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Investigation of Dyeing Characteristics of Merino Wool Fiber Dyed with Sustainable Natural Dye Extracted from *Aesculus hippocastanum*

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Abstract: Recently there has been growing interest in dyeing biomaterials using natural sustainable plant extracts classified as eco-friendly. The microwave-assisted method provides fast heating and energy efficiency, more homogenous heat distribution in dyeing baths, less use of chemicals, and less heat loss, resulting in this method being greener—more sustainable and ecological. Artificial neural networks (ANNs) are used to predict the dyeing properties of fibers, which are often complex and dependent on multiple variables. This saves time and reduces costs compared to trial-and-error methods. This study presents the green dyeing of merino wool fiber with natural dye extracted from *Aesculus hippocastanum* (horse chestnut) shells using the microwave-assisted method. Before dyeing, the merino wool fiber underwent a pre-mordanted process with aluminum potassium sulfate with different concentrations using the microwave-assisted method. Spectrophotometric analysis of the light, washing, and rubbing fastness of the dyed merino wool fibers was performed. The color strength, light, washing, and rubbing fastness of the dyed merino wool fiber were developed using the pre-mordanting process. After the pre-mordanting process, the light fastness of the samples improved from 1–2 to 3, the color change increased from 2 to 3–4, and the rubbing fastness developed from 2–3 to 4 according to mordant concentration, mordanting time, and dyeing time quantities. The spectrophotometric analysis results indicate that color coordinates vary based on mordant concentration, mordanting, and dyeing duration. Furthermore, the results proved that microwave energy significantly shortened the mordanting and dyeing duration, resulting in an eco-friendly dyeing process. In this investigation, a feed-forward neural network (FFNN) model with sigmoid hidden neurons and a linear output neuron was used to predict the color strength dyeing property of merino wool fiber. Experimental results showed that the proposed model achieved a regression value of 0.9 for the color strength dyeing property. As demonstrated, the proposed FFNN model is effective and can be utilized to forecast the color strength dyeing properties of merino wool fiber.

Keywords: natural dye; *Aesculus hippocastanum*; merino wool; spectrophotometric analysis; fastness; feed-forward neural network

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1. Introduction

Globally, many different types of man-made dyes are utilized for coloring various textiles. It is well known that some man-made dyes that involve azo groups are carcinogenic and cause environmental pollution. To solve these problems with synthetic dyes, many investigators have studied the development of natural dyes, which are safer for dyers and users and are more environmentally friendly [1–3]. The main aim of researchers is to identify novel sources of natural dyes, optimize the dyeing and extraction process, and improve fastness properties. Many studies have been carried out to obtain new natural dyes from different plant materials [4–7]. In order to optimize the dyeing process, different technologies have been evaluated, such as gamma radiation, ultrasonic, and microwave energy [8–10]. Some substrates have been investigated for their ability to improve fastness properties [11].

Natural dyes are commonly defined as color molecules that are extracted from insect or plant sources without the use of chemical production processes. In the present day, interest in natural dyes is rising because environmental pollution threatens all living things. It is estimated that the demand for natural dyes will grow rapidly all around the world in the near future. Natural dyes are gradually taking a share of the global market, and natural dyed eco-textile products are accepted as beneficial in protecting the environment from dangerous man-made dyes [12]. However, the low cost and high fastness properties of synthetic dyes make them attractive in comparison. Natural dyes have nevertheless continued to capture the hearts of consumers aware of maintaining a healthful lifestyle because of their non-carcinogenic, bio-degradable, eco-friendly, sustainable, and non-toxic characteristics [13,14].

Since prehistoric times, merino wool fiber has been utilized as a source of textile fabric, insulation material, and artistic expression. Merino wool is known as the softest and best wool in the world. It is obtained from merino sheep through sustainable agricultural practices. These wool fibers are industrially produced using non-renewable fossil energy. Merino sheep are sustained with natural elements such as sunlight, fresh air, water, and grass over many years. Therefore, the wool of merino sheep is also extremely high quality. These natural fiber wool coats are renewed by sheep every year. As a result, merino wool fiber is a renewable and sustainable fiber. Merino wool fibers are a type of keratin fiber made up of an outer fatty acid monolayer and an inner layer of cortical cells encircled by a cuticular layer. The fiber is largely hydrophobic due to the fatty acid layer, which inhibits dyeing and polymer adhesion. Alkalis, amines, and chloride treatment are used to enhance the wettability of the fiber [15]. The literature on the natural dyeing of merino wool fibers is limited. Gong et al. investigated the dyeability behaviors of merino wool fibers using natural dye obtained from middle-aged/mature *Cinnamomum camphora* leaves with different bio-mordants (pomegranate peel, gallnut, chlorophyll extract, arjun bark, and citric acid) and metal mordants (sodium dichromate, stannous chloride, ferrous sulfate, and copper sulfate). In general, pre-biomordanted merino wool fibers had comparable color and fastness results compared to those of metal salt-treated samples [12]. Another study used biocolorants extracted from waste/fallen *Cinnamomum camphora* leaves and dyed merino wool fibers with metal salt combination pre-mordanting. The results demonstrated the manufacture of 19 shades of different hues and tones according to mordant agent type [16]. Thakker and Sun dyed merino wool fibers with mango ginger, alkanet, chilly, henna, birch, cloves, munro, goldenrod, walnut, and poplar. The maximum K/S values were determined for henna, cloves, and mango ginger [17]. In another study, merino wool yarns were dyed with natural dyes extracted from green tea, tulsi, and manjista. The maximum K/S value was determined to be 8.87 for green tea natural dye [18].

Due to increasing ecological concerns, significant attention has been paid to the textile finishing process since it utilizes less energy, fewer chemicals, and less water. The utilization of microwave energy is accepted as an eco-friendly and green technique in textile finishing operations. Microwaves are defined as electromagnetic waves that include magnetic and electric fields with 1–1000 mm wavelengths. Microwave energy is converted into heat energy when it interacts with metals. This energy has been applied in various processes since the 1950s, including drying and synthesis treatments, waste sterilizations, sintering, dyeing, modification processes, and advanced material processing applications. In textile finishing operations, microwave energy is generally used for its time- and energy-saving advantages, effectiveness in heating, and eco-friendly properties. This method heats the dye bath with microwave energy, resulting in a more even distribution of heat than traditional heating methods. Unlike conventional heating processes, where heat moves inward, microwave heating in textile finishing treatments begins from the center of the dye bath. This leads to the liquid increasing in temperature faster than when using traditional methods. As a result, microwave energy reduces energy consumption and saves time relative to conventional heating methods [19–21].

Aesculus hippocastanum, commonly known as horse chestnut, belongs to the *Hippocastanaceae* family. It originates from Greece and the Balkans and is widely cultivated in North America, Europe, and Asia. Due to its active ingredients, the seeds of *Aesculus hippocastanum* have been widely used in treatments for conditions such as varicose veins, hemorrhoids, phlebitis, fever, rheumatism, neuralgia, and rectal complaints [21]. The horse chestnut plant is cultivated throughout Türkiye, where it is abundant. The fruit shells of horse chestnuts are treated as waste, providing a renewable and sustainable resource. In the literature, a natural dye extracted from horse chestnut has been used to dye organic cotton. The samples were pre-mordanted with copper sulfate, potassium aluminum sulfate, potassium tartrate, and citric acid. The color strength of the sample was found to be approximately 0.5, with light fastness rated at 3–4 and washing, rubbing, and perspiration fastness rated at 5 with aluminum potassium sulfate as the pre-mordant [22]. In another study, horse chestnut was considered a biomordant agent due to its tannin content [23]. Horse chestnut was also used as a natural colorant in additional research, where cotton and mercerized cotton fabric samples were dyed with horse chestnut natural dye. The results indicate that the dyes have optimal fastness and color yield. Furthermore, horse chestnut natural dye gives the samples UV-blocking properties [24].

Artificial neural networks (ANNs) can be used to predict the dyeing properties of fibers as they are often complex and dependent on multiple variables. Many parameters, including the structure of the fibers, chemical properties of the dye, environmental conditions, and the physical conditions of the process, affect dyeing outcomes. By learning the complex relationships between these parameters, ANNs can analyze non-linear and multi-dimensional data, enabling highly accurate predictions of dyeing properties They also save time and reduce costs compared to trial-and-error methods. Since the interactions between many variables in the dyeing process involve non-linear structures, ANNs can model these relationships more accurately, enhancing generalization capacity by minimizing the risk of overfitting. Additionally, they offer a more flexible and adaptable structure that can adjust to continuous data flow and process changes.

The main objective of this study was to demonstrate that the horse chestnut shell extract can serve as a natural, sustainable textile dye and an alternative to synthetic dyes, thereby helping to reduce environmental pollution. Furthermore, this investigation aims not only to evaluate the coloring potential of horse chestnut on merino wool fiber under the influence of green microwave radiation but also to enhance color fastness and introduce new shades by using different concentration mordants. To the best of the authors' knowledge, no study in the literature has investigated the dyeing of pre-mordanted merino wool fiber with aluminum potassium sulfate using natural dye extracted from *Aesculus hippocastanum* shell via the microwave-assisted method with varying mordant concentration, mordanting, and dyeing durations. In this study, merino wool fiber was colored with natural dye obtained from horse chestnut shells using the microwave-assisted method for different durations. Prior to dyeing, merino wool fibers were mordanted with aluminum potassium sulfate at various concentrations and for different durations. Following spectrophotometric analysis, the dyeing process, as well as the rubbing, washing, and light fastness of specimens, were evaluated. ANNs were used to make the dyeing process more cost-, energy-, and time-efficient. Additionally, a feed-forward neural network (FFNN) model for estimating the K/S dyeing property of merino wool fiber was introduced. The proposed FFNN-based model's results indicate that it may be useful for predicting the K/S dyeing characteristics of merino wool fiber.

2. Materials and Methods

2.1. Materials

Merino wool fiber was sourced from merino sheep in the Central Anatolia region (Türkiye), and *Aesculus hippocastanum* was collected in the Black Sea region (Türkiye). Potassium aluminum sulfate was purchased from Sigma Aldrich (St. Louis, MO, USA).

2.2. Method

Before the mordanting and dyeing treatments, merino wool fiber was washed with 1 g/L anionic detergent (Denraw NBP, Denge Chemical Incorporated Company, Istanbul, Türkiye) at a 1:40 liquor ratio at 70 °C for 30 min. First, merino wool fiber was mordanted with varying concentrations of potassium aluminum sulfate using a microwave-assisted method for 1, 2, 3, 4, and 5 min. After the pre-mordanting treatment, merino wool fibers were colored with *Aesculus hippocastanum* natural dye using a microwave-assisted method for 2, 4, 6, 8, and 10 min at 70 °C at 90 watts. Furthermore, a sample was dyed without mordant at boiling temperature for 1 h, which served as a reference sample in the colorimetric and fastness analysis. After the dyeing operation, the spectrometric and fastness behaviors of specimens were analyzed.

2.2.1. Dye Extraction

To obtain the natural dye, 100 g of dried *Aesculus hippocastanum* shell was mixed with 1000 mL of distilled water. The extraction dye bath was maintained at boiling temperature for 1 h and then filtered.

2.2.2. Pre-Mordanting Process

Prior to the dyeing process, the merino wool fiber was mordanted using 0.5 g/L, 0.75 g/L, 1 g/L, 1.25 g/L, 1.50 g/L, 1.75 g/L, and 2 g/L potassium aluminum sulfate (based on fiber weight) for 1, 2, 3, 4, and 5 min in a microwave oven (Whirlpool-jet Stream, Whirlpool Company, Benton, MI, USA) at 90 watts and with a 1:40 liquor ratio and then the fiber was eliminated from the pre-mordanting liquor without washing. Pre-mordanting ensures that the dye spreads evenly and adheres homogeneously to the fibers during the dyeing process, which is why the pre-mordanting method was selected for this study. Aluminum potassium sulfate is a mordant widely used in natural dyeing that offers numerous benefits. It increases color permanence by helping natural dyes bind more effectively to fibers. This mordant makes it possible to obtain various color tones when used with different natural dyes. Fibers, yarns, or fabrics mordanted with aluminum potassium sulfate become more resistant to factors like washing and sunlight exposure. The chemical structure of merino wool fiber mordanted with aluminum potassium sulfate is shown in Figure 1 [25].

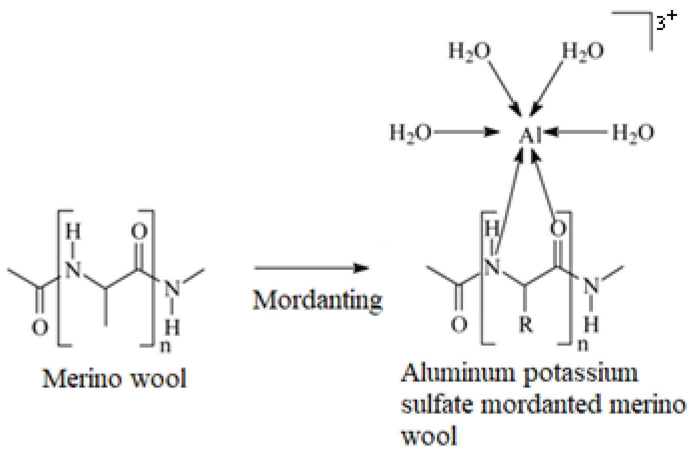

Figure 1. Chemical structure of merino wool fiber mordanted with aluminum potassium sulfate.

2.2.3. Dyeing Process

Merino wool fiber was colored with the natural dye obtained from *Aesculus hippocastanum* shell using a microwave oven (Whirlpool-jet Stream) at 90 watts and a 1:50 liquor ratio for 2,4,6, 8, and 10 min. After the dyeing process, the specimens were left in the dye bath for 12 h. They were treated with a cold rinse overflow, followed by soaping with 1 g/L anionic detergent (Denraw NBP, Denge Chemical Incorporated Company, Tekirdağ, Türkiye) at a 1:50 liquor ratio at 70 °C for 10 min, followed by a final cold rinse. The study's experimental process is illustrated in Figure 2.

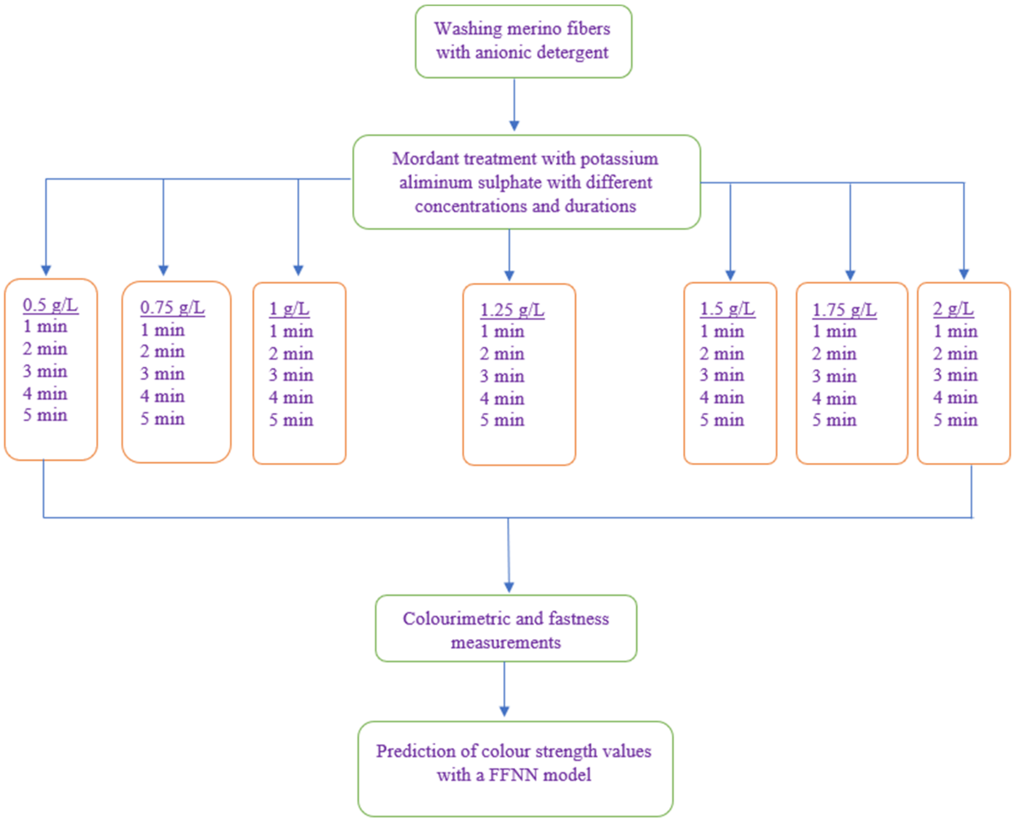

Figure 2. The experimental process.

2.2.4. Colorimetric Analysis

Colorimetric analysis of the samples was carried out using a Gretaq Macbeth—Color Eye 2180 UV spectrophotometer (Datacolor Campany, Lawrenceville, NJ, USA), and the CIELab values were calculated using a D65 illuminant at a 10° standard observer, assessing the L, a, b, C, ho, and K/S values of the dyed merino wool fiber samples. To calculate the color strength (K/S) values of the specimens, the Kubelka–Munk equation was applied as follows:

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

where K is the absorption coefficient, S is the scattering coefficient, and R represents the reflectance of the dyed merino wool fiber, respectively.

2.2.5. Fastness Properties Analysis

The Atlas Alfa 150 S test instrument was utilized to determine the light fastness of the colored merino wool fiber in compliance with EN ISO 105–B02 [26]. Washing fastness was assessed following the ISO 105-C06 standard [27]. The samples were washed using a Gyrowash/James H. Heil Co. Ltd. (Sterling, VA, USA) test apparatus with 5 g/L soap at 40 °C for 30 min. Rubbing fastness was analyzed using the James H. Heal 255 crockmeter, performing ten cycles with both dry and wet standard crocking fabric in accordance with ISO 105 X12 [28].

2.2.6. Feed-Forward Neural Network Modeling

In this study, an FFNN model with sigmoid hidden neurons and a linear output neuron was utilized to forecast the K/S dyeing property of merino wool fibers. Figure 3 depicts the proposed FFNN model, which comprises input, hidden, and output layers. In the model, the dyeing time, mordant concentration, and mordanting time are the inputs, and the K/S dyeing property is the output. The number of neurons in the hidden layer was set to 10, as in various previous studies [29–32]. The network is optimized through the hidden layer, and the number of neurons in this layer depends on the nature of the problem [31–34]. In other words, for different estimation problems, the number of neurons varies according to the input variables and output variables in the dataset. In FFNNs, the output layer displays the network's outputs and measured values, while the input layer takes data from the outside and feeds it into the network. Each input and output layer has the same number of neurons as there are input and output characteristics, respectively [31,32,35].

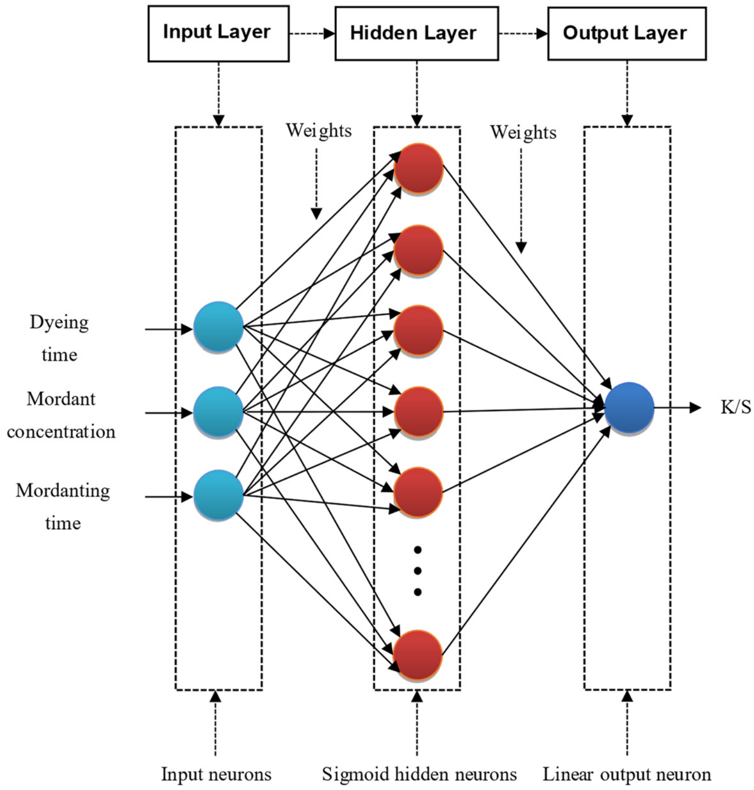

Figure 3. The proposed FNNN model.

The Levenberg–Marquardt algorithm [35] was employed to train the proposed model, chosen because of its effectiveness. First, the FFNN model was fed with the training data. The network parameters were then updated based on the discrepancies between the outputs and the targets, and the testing dataset was utilized to evaluate the trained network. This cycle was repeated until the desired level of training accuracy was achieved. The mean squared error (MSE), which represents the average squared variance between outputs and targets, was utilized throughout the training process until the minimum error value was achieved. The MSE is computed as follows:

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

where Y is an observed value vector, $\hat{Y}$ is a predicted value vector, and n is the number of predictions [32–34,36].

3. Results and Discussion

3.1. Colorimetric Measurement

The color parameters of the samples are listed in Table 1.

Table 1. The color parameters of the samples.

	L	**a**	**b**	**C**	**ho**
Reference	72.1	1.2892	15.8550	15.9073	85.3516
Effect of Mordant Concentration (Samples Dyed for 2 min After Mordanting for 4 min)					
Concentration (%)					
0.5	57.3	4.1	19.7	20.1	78.2
0.75	58	1.4	12.3	12.3	83
1	57.3	1.8	13.9	14	82.2
1.25	56.1	1	9.1	9.2	83.4
1.50	58.4	1.1	10.8	10.9	83.8
1.75	60.1	1.6	11.5	11.6	82
2	56.7	1.8	11.9	12.1	81
Effect of Mordanting Time (Samples Dyed for 2 min After Mordanting with 1.75% Aluminum Potassium Sulfate)					
Time (min)					
1	56.6	1.3	10.4	10.4	82.3
2	59.2	2	11.9	12	80.1
3	57.8	1.8	12	12.1	81.4
4	60.1	1.6	11.5	11.6	82
5	59.2	1.5	11.6	11.7	82.5
Effect of Dyeing Time (Samples Mordanted for 4 min with 1.75% Aluminum Potassium Sulfate)					
Time (min)					
2	60.1	1.6	11.5	11.6	82
4	55.9	1.7	11.1	11.2	80.8
6	56.8	1.6	8.9	9.1	79.4
8	55.9	1.3	9.8	9.9	82.2
10	53.3	1.3	11.1	11.1	83.1

Natural dyes have significant advantages over synthetic dyes in terms of performance. These advantages can be listed as follows. Natural dyes are typically derived from plant or mineral sources, meaning they contain fewer chemicals. Since they do not contain volatile organic compounds, natural dyes are generally not harmful to human health. Additionally, natural dyes can offer unique and warm tones, creating a distinct aesthetic. The dyeing of merino wool fibers with natural dye extracted from horse chestnut shell at pH 5 revealed that electrostatic attraction forces play a role in the dyeing of ionic wool fibers. In addition to electrostatic attraction forces in wool fiber dyeing, hydrogen bonds, Van der Waals forces, coordinates, and covalent bonds may also contribute, depending on the structure of the natural dye's source. All wool fibers consisting of polypeptide macromolecules contain free amino (NH_2) and carboxyl (COOH) groups. Around the iso-ionic point of wool fibers (pH 5), these amino and carboxyl groups exist in ionic form, with equal numbers of (+) and (−) charged groups ($^{+}H_3N$ and COO^{-}). Natural dye molecules interact with these (+) and (−) charged groups on the wool fiber through the mordant substance.

The CIELab system is generally utilized in spectrophotometric measurements, with L, a, b, C, and ho as the parameters. According to the literature, L represents the black–white scale (lightness), while a, b, C, and ho represent the red–green scale, yellow–blue scale, chroma, and hue angle, respectively. Generally, color can be defined by three main characteristics: lightness (or luminance), which measures a color's reflection of light; saturation, which indicates a and b colorfulness; and hue, which corresponds to the dominant reflected wavelength of the color. If the L value of a reference sample is smaller than that of another sample, this indicates that the color is darker than the reference. According to Table 1, the color coordinates (L, a, b, C, and ho) vary with mordant concentration, mordanting time, and dyeing time. With the increase in mordant concentration and mordanting time, better binding of the dye molecules to the fibers is achieved, which affects the CIElab values of the samples. The dyeing time determines how long the fibers are exposed to the dye molecules, with color intensity generally increasing as the time lengthens. The reason for these results is thought to be that mordant concentration, mordanting time, and dyeing time lead to the samples absorbing different light wavelengths [31]. Color quality can express the brightness, fastness values, and color tone of the textile material resulting from the dyeing process.

When the samples were evaluated in terms of color quality, mordant concentration, mordanting time, and dyeing time were found to improve the color quality of the samples. Pre-mordanting with aluminum potassium sulfate resulted in significant darkening and color change effects, as evidenced by the lower lightness (L*) values compared to the unmordanted samples. According to Table 1, after the pre-mordanting process, the L values of the samples decrease with increases in mordanting time, mordant concentration, and dyeing time. This is due to the coloring effect of the horse chestnut shell's natural dye, which occurs as dye molecules penetrate into the merino wool fibers. It can be concluded that the pre-mordanting process with aluminum potassium sulfate leads to an increase in the darkness of the samples.

Aluminum potassium sulfate, a double sulfate of potassium and aluminum, is the most extensively used aluminum mordant for natural dyeing. The amount of mordant required depends on the desired shade, with more mordant needed for darker hues. Due to ecological aspects, the recommended concentration for using aluminum potassium sulfate is 2%. Mordanting is a treatment that is utilized not only to modify the shade but also to firmly bond the colorant with the fabric via complex formation. In addition, microwave irradiation during the dyeing process does not alter the chemistry of the merino wool fiber; rather, it physically modifies the wool's surface to achieve maximum dye yield. Furthermore, adding metal salts is also beneficial for merino wool fiber dyeing because they enhance the process by transferring the colorant from the medium to the fibers. A small amount is insufficient to complete the task, while too much can cause wear, aggregation, or inadequate fixing during the finishing phase [37]. The values of a, b, C, and ho did not show any correlation. Furthermore, it can be concluded that the sustainable natural dye extracted from horse chestnut shells can be used as a potential substitute for synthetic dyes like olive drab. The major components of horse chestnut include a complex mixture of triterpene saponin glycosides known as escin 3, along with other compounds such as flavonoids, tannins, and oligosaccharides. The dyeing mechanism of natural dye obtained from horse chestnut shells involves tannin interactions with merino wool fibers. Tannin penetrates the merino wool fabric during the dyeing process and forms additional hydrogen bonds with the merino wool fibers, thus aiding in color fixation and the dispersion of dye molecules [38,39]. The use of microwave radiation in the dyeing and mordanting process ensures energy and time savings compared to conventional methods. Furthermore, the dyeing and mordanting processes are more eco-friendly, sustainable, and green when using the microwave-assisted method [40].

3.2. Effect of Mordant Concentration on Color Strength

The color strength value of the samples dyed for 2 min after being mordanted for 4 min was examined to determine the effect of mordant concentration on the color strength of the dyeing. The color strength of the samples is given in Figure 4.

Figure 4 indicates that the color strength generally improves with a rise in mordant concentration, and the optimum mordant concentrations were found to be 0.5, 1, and 2 g/L. The literature reports that metal salts form a more favorable and stronger interaction between fibers and natural dyes. According to earlier studies, color strength value increases as a result of chelate complex formation between metal salt-mordanted merino fiber and dye molecules [41]. Such strong interactions between the merino fiber and dye molecules lead to higher dye uptake [42]. In the mordanting process, the use of metal salts at a maximum concentration of 2% is considered a green and ecological dyeing practice. During the dyeing operation, these metals form stable complexes on the merino wool fiber. Low concentrations of metal salt mordants do not form strong complexes, while high concentrations may lead to aggregates of the complex on the fibers, which may result in unfixed substrates and lower K/S values after washing [40]. In addition, the increased interaction (higher color strength values) between aluminum potassium sulfate-mordanted merino wool fibers and natural dye molecules is due to stable chelate complex formation, as reported in our earlier studies [12]. Rather et al. dyed merino wool fibers with *Cinnamomum camphora* waste leaves extract after pre-mordanting with alum at a 5% concentration. The K/S values of the samples were determined to be 4.83 for conventional dyeing and 5 for the ultrasonic-assisted method [43]. Benli et al. dyed wool fabric with different natural dye sources, including *Humulus lupulus* L., *Rubia tinoctorum* L., and *Quercus* L., after alum pre-treatment at a 3% concentration. The K/S values of the samples were found to be 3.1 for *Humulus lupulus* L., 20.7 for *Rubia tinoctorum* L., and 22.2 for *Quercus* L. [44]. In another study, wool fabric was dyed with natural dye extracted from Dalbergia Sissoo. The samples were mordanted with Aloe Vera and Amla using pre-mordanting, post-mordanting, and meta-mordanting processes. The K/S values of the samples ranged from 5.417 to 13.680 [45]. Mirnezhad et al. investigated the dyeing properties of 5% alum-mordanted wool fabric dyed with natural dye extracted from Ziziphus bark. The K/S value of the sample was predicted to be 3.75 [46]. These studies show that the K/S values of dyed samples vary depending on factors such as mordant concentration, mordanting time, dyeing time, mordant type, natural dye properties, fiber behavior, dyeing method, mordanting method, and more.

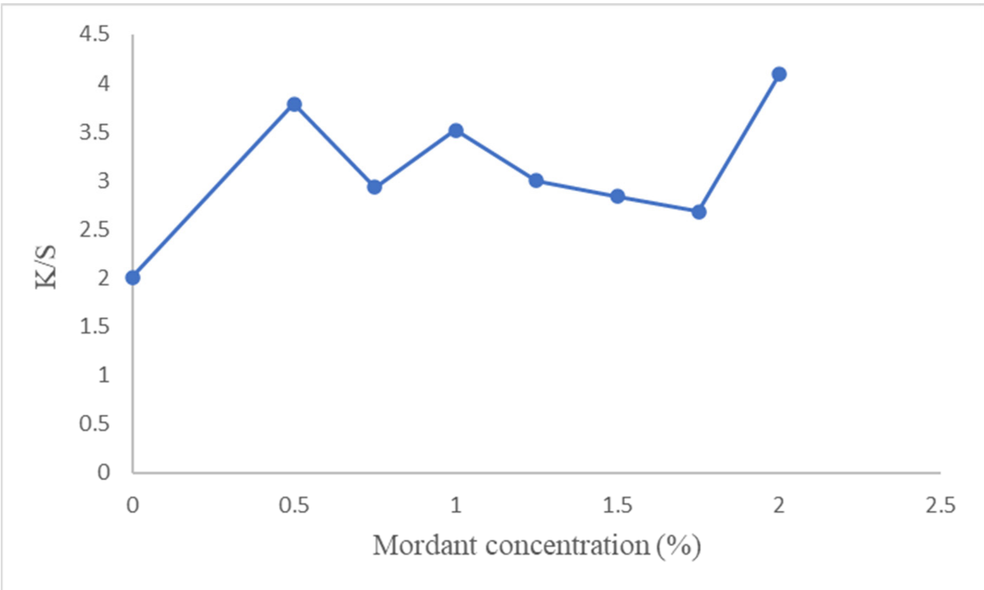

Figure 4. The effect of mordant concentration on color strength value of the samples dyed for 2 min after being mordanted for 4 min.

3.3. Effect of Mordanting Duration on Color Strength

The color strength value of the samples dyed for 2 min after mordanting with 1.75 g/L aluminum potassium sulfate salt was examined to determine the effect of mordanting duration on the color strength of the dye. The color strength of the samples is shown in Figure 5.

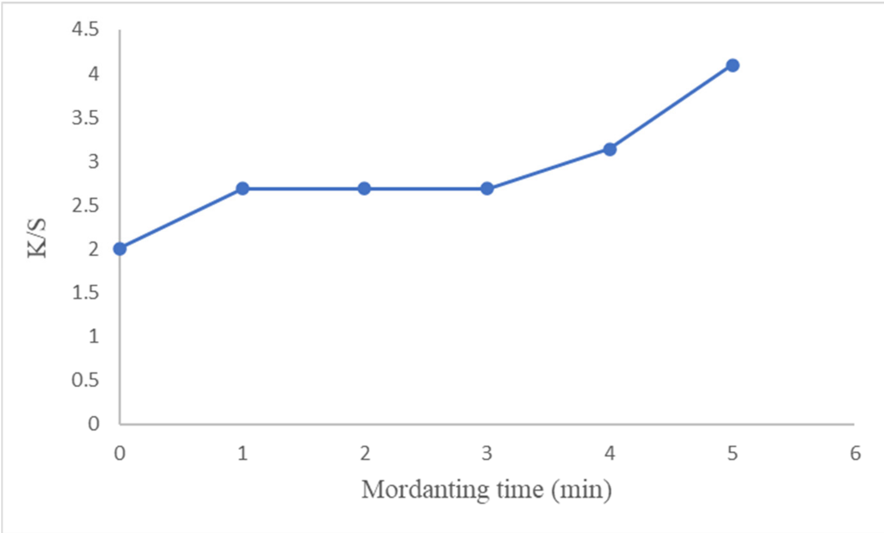

Figure 5. The effect of mordanting duration on color strength value of the samples dyed for 2 min after mordanting with 1.75 g/L aluminum potassium sulfate salt.

In this study, the mordanting and dyeing processes were carried out using a microwave-assisted method. In the microwave-assisted method, mass transfer kinetics driven by microwave-assisted treatment during mordanting and dyeing made the natural coloring process more sustainable, green, ecological, time-efficient, and cost-effective [40]. According to Figure 5, the color strength value of the samples generally increases over time, and the optimum mordanting duration is 4 min. Typically, metal salts form more favorable and stronger interferences between fibers and natural dyes. With an increase in mordanting duration, the interactions between wool fibers and natural dyes are thought to increase [41]. During mordant treatment, the metal ions may create coordinate complexes with amino groups of merino wool fiber. Certain coordination sites become available to form complexes with dye molecules once they interact with the fiber. These metals combine with the dye-infused fiber to form a binary complex. The increased dye uptake was caused by enhanced contact between the fiber and the dye molecule as a result of this strong coordination tendency [42]. Furthermore, the mordanting duration increases the interaction between merino wool fiber functional groups (amine functionality) and dye functional groups (hydroxyl and carbonyl groups), leading to increased dye exhaustion values, which can be directly correlated with the higher color strength values of the dyed merino wool fibers [47].

3.4. Effect of Dyeing Duration on Color Strength

The color strength value of the samples mordanted for 4 min with 1.75 g/L aluminum potassium sulfate salt was examined to determine the effect of dyeing duration on the color strength. The color strength of the samples is shown in

Figure 6 shows that the color strength increases with increasing dyeing duration; the optimum dyeing duration was determined to be 10 min. In the literature, it has been reported that microwave application provides greater values in obtaining darker shades through mass transfer kinetics. Instead of using traditional heating, microwave radiation causes the dye to move quickly from the plant cell wall to the solvent without altering its physiological properties. This process is known as mass transfer kinetics. Longer microwave treatment times may lead to greater interaction between dyes and fibers compared to shorter treatment times. Additionally, the duration of contact between the merino wool fiber and the dyeing bath generally plays a significant role in achieving effective natural dyeing of merino wool. Insufficient contact time reduces the kinetic energy of dye molecules, limiting their movement toward the fiber [48,49]. Microwave irritation, through mass transfer kinetics, helps release the colorant from the horse chestnut shell into the dye bath, resulting in a high color strength yield as the dyeing time increases. Furthermore, electrostatic and ion-dipole forces of attraction are basically responsible for improved dye exhaustion rates as a result of the protonation of amine (-NH_2) functional groups of the merino wool fiber (formation of ammonium (-NH_3^+) ions) [43]. Longer dyeing times allow the natural dye molecules to adhere more effectively to the merino wool fibers, resulting in an increase in the K/S value. Merino wool fiber samples were dyed with horse chestnut shell natural dye using a microwave-assisted method for different durations ranging from 2 to 10 min. The absorption of the natural dye increased with the dyeing time. In addition, at 10 min, the amount of dye absorption in the merino wool fiber is complete, and the sites of the merino wool fiber are saturated [46]. Figure 6.

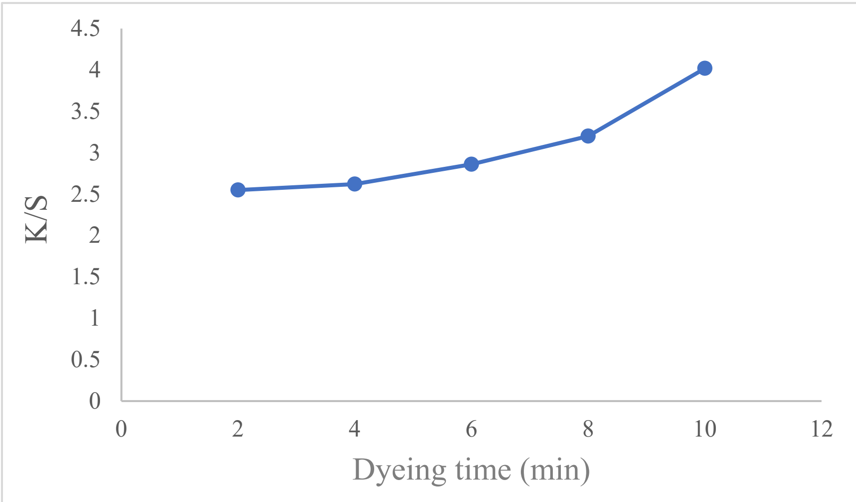

Figure 6. The effect of dyeing duration on color strength value of the samples mordanted for 4 min with 1.75 g/L aluminum potassium sulfate.

3.5. Fastness Properties

The fastness behaviors of the samples are presented in Table 2.

The light, washing, and rubbing fastness behaviors of the samples are given in Table 2. Fastness results were evaluated using a grey scale, ranging between 1 and 5. Table 2 indicates that the light fastness of the dyed merino wool fibers is not significantly affected by the dyeing and mordanting parameters, and it is generally good. In the literature, it has been reported that one of the main challenges of natural dyes is their low light fastness. Additionally, light fastness can vary depending on several factors, including the nature of the fiber, the physical and chemical structure of the dye, the mordant type, and the dye concentration [50]. After the mordanting process, the light fastness of the samples slightly improves because of stronger interactions between the fiber and the dye particulars (ionic/covalent bonding) during mordanting treatment [51,52].

Table 2 indicates that the washing fastness of the samples generally improves with mordant treatment and increasing mordant concentration. Color staining on the wash of the samples is rated as good for all the multi-fiber fabrics, with the color change of the dyed samples ranging from 3 to 4. Mordanting and dyeing parameters did not significantly alter the washing fastness of the samples. Furthermore, the pre-mordanting process using aluminum potassium sulfate improves the washing fastness of the dyed merino wool fibers because of the stronger interactions between the fiber and dye.

The rubbing process tends to move dye molecules away from the surface of the fiber. However, the dye molecules are not easily removed by rubbing when intermolecular hydrogen bonding and metal complexation occur between the dye molecules and the fiber through mordants. After the mordanting process, merino wool fibers demonstrate better rubbing fastness, as the bonding with the metal salt mordant is enhanced [51].

Table 2. The fastness properties of the samples.

	Light Fastness	Wash Fastness							Rub Fastness	
		Color Change	CA	CO	PA	PET	PAN	WO	Dry	Wet
Reference	1–2	2	2	2–3	3	3	3	2–3	2	2–3
Effect of Mordant Concentration (Samples Dyed for 2 min After Mordanting for 4 min)										
Concentration (%)										
0.5	2–3	3	3	3–4	4–5	4–5	3–4	4	2–3	3–4
0.75	2–3	3	3–4	3–4	4–5	4–5	4	4	2–3	3–4
1	2–3	3–4	3–4	4	4	4–5	4–5	4–5	3	3–4
1.25	2–3	3–4	3–4	3	3–4	3–4	3–4	3–4	3	4
1.50	2–3	3–4	3–4	4	4–5	4–5	4	4	3–4	4
1.75	3	3–4	3–4	3–4	3–4	4–5	4–5	3–4	3–4	4
2	3	3	4	3–4	4	4–5	4–5	4–5	3–4	4
Effect of Mordanting Time (Samples Dyed for 2 min After Mordanting with 1.75% Aluminum Potassium Sulfate)										
Time (min)										
1	3	3	3–4	3–4	3–4	4	4	3–4	3–4	4
2	3	2–3	4	3–4	4	4	4	4	3	3
3	3	4	4	3–4	3–4	4	4	3–4	3	4
4	3	3–4	3–4	3–4	3–4	4–5	4–5	3–4	3–4	4
5	3	4	3–4	4–5	4	4–5	4–5	4–5	3	3–4
Effect of Dyeing Time (Samples Mordanted for 4 min with 1.75% Aluminum Potassium Sulfate)										
Time (min)										
2	3	3–4	3–4	3–4	3–4	4–5	4–5	3–4	3–4	4
4	3	3–4	4–5	3–4	4–5	4	4–5	4	3–4	4
6	3	3–4	4	4	4	4–5	4–5	4	3	3–4
8	3	3	4–5	3–4	3–4	4–5	5	4	3–4	3–4
10	3	2–3	4–5	4	5	5	5	4–5	3	3–4

Haji dyed wool yarn with a natural dye extracted from Berberis vulgaris root using aluminum potassium sulfate mordant at a 2% concentration. Washing fastness, rubbing fastness (wet), rubbing fastness (dry), and light fastness were determined as 5, 3–4, 4–5, and 5–6, respectively [41]. Bulut et al. investigated the fastness properties of wool yarn dyed with natural dye obtained from Rosa Damascena Mill, with fibers mordanted using biomordants and metal salts, including aluminum potassium sulfate at a 2% concentration. The light fastness of the sample was found to be 4 [53]. Gautam and Goel investigated the color fastness of merino yarn dyed with natural dye obtained from Tesu flowers after pre-mordanting with aluminum potassium sulfate at a 0.5% concentration. While the light fastness of the samples was rated as fair, dry rubbing fastness was excellent [54]. Yusuf et al. dyed wool fibers with madder root natural dye after pre-mordanting with different concentrations of aluminum potassium sulfate. For a 2% concentration alum pre-treatment, light fastness was rated as 5, washing fastness as 4–5, and rubbing fastness (wet) and rubbing fastness (dry) as 5 and 4–5, respectively [55]. Miah et al. investigated the fastness properties of alum pre-mordanted wool fabric dyed with the natural dye of an onion's outer shell. The pre-mordanting process was carried out with a 2% concentration. Washing fastness was determined to be 4, while rubbing fastness in dry and wet conditions was determined to be 5 [56]. Cuce dyed pre-mordanted wool fabric with aluminum potassium sulfate and Rose canina leaf natural dye at a 20% mordant concentration. The light, rubbing (dry and wet), and washing fastnesses of the samples were estimated to be 2–3, 4, 3–4, and 3, respectively [57]. The fastness behaviors observed in the present study's samples are similar to those found in other studies using different natural dye sources and 2% aluminum potassium sulfate pre-mordanting.

3.6. Feed-Forward Neural Network Results

The proposed FFNN model was performed in MATLAB R2020b. In the model, the input dataset is symbolized by a 105 × 3 matrix—105 samples of three elements—while the output dataset is represented by a 105 × 1 matrix for the K/S dyeing property output—105 samples of one element. Training (70%, 73 samples), validation (15%, 16 samples), and testing (15%, 16 samples) were the three subsets generated by randomly splitting the input dataset into these three categories. The experiments were repeated many times, and the success of the proposed model was observed for different subsets. Thus, it was found that the model achieves close and successful results for different subsets, and the replicability of the model is guaranteed. The neural network was educated utilizing specimens from the training subset during the training process, and the network was then arranged based on its error. The generalization of the network was evaluated using samples from the validation subset, and the training process was stopped when further improvements were no longer observed. Consequently, the samples from the testing subset procure a separate measure of the network's performance both during and after training. The regression results of the suggested model are given in Table 3 in terms of training, validation, testing, and all subsets. The correlation between estimated outputs and actual targets is specified using regression values. A regression value of zero denotes a random relationship, whereas a regression value of one denotes a close relationship. By selecting 10 neurons in the hidden layer, the regression value of 0.90355 for the entire dataset was achieved for the K/S dyeing property, as shown in Table 3. Additionally, the regression results for training, validation, and testing subsets were 0.93955, 0.91319, and 0.87557, respectively. Figure 7 depicts the experimental and estimated values for the K/S dyeing property. This graph illustrates that the experimental and estimated results are relatively similar to one another or equal in the majority of tests. The FFNN-based results all reveal that the proposed model can accurately predict the K/S dyeing property of samples.

Table 3. Regression results of the proposed FFNN model.

Training	Validation	Testing	All
0.93955	0.91319	0.87557	0.90355

The model used in this study used the dyeing time, mordant concentration, and mordanting time as input variables and the K/S dyeing property as the output variable. The success of this proposed model may vary depending on the type of fiber, as different fibers interact differently with the chemical structure of the dye and the parameters in the dyeing process. Factors such as fiber structure, absorption capacity, and other fiber properties may limit the generalization capacity of the model. In addition, the chemical structures of different dye types and the ways they interact with fibers also differ. Since the proposed model was trained with merino wool fiber and a specific dye type, it may not achieve the same performance for other dye types. Additionally, since the model was trained with a specific dataset, it may have learned patterns specific to this dataset. In this case, its generalization ability may be limited when faced with new datasets with different dyeing conditions. To overcome these limitations, we plan to train the model with an expanded dataset on different dye and fiber types in future work. On the other hand, similar models that we have previously developed using different parameters for various fibers [30,31] have also shown successful results in predicting dyeing properties. This shows that the model proposed in this study is promising.

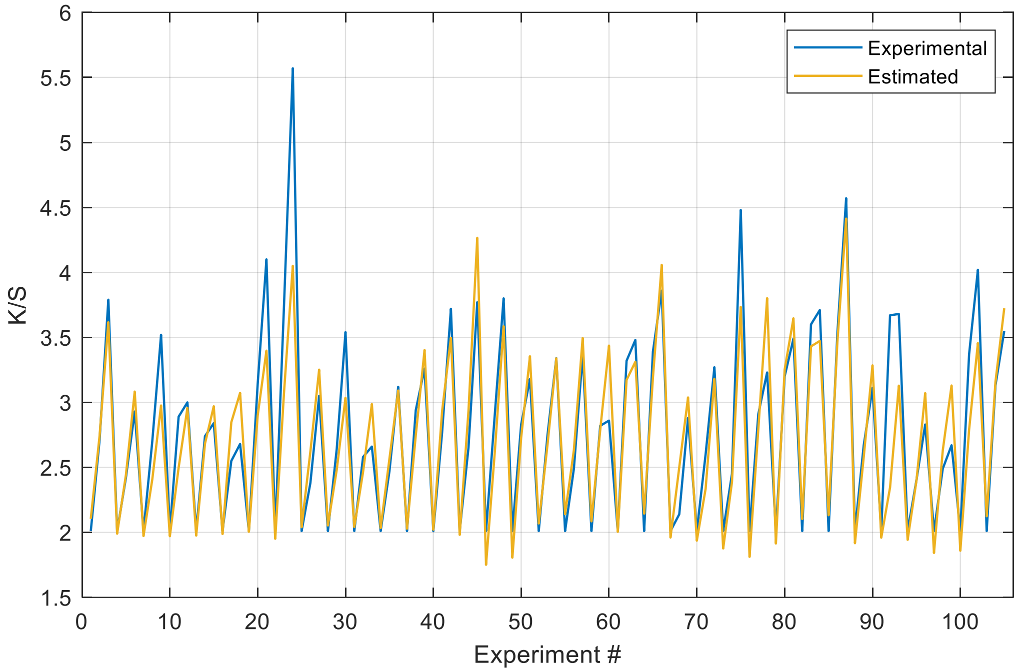

Figure 7. Comparison of experimental and estimated *K/S* values of dyed samples.

3.7. Environmental Benefits and Results

The use of natural dyes in the process of dyeing textile fibers offers numerous environmental advantages. Natural dyes typically contain fewer chemicals than synthetic dyes, resulting in reduced toxin emissions and environmental pollution. Additionally, natural dyes are biodegradable, and their production often involves raw materials derived from a variety of plants and other natural resources, which supports biodiversity. Less water is required in the production of natural dyes compared to synthetic dyes, making natural dye use beneficial for protecting water resources. Natural dyes generally have biodegradable and recyclable properties, making waste management easier. While synthetic dyes can sometimes cause skin irritation or allergic reactions, natural dyes are generally safer and skin-friendly. These advantages reflect the potential for natural dyes to be integrated into the sustainable and green textile industry. In addition, natural dyes are usually biodegradable because they are derived from plant or animal sources. Synthetic dyes are often derived from petroleum, increasing the risk of fossil fuel dependency and environmental pollution. Furthermore, when the mechanism for dyeing wool fibers with many different synthetic dyes is examined, the dyeing process is usually carried out at 100 °C and takes up to 150 min, including the rinsing process. In terms of sustainability, natural dyes appear to be more advantageous in terms of the environment, dyeing processes, and health. In this study, horse chestnut shells were used as a natural dye source for dyeing merino wool fiber. According to the results, it can be deemed that the green and sustainable natural dye extracted from horse chestnut shells could potentially act as a substitute for synthetic dyes, such as olive drab. Notably, no existing studies use the natural dyes extracted from horse chestnut to color merino wool fibers using a microwave-assisted method. In the dyeing and mordanting process, the microwave-assisted method offers numerous benefits, including fast heating, energy efficiency, more homogenous heat distribution in dyeing baths, reduced chemical use, and minimized heat loss. These characteristics make the microwave-assisted method greener, more sustainable, and more ecological. All these advantages of the microwave-assisted method improve the efficiency of the dyeing and mordanting processes. The primary contribution of microwave energy to textile wet processing is its ability to save energy and time. In textile wet processing, microwave heating is faster, more uniform, and more efficient than conventional methods. In addition, microwave energy provides easy penetration of particles into fibers, thereby reducing heat transfer problems [38]. Predicting dyeing properties using ANNs offers a number of advantages, especially in sectors such as the textile industry. Color prediction and harmony, color quality and consistency, color-compatible product development, and cost reduction in color laboratories are some of the advantages of ANNs for the textile industry. To achieve accurate and reliable color predictions, it is essential to train ANNs correctly and feed them with high-quality datasets. Additionally, selecting models that meet industry-specific requirements and usage scenarios is crucial. In this study, an FFNN model was used to predict the K/S dyeing property of merino wool fiber.

4. Conclusions

The increasing demand for green production methods has encouraged a shift toward their widespread usage in many fields. Horse chestnut is a resource for sustainable natural dyes in textile materials, known for its antiviral, antioxidant, and antibacterial properties. In this study, an eco-friendly dyeing process was performed on merino wool fiber via the use of natural dye extracts from the *Aesculus hippocastanum* (horse chestnut) shell using the microwave-assisted method. Before the dyeing process, the merino wool fiber underwent a pre-mordanting process with aluminum potassium sulfate using the microwave-assisted method with different concentrations, which are 0.5, 0.75, 1, 1.25, 1.50, 1.75, and 2% for 1, 2, 3, 4, and 5 min, respectively. Afterward, the samples were dyed for 2, 4, 6, 8, and 10 min using the microwave-assisted method. The dyeing process, spectrophotometric analysis, and light, rubbing, and washing fastnesses of the samples were analyzed in terms of mordant concentration, mordanting time, and dyeing time. The results show that the color strength, light, washing fastness, and rubbing fastness of the dyed merino wool fiber increase with the pre-mordanting process. The spectrophotometric measurement results demonstrate that mordant concentration, mordanting, and dyeing duration affect the color coordinates. Owing to strict environmental regulations, there has recently emerged a dire need for eco-friendly products in textiles using modern tools. Being a clean, sustainable, and uniform heating source, microwave radiation made good use of natural dyes for merino wool dyeing under mild conditions. Microwave-assisted dyeing creates energy and time savings compared to conventional methods. In addition, an FFNN-based model consisting of input, hidden, and output layers for forecasting and evaluating the K/S dyeing property of merino wool fiber was proposed. This model's output is the K/S dyeing property, whereas its inputs are the dyeing time, mordant concentration, and mordanting time. For the K/S dyeing property, the proposed model achieved regression values of 0.93955, 0.91319, 0.87557, and 0.90355 for training, validation, testing, and all subsets, respectively. The experimental results of this investigation indicate that the proposed model can be utilized to forecast the K/S dyeing property of merino wool fiber.

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