness of samples was found to be bad, as shown in Table 3. Low light fastness is the most significant disadvantage of natural dyes. The physical and chemical structure of the dye, dye concentration, kind of fiber, mordant agents, and fiber nature all have an impact on the light fastness of dyeing. According to the analysis of natural dyes, all flavonoid compounds used to color textiles are natural dyes and have a poor light fastness value [32].

Table 3 Fatness properties of samples

Mordant type	Mordant concentration	Mordanting time	Light fastness	Washing fastness							Rubbing fastness	
				Color change	CA	CO	PA	PET	PAN	WO	Dry	Wet
Raw	–	–	1	4	4	4	3/4	4	4	3/4	4	3/4
A. acid	5	5	1/2	3/4	4/5	4/5	4/5	4/5	4/5	4/5	5	4/5
A. acid	7.5	5	1/2	3/4	4/5	4/5	4/5	4/5	4/5	4/5	4	3/4
A. acid	10	5	1/2	3	4/5	4/5	4/5	4/5	4/5	4/5	4	3/4
A. acid	5	10	1/2	2/3	4/5	4/5	4/5	4/5	4/5	4/5	4	3/4
A. acid	7.5	10	1/2	2/3	4/5	4/5	4/5	4/5	4/5	4/5	4	3/4
A. acid	10	10	1/2	2/3	4/5	4/5	4/5	4/5	4/5	4/5	4	3/4
A. acid	5	15	2	2/3	5	5	5	5	5	5	4	3/4
A. acid	7.5	15	2	2/3	5	5	5	5	5	5	4	3/4
A. acid	10	15	2	2/3	5	5	5	5	5	5	4	3/4
S. carbonate	5	5	1	3	4/5	4/5	4/5	4/5	4/5	4/5	4/5	4/5
S. carbonate	7.5	5	1	3	4/5	4/5	4/5	4/5	4/5	4/5	4/5	3/4
S. carbonate	10	5	1	3	4/5	4/5	4/5	4/5	4/5	4/5	4/5	3/4
S. carbonate	5	10	1	2/3	4/5	4/5	4/5	4/5	4/5	4/5	4	4
S. carbonate	7.5	10	1	2/3	4/5	4/5	4/5	4/5	4/5	4	4	4
S. carbonate	10	10	1	2/3	4/5	4/5	4/5	4/5	4/5	4	4	4
S. carbonate	5	15	1	2	4/5	4/5	4	4/5	4/5	4	4	4
S. carbonate	7.5	15	1	2	4/5	4/5	4	4/5	4/5	4	4	4
S. carbonate	10	15	1	2	4/5	4/5	4	4/5	4/5	4	4	4
T. acid	5	5	1	4	2	4/5	4/5	4/5	4/5	4/5	4/5	4/5
T. acid	7.5	5	1	3–4	2	4/5	4/5	4/5	4/5	4/5	4/5	4/5
T. acid	10	5	1	4	2	4/5	4/5	4/5	4/5	4/5	4/5	4/5
T. acid	5	10	1	4	2	4/5	4/5	4/5	4/5	4/5	4/5	4/5
T. acid	7.5	10	1	4	2	4/5	4/5	4/5	4/5	4/5	4/5	4/5
T. acid	10	10	1	4	1/2	4/5	4/5	4/5	4/5	4/5	4/5	4/5
T. acid	5	15	1	4	1/2	5	5	5	5	5	5	5
T. acid	7.5	15	1	4	1/2	5	5	5	5	5	5	5
T. acid	10	15	1	4	1/2	5	5	5	5	5	5	5

A. acid Ascorbic acid, *S. carbonate* Sodium carbonate, *T. acid* Tannic acid

Table 3 shows that the bio-mordanting process generally develops fastness properties (light, washing, and rubbing) of samples. The mordanting mechanism is essentially based on the hypothesis that the creation of fiber-mordant-dye complexes is performed through coordination, ionic, and hydrogen bonds between amino, hydroxyl, and carboxyl groups in the fiber, dye, and mordant substance, which develops color fastness of dyeing [47]. After the pre-bio-mordanting process, it is seen from Table 3 that the light fastness of samples mordanted with ascorbic acid develops compared with the standard sample. Ascorbic acid is known as one of the strongest antioxidants. The light waves play a significant role in the weakening of fibers, discoloration of dyes, and photodegradation of substances. In many instances, antioxidant molecules like ascorbic acid can prevent harmful impacts of light [14]. Furthermore, it was reported that the fastness properties of a mordant dye depend on the mordant agent type and mordanting method so that different tannin-dye complexes occur, which may differ in their stability to rubbing, washing, or light and also because tannins can have a positive or negative catalytic effect on the photochemical degradation of natural dye.

Tannins were utilized as mordant agents by Perkin to increase fastness properties. Without a mordanting process, many natural dyes have poor fastness properties. Ali et al. investigated the effect of tannic acid mordant on the dyeability of wool fabric with natural dye. After mordanting with tannic acid, fastness properties of wool fabric samples increase, which in turn is attributed to the development of the interaction of dye and fiber [43]. Tannic acid was used as an after-treatment to develop the fastness properties of protein and polyamide fibers owing to polyphenolic compounds [44–46].

Bio-mordant agents have active –OH groups. It is thought that these groups cause strong interactions between dye molecules and fibers in the mordanting process. In addition, the fastness to washing and rubbing indicates satisfactory results owing to using ultrasonic energy during the dyeing and mordanting process that enables more interactions that ensure improved contact between the wool fabric and the dye molecules [53].

3.4 Neural Network-Based Results

The suggested FFNN model was developed in MATLAB R2020b on a PC equipped with an Intel Core i7-8565U (1.80 GHz) CPU, Windows 10 64-bit, and 8 GB of RAM. The input dataset in this model is described by a 28×3 numeric matrix, while the output dataset is described by a 28×1 numeric matrix for each dyeing characteristic output. The three subsets formed by randomly partitioning the input dataset into these three categories are training (70%, 20 samples), validation (15%, 4 samples), and testing (15%, 4 samples). During the training stage, the neural network is trained by training samples, and the network is adjusted depending on its error. The network generalization is assessed using samples from the validation subset, and the training process is halted when generalization inhibits further development. At last, the testing subset samples acquire a different measure of the performance of the network both during and after training.

Table 4 shows the proposed model's regression results for training, validation, testing, and all subsets. Regression values are used to specify the correlation between outputs and goals. A regression value of 1 points out a close relation, whereas a regression value of 0 indicates a random relation. As shown in Table 4, the best regression values for the whole dataset range from 0.97297 to 0.99999 and are larger than 0.97 for all dyeing characteristics. The regression values of 0.97485, 0.97473, 0.98329, 0.97297, 0.97705, 0.99999, and 0.99994 for *L*, *a*, *b*, *K/S*, color change, dry rubbing fastness, and wet rubbing fastness were achieved, respectively, by choosing ten neurons in the hidden layer. The experimental and estimated values for dyeing characteristics are shown in Figs. 6, 7, 8, 9, 10, 11, 12. These figures indicate that in the vast majority of tests, the experimental and estimated outcomes are very close or equal. This study's FFNN-based findings all show that the suggested FFNN model can be used to properly predict the dyeing features of samples.

4 Conclusions

In this investigation, wool fabric was colored with ecological dye extracted from *Beta vulgaris* (beetroot) via an ultrasonic-assisted method. Prior to the dyeing, the samples were subjected to bio-mordanting treatment with different bio-mordant agents, which are ascorbic acid, sodium carbonate, and tannic acid, with different concentrations and durations with the ultrasonic-assisted method. After the dyeing treatment, the fastness and colorimetric features of the samples were investigated. According to the FTIR spectra results, there are no important variations in the functional groups of

Table 4 Regression results of the suggested model

Dyeing characteristics	Regression results			
	Training	Validation	Testing	All
L	0.97181	0.9514	0.99363	0.97485
a	0.95983	0.99539	0.99332	0.97473
b	0.99228	0.96593	0.98194	0.98329
K/S	0.96732	0.99459	0.98806	0.97297
Color change	0.97318	0.99463	0.97428	0.97705
Dry rubbing fastness	1	1	0.99998	0.99999
Wet rubbing fastness	1	0.99992	0.99967	0.99994

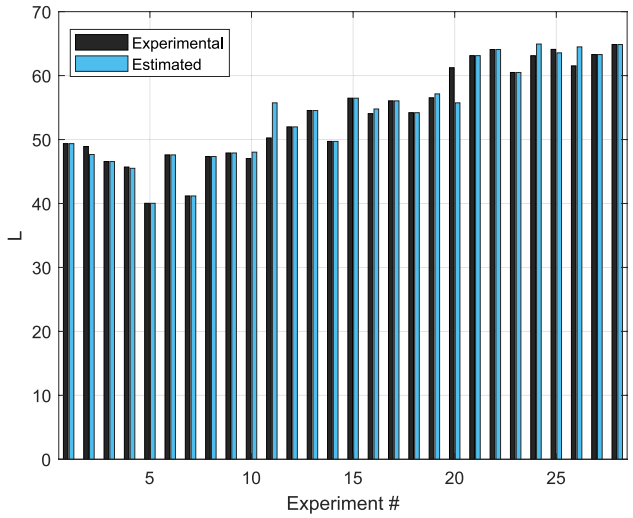


Fig. 6 Estimated and experimental *L* value comparison

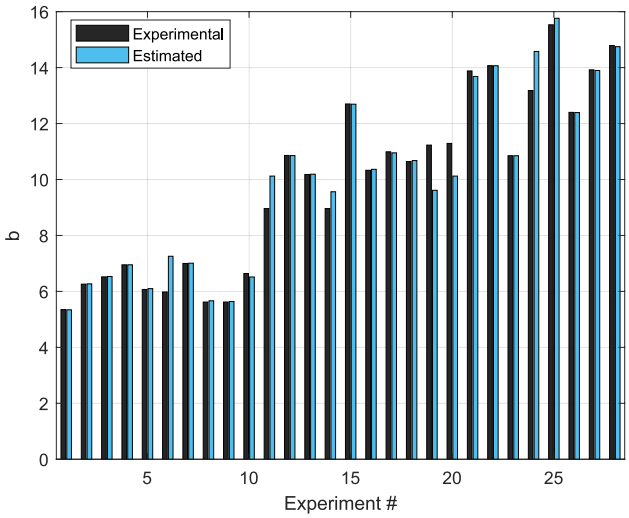


Fig. 8 Estimated and experimental *b* value comparison

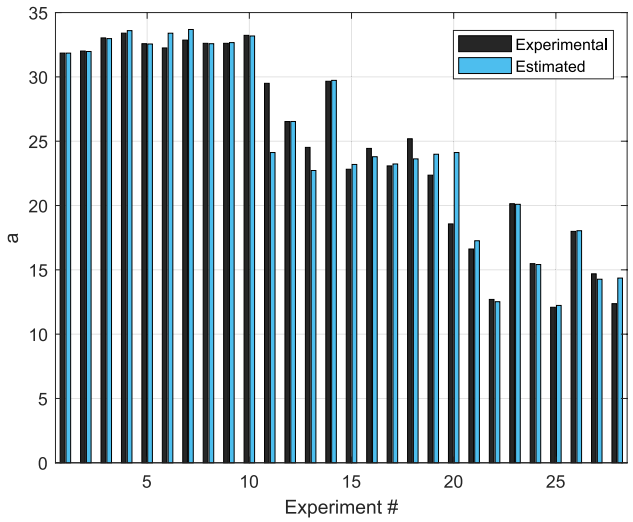


Fig. 7 Estimated and experimental and *a* value comparison

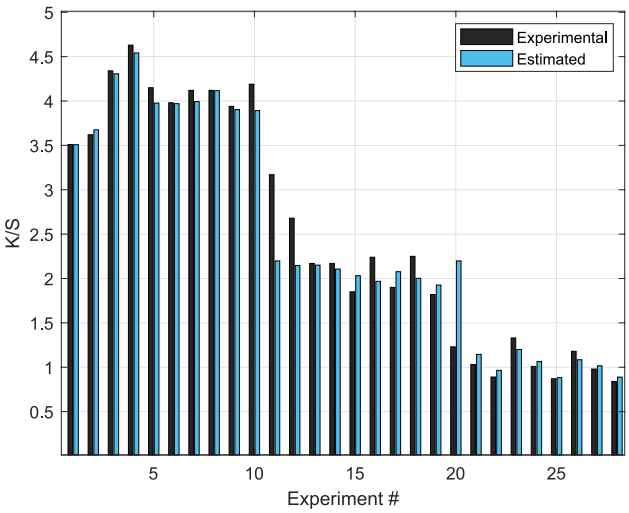


Fig. 9 Estimated and experimental *K/S* value comparison

wool fabric samples with the pre-mordanting process. The experimental results show that the pre-mordanting process, mordant agent type, mordant concentration, and mordanting time had an effect on the colorimetric and fastness characteristics of the samples. Color strength results demonstrated that ascorbic acid mordant improved the color strength of the samples (*K/S* increased from 3.51 to 4.63). The light fastness of samples improved from 1 to 2 with the use of ascorbic acid for 15 min mordanting time. An FFNN model was also presented to forecast and evaluate the dyeing characteristics of wool fabrics. This model has three layers: input, hidden, and output. The dyeing characteristics are the outputs of this network, whereas the inputs are mordant type, mordant concentration, and mordanting time. The suggested model attained regression values of 0.97485, 0.97473, 0.98329, 0.97297, 0.97705, 0.99999, and 0.99994 for *L*, *a*, *b*, *K/S*, color change, rubbing fastness (dry), and rubbing fastness (wet) characteristics, respectively. The experimental results of the current study indicate that the suggested model can be utilized to predict the dyeing characteristics of wool fabrics. It is hinted at on the study basis that an extract of beetroot that is eco-friendly can be utilized to dye wool fabric using ascorbic acid, sodium carbonate, and tannic acid as a mordant.

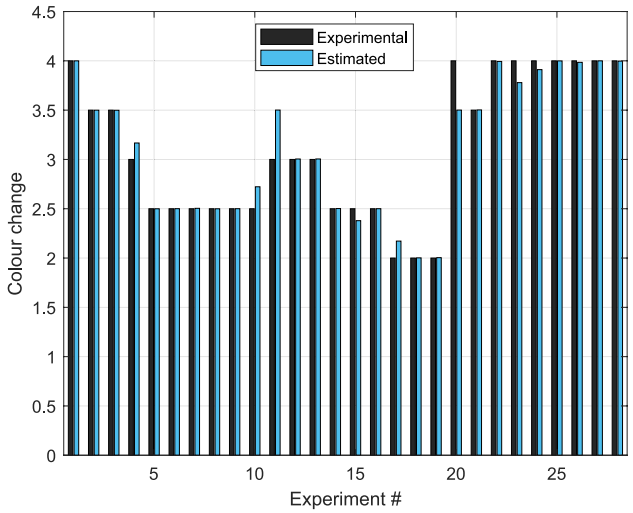


Fig. 10 Estimated and experimental color change value comparison

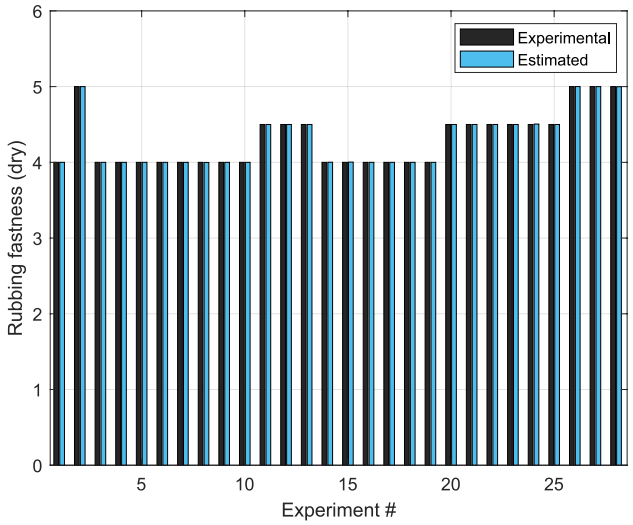


Fig. 11 Estimated and experimental dry rubbing fastness value comparison

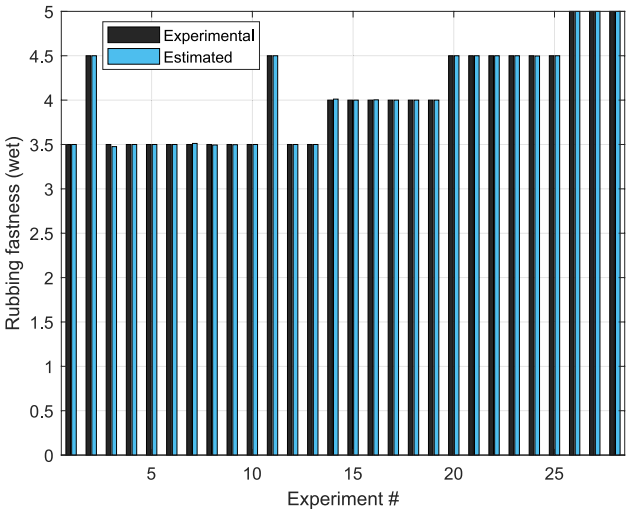


Fig. 12 Estimated and experimental wet rubbing fastness value comparison

Declarations

Conflict of interest None.

Ethical Approval The authors confirm that there were no ethical issues in preparing this manuscript.

Consent to Participate All authors consent to participate in this work.

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