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Broadening Color Shade of Dyed Wool Fibre with Binary and Ternary Natural Plant Dye Combinations

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ABSTRACT

A limited selection of natural dyes' color impedes the development of textile dyeing with natural plant dyes. Inspiring by the conventional coloration of textiles with a combination of three synthetic dyes generally, the present work is to investigate the broadening color shade of dyed wool fiber with ternary natural dye combinations of madder red (MR), gardenia yellow (GY), and gardenia blue (GB) without mordants in a decamethylcyclopentasiloxane (D5) medium. The wool yarn was wetted in an aqueous solution of pH 3 to own a 300% pickup rate, followed by immersion in a D5 medium containing 2% of alcohol ethoxylate (AEO-3) and solid natural dyes at 90°C for 90 min for coloration. The colorfastness to washing was achieved at a 4–5 for fading and a 5 rating for staining for all colors. The XRD patterns and TGA analysis confirmed that the dyeing procedure did not affect the crystallinity nature and stable thermal tendency. SEM images and cross-sections showed that the dyeing procedure did not damage the morphological structure of the wool fiber surface, and the dyes were evenly distributed. Finally, many color shades of dyed fibers were prepared with various dyes' ratios.

摘要

天然染料的颜色选择有限，阻碍了天然植物染料染色的发展. 受三种合成染料组合对纺织品常规染色的启发，本工作研究了在十甲基环五硅氧烷（D5）介质中，用茜草红（MR）、栀子黄（GY）和栀子蓝（GB）三元天然染料组合对染色羊毛纤维的增宽色度. 将毛纱在pH为3的水溶液中润湿，使其具有300%的拾取率，然后在90℃下浸入含有2%乙氧基醇（AEO-3）和固体天然染料的D5介质中90分钟进行着色. 所有颜色的褪色色牢度为4-5，染色色牢度为5. XRD图谱和TGA分析证实，染色过程不影响结晶性质和稳定的热趋势. SEM图像和横截面显示，染色过程没有破坏羊毛纤维表面的形态结构，染料分布均匀. 最后，用不同的染料配比制备了多种颜色的染色纤维.

KEYWORDS

D5 solvent; wool dyeing; natural dye; color shade; crystallinity; thermal stability

关键词

D5溶剂; 羊毛染色; 天然染料; 色调; 结晶度; 热稳定性

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Introduction

The ecosystem and human health are negatively affected by the continuous discharge of untreated wastewater containing synthetic dyes from textile industries (Al-Tohamy et al. 2022; Fazal Ur et al. 2022; Pervez et al. 2019; Pervez, Balakrishnan, et al. 2020; Riza, Ehsan, and Hoque 2021). Specifically, aromatic azo dyes based on benzidine are carcinogenic and harmful to human health; therefore, their production is banned in several countries. This has encouraged scientists to look for relatively safe and biodegradable natural dyes as alternatives to synthetic dyes (Ding and Freeman 2017; Fang, Meng, and Zhang 2022; Jiang et al. 2022; Jose, Gurumallesh Prabu, and Ammayappan 2017; Pervez, Balakrishnan, et al. 2020). Natural dyes are recognized to be environmentally safer than synthetic colorants because of their non-toxicity and biodegradability (Haji 2020; Pervez et al. 2020, 2021, 2023). Some natural dyes also have antimicrobial and antioxidant properties, and the textile treated with these dyes can protect human skin from UV rays. Moreover, they may biodegrade easily in the environment, and some of them are used in herbal medicine (Eyupoglu et al. 2023; Ibrahim et al. 2010; Peng et al. 2023; Shafiq et al. 2021; Zhou et al. 2015). However, one notable drawback of natural dyes is their low affinity for textile fibers (Eyupoglu, Eyupoglu, and Merdan 2022; Nambela, Haule, and Mgani 2020). In addition, natural dyes have limited practical use because of limitations such as the difficulty in achieving a good number of deep shades, a limited number of shades, and poor colorfastness properties (Mia et al. 2022).

In addition, in typical natural dyeing processes, heavy metal salts are commonly utilized as mordants to maximize the colorfastness of natural dyes. However, this mordant has the ability to have adverse effects on both the environment and public health (Rehman et al. 2022; Velusamy et al. 2021). In recent years, several methods have been devised to enhance the process intensification of natural dyeing, including microwave irradiation, plasma treatment, and other techniques (Batool et al. 2022; Mahboob et al. 2023; Shahid et al. 2023). Nevertheless, these approaches encounter challenges such as energy consumption, limited colorfastness, and solubility (Adeel et al. 2022; Haji and Vadood 2021). In contrast, non-aqueous dyeing with decamethylcyclopentasiloxane (D5) was recently developed as an alternative to the typical water-based method of dyeing textile materials. This technique reduces the need for metal mordants as well as other dyeing additives (Hossain, Liang, et al. 2021; Omerogullari Basyigit et al. 2023; Pei et al. 2019). Furthermore, the usage of natural dye in conjunction with an organic solvent such as D5 reduces the burden of pollution caused by synthetic colorants and the accompanying effluent treatment expenditures.

The D5's low surface tension, strong hydrophobicity, and superior thermal conductivity make it a safe and effective dyeing medium (Tang et al. 2018). D5 has been utilized in this dyeing method to accelerate the dye dispersion and the overall dyeing procedure (Alebeid et al. 2020; Hossain et al. 2021). Natural dyes within a D5 medium have garnered significant interest in contemporary times due to their notable sustainability attributes, efficiency, and economic feasibility. Previous studies have shown that natural dyeing in a D5 medium enhanced the fixing rate, adsorption kinetics, and color strength (Hossain et al. 2022; Hossain, Liang, et al. 2021). Moreover, using a D5 medium in the process of natural dyeing resulted in the production of water that is free from effluent, contributing to environmental sustainability (Hossain, Liang, et al. 2021; Pervez et al. 2023).

Therefore, this present work aims to determine the feasibility of employing natural dyes such as madder red (MR), gardenia yellow (GY), and gardenia blue (GB) in the D5-based solvent dyeing of wool yarns. In this work, the wool yarns were dyed with various dye combinations (GY, GB, and MR), and the effects on shades produced, color strength (K/S), colorfastness to washing, and mechanical properties of colored wool yarns were evaluated. X-ray diffraction (XRD) and thermogravimetric (TG) analyses were used to determine the effect of the D5 medium on the crystallinity and thermal stability of the wool fibers, respectively. In addition, scanning electron microscopy (SEM) was used to determine the extent of surface morphological damage of wool fibers. The light microscope image of the cross-section of the dyed wool fiber was examined to investigate the distribution of ternary natural dyes in the dyed wool fibers. Finally, the color shade of dyed wool fiber was successfully broadened without mordants, which hints that the general issue of excessive metal compounds in the natural plant dyed wool fiber was sorted out as well.

Experimental

Materials

Scoured pure wool yarns (48 NM/2) were selected for this study and purchased from Hengyuanxiang (Group) Co., Ltd., Shanghai, China. Madder red (MR) was supplied by Henan Zhongda Hengyuan Biotechnology Co. Ltd (China), and gardenia yellow (GY) and gardenia blue (GB) were sourced from Wuhan Green Food Biological Engineering Co., Ltd. (China). Sinopharm Chemical Reagent Co., Ltd has supplied acetic acid ($C_2H_4O_2$) and sodium acetate trihydrate ($CH_3COONa{\cdot}3\,H_2O$). A primary alcohol ethoxylate (AEO-3) (RO $(CH_2CH_2O)_nH$) and a nonionic detergent (Luton 500) were purchased from Shanghai Aladdin Biochemical Technology Co., Ltd. (China) and Dalton Company (China), respectively. The D5 solvent (decamethylcyclopentasiloxane) was purchased from Jiangxi Bluestar Xinghuo Silicones Company (China).

Dyeing process

The dye mass ratios are from 1:0 to 0:1 with an interval of 0.1 for the binary dyes' mixtures of MR:GY, MR:GB, and GY:GB. Meanwhile, the dye mass ratios (MR: GY: GB) of 0.6:0.2:0.2, 0.4:0.3:0.3, 0.2:0.6:0.2, and 0.2:0.2:0.6 were selected for the ternary dyes' mixture. The total dye mass of mixtures was 3% o.m.f (on the mass of fiber). The AEO-3 (2%, v/v) was added to D5 to enhance the dispersibility of natural dyes in the D5 solvent. Powdered MR, GY, or/and GB dyes were added to it, and they were dispersed in the D5 solvent by sonication (Ultrasonic Cleaner, YM-040S, Shenzhen FANGAO Microelectronics Co., Ltd., China) until an even dye suspension system was prepared. For all dyeings, the wool yarn was fixed at 2 g. The wool yarn was wet in an aqueous solution at pH 3 (adjusted by a mixture solution of acetic acid and sodium acetate trihydrate) and 25°C and then padded with a padder (p-BO, Foshan Shunde Yaliruo Precise Machinery Manufacturing Factory Limited, China) to obtain 300% pickup rate. The wet wool yarn was then immersed inside the dye suspension system at a liquor ratio (LR) 25:1 (i.e., 50 mL). Subsequently, the dyeings were performed in an infrared-heated rotary sample dyeing machine (Automatic Prototype, Model: A-12, AQUA, China) at 90°C (with a heating rate of 2°C min^{-1}) for 30 min. After completion of dyeing, wool yarns were treated by a spin-drier (HB-T180, Foshan Shunde HUI BAO RAN ZHENG JI XIE Factory, China) for 30 s to remove most of the residual D5 and dye solution. A short ethanol rinse was done to wash off the D5 solvent further, followed by drying at 60°C in an oven dryer. The dried-colored yarn was washed in a nonionic detergent (2 g L^{-1}) at an LR of 50:1 for 15 min at 90°C. Finally, the wool yarn was dried in an oven at 60°C to measure the K/S value.

Measurement and characterisations

Color strength and uniformity

A spectrophotometer (CHN-Spec CS-650A, Hangzhou Colour Spectrum Technology Company, China) was used to measure the CIE *Lab* values, chroma (C*), hue (h^o), and K/S at 20 randomly chosen different spots, and the average results are reported. The color difference (ΔE) of dyed samples before and after washing was calculated by Eq. 1, where b and a represent before and after wash samples, respectively. The standard deviation of K/S meant the color uniformity; this standard deviation is denoted by ($\sigma_{K/S}$).

$$\Delta E = \sqrt{\left(L_b^* - L_a^*\right)^2 + \left(a_b^* - a_a^*\right)^2 + \left(b_b^* - b_a^*\right)^2} \quad (1)$$

Colorfastness to washing

The colorfastness to washing was measured according to the ISO Test Method 105-C06:1997 standard (Test number: C2S).

Tensile strength

A yarn strength testing machine (YG061F/PC, Laizhou Electron Instrument Co., Ltd) was used to determine the tensile strength of wool yarns 10 times, according to the standard GB/T3916–2013.

X-ray diffraction analysis

The XRD pattern of the meticulously scissored wool yarn powder was determined with an X-ray diffractometer (Rigaku Ultima III, Tokyo, Japan), which was recorded from 2θ = 5–80° by increments of 0.2° min^{-1}. The crystallinity index (CI) of wool fibers was calculated with Eq. 3 (Long et al. 2013), where I_9 and I_{14} refer to the maximum and minimum intensities of the XRD pattern at around 9° and 14° of 2θ, respectively.

$$\mathrm{CI} = \frac{I_9 - I_{14}}{I_9} \times 100\% \tag{2}$$

Thermal stability analysis

The thermal stability of dyed wool fibers was assessed with a thermogravimetric analyzer (TGA/DSC1, Mettler-Toledo, LLC, Shanghai, China) under a nitrogen atmosphere (flow rate of 50 mL min^{-1}) by maintaining a heating rate of 10°C min^{-1} from 30 to 700°C.

Surface morphology analysis

The surface morphology of dyed wool fiber was examined by a field-emission scanning electron microscope (SEM) (Philips SEM 515, Germany). The wool fiber was gold sputter-coated (JSM-560, Rigaku, Japan) and then examined at 5 kV accelerated voltage after gold sputtering. Microscope images of the cross-section of wool fiber were detected with a light microscope (Leica Microsystems, DM 2500, France).

Results and discussion

Wool dyed with binary combinations of MR and GY dyes

The chromatic values (L*, a*, and b* values of CIE *Lab* color space) and color strength values of wool yarn dyed with the binary combinations of MR and GY after washing are listed in Table 1, and the dyed samples are shown in Figure 1. It is visually detected that the color shade of dyed wool samples was gradually changed from red to yellow, but the primary shade of dyed wool yarns was red color when MR was included in the combinations, which was possibly ascribed to the higher tinctorial strength of MR compared to that of GY (Lin et al. 2022). This result was also supported by the wavelength of maximum K/S values (λ_{max}) of the dyed wool samples containing MR dye, which were all at 530 nm and the same as the MR-dyed wool fiber (Sample 1 in Figure 1). Besides, when the mass ratio of MR dye went down, and the GY mass ratio went up in dyed sample combinations, the a* values went down, and the L* and b* values went up. This result shows that a positive value contributes to the yellowness of the shade, while a decreasing a* value indicates a proportionate decrease in the shade of red (Figure 1). The increase in the GY dye ratio also decreased the K/S values of the dyed samples. After washing, dyed wool yarns' color uniformity is high since the $\sigma_{K/S}$ values are lower than 1 (Lin et al. 2021).

Wool dyed with binary combinations of MR and GB dyes

Table 2 represents the chromatic values of the binary combinations of MR and GB dyes. In this part of the experiment, decreasing the MR dye by 10% while increasing the GB dye by 10% was used to

Table 1. Chromatic values and color strength of wool fibers dyed with binary combinations of MR and GY.

Sample number	Dye mass ratio		Chromatic value			Color strength		
	MR	GY	L*	a*	b*	K/S	$\sigma_{K/S}$	λ_{max} (nm)
1	1	0	36.08	41.31	1.05	14.53	0.51	530
2	0.9	0.1	35.79	40.58	1.61	13.64	0.57	530
3	0.8	0.2	37.64	41.00	1.51	11.99	0.78	530
4	0.7	0.3	37.07	40.06	1.67	12.18	0.53	530
5	0.6	0.4	37.81	40.56	3.01	11.74	0.95	530
6	0.5	0.5	40.89	37.49	3.10	8.28	0.36	530
7	0.4	0.6	40.16	35.57	1.62	8.18	0.82	530
8	0.3	0.7	45.15	30.59	3.52	5.00	0.24	530
9	0.2	0.8	46.39	30.68	4.32	4.67	0.30	530
10	0.1	0.9	52.28	20.86	7.14	2.51	0.27	530
11	0	1	72.97	−0.31	33.61	1.81	0.07	440

Figure 1. Wool yarns dyed with various dye mass ratios of binary combinations of MR and GY (samples 1–11), MR and GB (samples 1, 12–21), GY and GB (samples 11, 21–30), and ternary combinations of MR, GY, and GB (samples 31–34).

Table 2. Chromatic values and color strength of wool fibers dyed with binary combinations of MR and GB.

Sample number	Dye mass ratio		Chromatic value			Color strength		
	MR	GB	L*	a*	b*	K/S	$\sigma_{K/S}$	λ_{max} (nm)
1	1	0	36.08	41.31	1.05	14.53	0.51	530
12	0.9	0.1	35.08	38.77	−0.89	13.47	0.57	530
13	0.8	0.2	35.98	35.81	−3.10	11.31	0.40	530
14	0.7	0.3	35.15	33.92	−4.15	11.47	0.50	530
15	0.6	0.4	33.92	29.50	−6.43	10.88	0.60	530
16	0.5	0.5	34.00	27.95	−7.17	10.14	0.47	530
17	0.4	0.6	36.19	22.78	−7.27	7.43	0.36	530
18	0.3	0.7	36.64	17.13	−10.57	6.13	0.45	530
19	0.2	0.8	32.20	16.57	−11.49	8.35	0.42	530
20	0.1	0.9	36.15	8.66	−12.69	5.79	0.24	570
21	0	1	42.07	−7.33	−11.34	5.37	0.25	610

observe the effect of binary mixtures, and the dyed samples are shown in Figure 1. Compared to dyeing with binary combinations of MR and GY, the L*, a*, and b* values tend to change similarly. The increasing negative value of b* indicates that the hue became bluer as the GB dye concentration increased (Luo et al. 2019). Samples 12 (MR: GB = 0.9:0.1) and Sample 20 (MR: GB = 0.1:0:9) had K/S values of 14.53 and 5.79, respectively. These results show that MR dye had a stronger color intensity

than GB dye, which is consistent with the previous report (Lin et al. 2022). It is worth noting that the λ_{max} of Sample 20, dyed with a ratio of 0.1:0.9 (MR: GB) changed to 570 nm, while it did not change in Sample 10 dyed with a ratio of 0.1:0.9 (MR: GY). This performance indicates that the absorbed GB mass in Sample 20 could obviously change the shade of color. Interestingly, the color shade of Sample 21 is close to the cyan hue rather than the blue hue, despite the single GY dye being used, and the a* and b* values of Sample 21 fit the cyan color character. Finally, all the dyed samples exhibited color even properties since the $\sigma_{K/S}$ values are lower than 0.60 (Lin et al. 2021).

Wool dyed with binary combinations of GY and GB

The wool yarns dyed with the binary GY and GB combinations are shown in Figure 1, and their corresponding chromatic values for the dyed wool yarns are listed in Table 3. The value of b* gradually decreases as the mass ratio of GY dye decreases from 90% to 10%. As the GB dye mass ratio increased, the value of L* gradually decreased, and K/S increased, showing that the absorbed GB mass in the dyed samples increased by 22–30 (Luo et al. 2019). The λ_{max} of K/S was changed to 610 nm at Sample 24, which hints that the GB dye became primarily contributed to the color shade of dyed wool fiber when the GB dye was only 30% of the binary mixtures. By contrast, the shift of λ_{max} did not occur in the binary combinations of MR and GY and occurred at 90% of GB in the binary combination of MR and GB. In an aqueous dyebath, the tinctorial strength of MR is the strongest, and GB is the weakest (Lin et al. 2022). However, after dyeing, the color compounds of these dyes in the dyed wool fibers contributed to the K/S values. Thus, it implies that GB dye was more easily absorbed than the GY dye due to its higher affinity under the dyeing conditions. Again, the uniform color of all dyed wool fibers was presented based on the low $\sigma_{K/S}$ values.

Wool dyed with ternary combinations of MR, GB, and GY

Table 4 illustrates the chromatic values of wool yarns dyed with ternary combinations of MR, GY, and GB. Apparently, decreasing MR concentration in the ternary mixtures created a lighter color shade, such as the L* value of Sample 31 (60% of MR) is 36.36, while it increases to 43.99 of Sample 33 (20% of MR). Meanwhile, increasing GB dye concentration in the ternary mixtures decreases the lightness, such as the L* value of Sample 33 (20% of GB) is 43.99, while it is 37.23 of Sample 34 (60% of GB). Increasing its concentration in the ternary mixtures for GY dye is beneficial for preparing a light color shade. Thus, it can be claimed that increasing the ratio of MR and/or GB dye in the mixtures has a positive rule for the preparation of deeper shade while increasing GY concentration is helpful for the preparation of lighter shade. This conclusion is similar to the previous research (Pervez et al. 2023). It also works for the K/S values of dyed wool yarns. Finally, the color uniformity of wool yarns dyed with the ternary dyes' mixtures is satisfactory; the dyed samples are shown in Figure 1.

Table 3. Chromatic values and color strength of wool fibers dyed with binary combinations of GY and GB.

Sample number	Dye mass ratio		Chromatic value			Color strength		
	MY	GB	L*	a*	b*	K/S	$\sigma_{K/S}$	λ_{max} (nm)
11	1	0	72.97	−0.31	33.61	1.81	0.07	440
22	0.9	0.1	62.64	−7.55	18.52	1.97	0.06	440
23	0.8	0.2	57.04	−9.25	9.50	1.96	0.09	440
24	0.7	0.3	51.94	−9.59	2.39	2.36	0.15	610
25	0.6	0.4	53.53	−9.89	5.20	2.08	0.11	610
26	0.5	0.5	49.06	−9.28	−2.61	3.07	0.16	610
27	0.4	0.6	47.04	−8.69	−4.51	3.54	0.20	610
28	0.3	0.7	45.88	−8.56	−6.41	3.96	0.33	610
29	0.2	0.8	44.49	−8.35	−8.49	4.46	0.20	610
30	0.1	0.9	43.58	−7.92	−9.81	4.79	0.14	610
21	0	1	42.07	−7.33	−11.34	5.37	0.25	610

Table 4. Chromatic values and color strength of wool fibers dyed with ternary mixtures of MR, GY, and GB.

Sample number	Dye mass ratio			Chromatic value			Color strength		
	MR	GY	GB	L*	a*	b*	K/S	$\sigma_{K/S}$	λ_{max} (nm)
31	0.6	0.2	0.2	36.36	34.05	−2.29	10.54	0.43	530
32	0.4	0.3	0.3	36.03	28.88	−4.47	9.09	0.59	530
33	0.2	0.6	0.2	43.99	19.80	−1.76	4.14	0.20	530
34	0.2	0.2	0.6	37.23	10.06	−10.44	5.33	0.15	570

Dyeing mechanism of wool fibre with the natural dyes in D5 medium

The wool yarn fiber was first wet with an aqueous solution at pH 3, and the amino groups of wool fiber were ionized to form cationic dye sites (Lewis and Rippon 2013). The solid natural dyes distributed in the D5 medium were immediately adsorbed onto the wet wool fiber surface due to the hydrophilic property when the wet yarn was immersed inside the dye suspension system (Fu et al. 2015; Tang and Kan 2020). During the dyeing at high temperatures, the wool fiber was swollen, the gap between the cuticular cells and pore size inside the wool fiber was enlarged (Lewis 1992), and the natural dyes migrated inside the fiber and fixed there (Cai et al. 2023; Lin et al. 2022). It is confirmed by the K/S values of soaped dyed wool fiber at different temperatures from 60 to 100°C, shown in Figure S1. Simultaneously, the D5 was adsorbed on the fiber surface, preventing the dye solution from escaping from the dyed wool fiber. However, due to the rotary process, some adsorbed water molecules leached into the D5 medium, which dissolved the residual suspended solid dye in D5. The AEO-3 surfactant in D5 was mixed and covered with the dye solution to form the microemulsion (Hossain, Liang, et al. 2021), which was adsorbed into the wet wool fiber due to its high affinity to cuticular scales. Meanwhile, the wool fiber surface scale fragments were removed, making a clear surface. Figure 2 illustrates the mechanics behind the coloring process.

Colorfastness to washing

Colorfastness is an essential criterion to assess the performance of dyed textile materials. When using natural dyes, colorfastness to washing is especially crucial. Natural dyes are typically used in conjunction with mordants due to their poor colorfastness to washing (Barani, Boroumand, and Rafiei 2017). The colorfastness to washing of all soaped dyed samples was evaluated, which results are listed in Table S1. All samples exhibited a rating of 4–5 to 5 for color staining (cotton) and 4–5 for color fading. Therefore, fastness ratings indicated that the dyed wool was resistant to dye bleeding.

XRD analysis

The XRD patterns of the original fiber and Sample 32 wool fiber are shown in Figure 3. A classic wool XRD pattern is displayed, which consists of two distinctive peaks located at $2\theta = 20.68°$ and $2\theta = 9.60°$, which are generated from the α-helix and β-sheet structures of the peptide chains in wool, respectively (Niu et al. 2012). The minimum intensity of the XRD pattern curve at $2\theta = 13.60°$ is considered an amorphous area of wool fiber (Long et al. 2013). In the dyed wool fiber, the two characteristic peaks of wool fiber are at the same 2θ degree. The prominent peak of the original and dyed wool fibers are not shifted, at $2\theta = 20.68°$, and the peak intensities of both samples are overlapped, whereby it is clear that interplanar distance in the crystallinity was not affected by the dyeing procedure (Long et al. 2013). However, the intensity of peak at $2\theta = 9.60°$ slightly reduced, which was 8418 from 8977, and its minimum intensity at $2\theta = 13.60°$ decreased as well from 6592 to 6393. Therefore, the CI of the original and dyed wool fibers is 26.6% and 25.6%, respectively. The slight decrease is ignorable, indicating that the wool fiber structure was not influenced during the dyeing in the D5 medium (Luo et al. 2022).

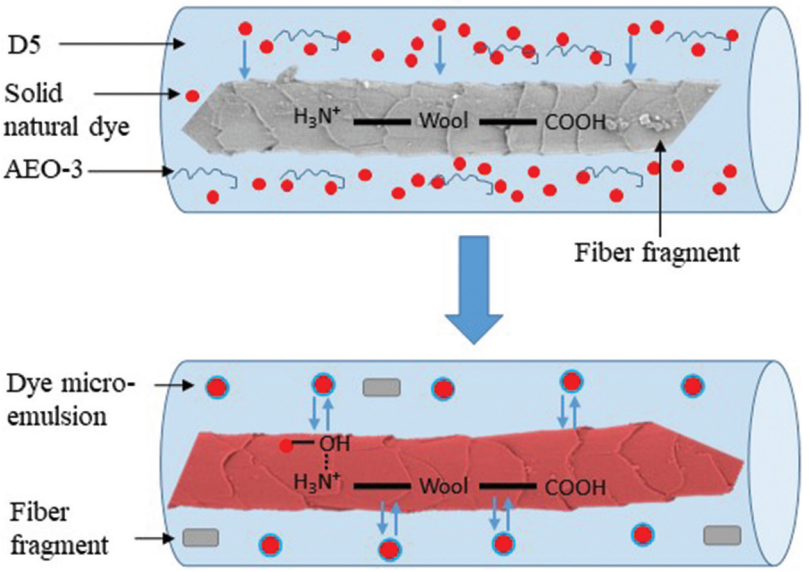

Figure 2. Dyeing mechanism of wool fiber in a suspension system.

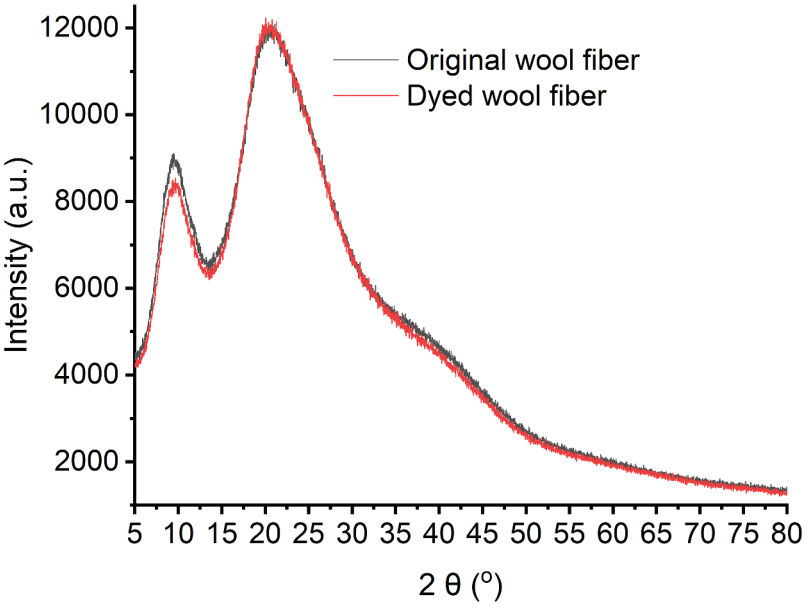

Figure 3. X-ray diffraction of the original and dyed wool fibers.

Thermogravimetric analysis

The original wool yarn and the wool yarn treated by wetting with a 300% water pickup rate at pH and 90°C for 90 min in a D5 medium were selected to do the thermogravimetric analysis. The TG curves of the original and treated wool fibers are similar, and both have two weight-loss intervals, as shown in Figure 4. In the first weight-loss interval, the strong-bond water content in the wool fiber was evaporated (Xia et al. 2016) with 10.9% of weight from 30 to 150°C, and the

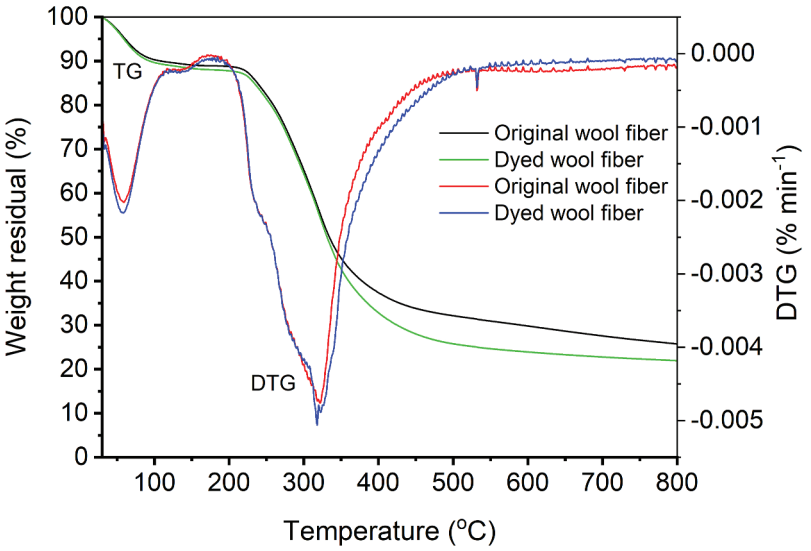

Figure 4. TG and DTG curves of the original and treated wool fibers.

fastest water loss is at 59°C. The second weight-loss interval of both wool fibers ranged from 180 to 500°C is a major weight loss, which was caused by the carbonation, fusion, and decomposed air-state release because during the primary thermal decomposition of the wool fibers, the hydrogen bonds and disulfide bonds between the molecular chains were damaged (Zhang et al. 2014). Besides, the highest water loss in the 2nd weight-loss internal is at 320°C with 56.3% and 55.3% of weight residuals for the original and treated wool fibers, respectively. The weight loss of both wool fibers shows a stable tendency in a range of 500–800°C, and the char residue masses were 25.7% and 21.9% for the original and treated wool fibers, respectively. The slightly low char residue was possibly ascribed to the volatilization of residual D5 solvent in the treated wool fiber (Pu, Zhong, and Wang 2016).

Tensile strength

The breaking force and elongation of the original wool yarn, the wool yarns treated in aqueous at pH 3 and 90°C for 90 min, and the wool yarn treated by wetting with a 300% water pickup rate at pH and 90°C for 90 min in D5 medium are showed in Figure 5. After treatment, the breaking force of wool yarn slightly decreased because the mechanical properties of the yarn deteriorated after treatment at high temperatures (Hasani 2010). However, the reduction in breaking force in D5 was greater than in water medium treatment. By contrast, the change in the elongation (%) shows a contrary tendency, and after the treatment in the D5 medium, the elongation of wool yarn mildly increased to 22.1% from 19.1%. Therefore, in terms of mechanical properties, D5 could be considered an alternative to water medium.

Surface structure and cross-section of wool fibre

Wool is a common, naturally occurring polymer with a very complex surface structure (Xue 2015). The SEM pictures of the raw wool fiber and the dyed fiber (Sample 32) are shown in Figure 6a,b. The original undyed wool fiber surface was covered by compact scales with many fragments of scales, as observed in Figure 6a. After dyeing, these fragments were removed, and thick and clear scales appeared in Figure 6b. It is suggested that the morphological surface of the wool was not

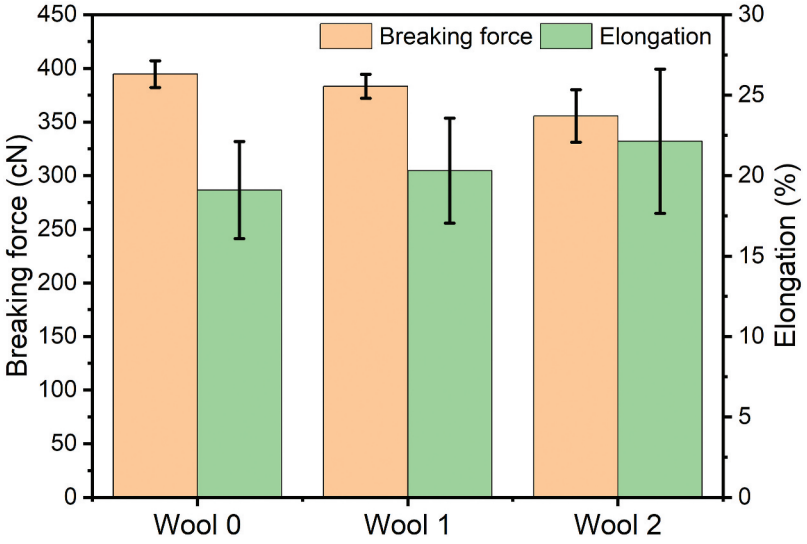

Figure 5. Breaking force and elongation of wool yarns, wool 1: original wool yarn, wool 2: the wool yarns treated in an aqueous at pH 3 and 90°C for 90 min, wool 2: the wool yarn treated by wetting with a 300% water pickup rate at pH and 90°C for 90 min in D5 medium.

compromised during the dyeing procedure, which is consistent with the previous works (Feng et al. 2022; Luo et al. 2022). In Figure 6c,d the microscope images of cross sections of dyed wool fiber (Sample 32) exhibit that after dyeing, the ternary natural dyes (MR:GY:GB = 0.4:0.3:0.3) evenly mixed and distributed in wool fiber interior, since the color shade of cross-section of individual wool fiber is uniform.

Conclusions

Three natural plant colorants, including MR, GY, and GB, are employed to prepare binary dyes' mixture and ternary dyes' mixtures, which mixtures were used to dye wool yarns without any mordants in D5 medium to explore the broadening color shades of wool fibers. With various dye mass ratios in the binary and ternary combinations, the color shades of dyed wool yarns gradually changed. Moreover, very good to excellent colorfastness to washing was achieved with these natural dyes. XRD and TGA analyses show that the morphology and structure of wool fibers remained for the wool fiber dyed with natural dyes in a D5 solvent. Similarly, wool yarns dyed in D5 solvent had a minimal impact on the yarn's mechanical properties. Most importantly, using an organic solvent also significantly reduces the amount of water in dyeing because only 300% o.m.f of water was needed for better dye adsorption. Moreover, SEM images and cross-section analysis indicate that the morphological surface of the wool was not compromised during the dyeing procedure, and dyes were mixed and distributed in the wool fiber interior. Furthermore, A D5-based dyeing method was developed to reduce the use of chemicals and eliminate the need for water. In addition, more than 99% of the used D5 organic solvent can be recovered and reused for multiple dyeing cycles. So far, laboratory-scale trials employing D5 as a dyeing medium indicate that this dyeing process has significant potential for the future. However, in order to use it in an industrial setting, it is essential to carry out large-scale experiments on different types of equipment using diverse combinations of natural dyes that produce varied colors.

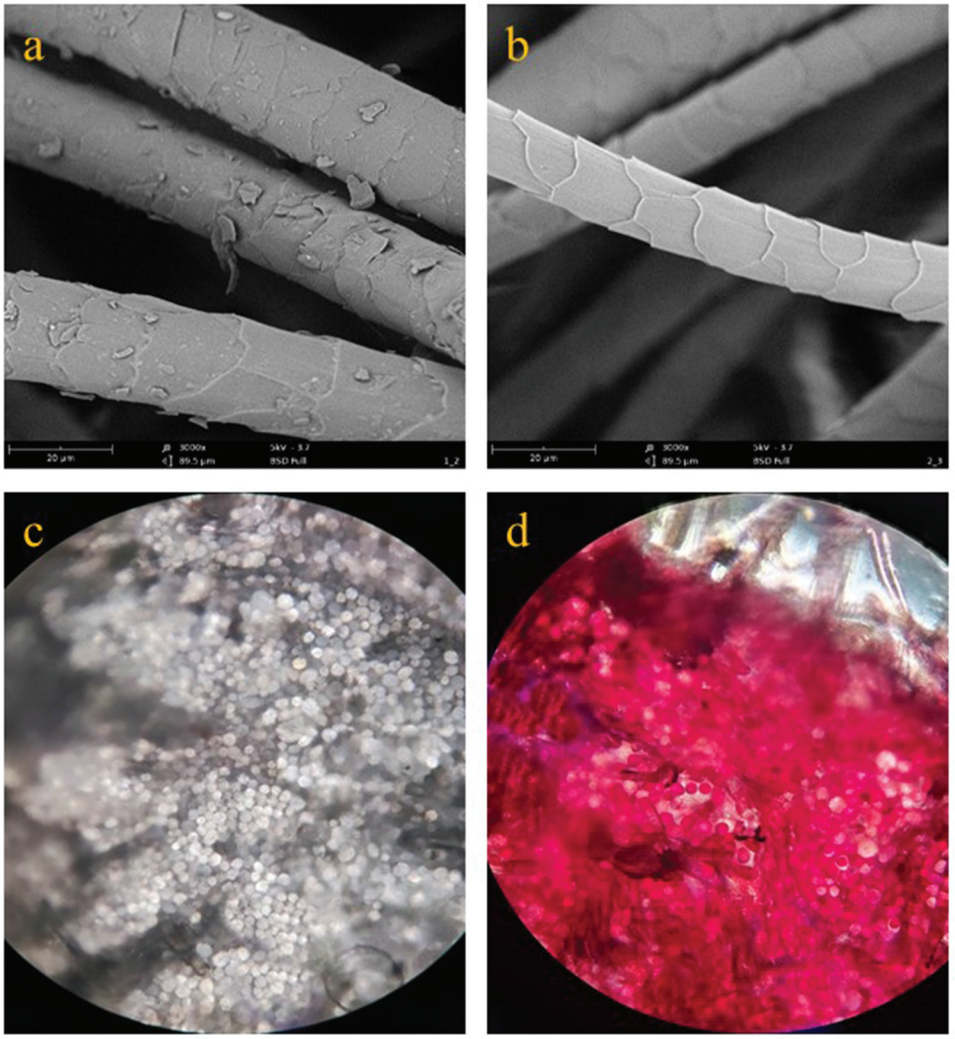

Figure 6. SEM images of the cross-section of (a) original undyed wool fiber and (b) dyed wool fiber, and the microscope images of the cross-section of (c) original wool fiber and (d) dyed wool fiber.

Disclosure statement

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Authors' contributions

Y.C. and S.H.: conceptualization, methodology, writing – original draft; X.H. and T.J.: conducted the experiments, analyzed the data; M.N.E., M.R., M.N.P., M.M.H., and V.N.: formal analysis, writing – review, and editing; S.H., M.M.H., and V.N.: project administration, funding acquisition, and supervision. All authors have read and agreed to the published version of the manuscript.

Highlights

- Coloration of wool yarn in decamethylcyclopentasiloxane (D5) solvent medium using natural dyes.
- Development of combination shade in a color triangle using madder red, gardenia yellow, and gardenia blue.
- The impacts of various dyeing conditions on the K/S value and the rate of fixation are discussed.
- Excellent color stability, good fixation rate, and acceptable mechanical characteristics were achieved.

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