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An attractive path to use of green resources for production of antibacterial and antioxidant wool yarns

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The worldwide agricultural and food industries produce serious environmental pollution problems as well as increased costs of waste disposal. While the agro-food wastes are able to provide valuable resources for the production of bio-materials to promote circularity of the agro-food chain obtaining a better balance between economic, environmental, and social aspects. Today's society is then becoming more and more sensitive to such important issues as resource optimization and ecologically sound waste management strategies for a sustainable future. Bio-finishing of textile materials emerges as an effective solution to management of agro-food wastes and alleviate environmental pollution. In this way, for the first time, a bio-mordant by-product was recovered from the mango seed kernel to replace metallic mordanting agents in the dyeing process of wool with natural dye extracted from the leaves of *S. rebaudiana* through the two conventional and ultrasound-assisted procedures. The influence of the main process parameters on the dyeing efficiency was evaluated and an efficient mathematical model was developed to provide the best dyeing conditions at minimum materials and energy costs. Results revealed that while the ultrasound method was able to improve extraction efficiency, the bio-materials produced in this study could further display good antioxidant activity, antibacterial performance, and enhanced color fastness. Moreover, the elaborated dyeing process reduced the environmental parameters (BOD_5 and COD). The findings foresee developing a new sustainable process for the resource optimization of mango seed kernel and *S. rebaudiana* leaves to decrease the agro-food wastes disposal impacts as well as close he loop in the circular bio economy.

Keywords Bio-art textiles, *S. rebaudiana* leaves, Mango seed kernel, Ultrasound extraction, Antioxidant activity

Nowadays, a huge amount of agro-food wastes has generated to meet the rapidly expanding population's increasing demands, which is destroying our environment. It is well known that the practice of open burning of agro-food wastes produce a lot of harmful chemicals like CH_4, SO_2, N_2O, and smoke, contributing to air pollution seriously. Besides air pollution, inadequate management of agro-food wastes has also been claimed to have several disadvantages, such as the increasing of carbon dioxide emissions, contamination water and soil resources, posing human health risks, and the economic losses[1–3]. As a result, the agro-food wastes sectors are worrying in the space of sustainability, and the future generations are becoming more and more sensitive to such important issues as resource optimization, innovative conversion technologies, and proper management techniques for a sustainable future. In this context, there is an urgent need to explore more green and safer bio-based alternatives. Among the reported strategies to waste management and environmental protection, utilization of agro-food wastes as green resources in the textile bio-finishing process can contribute to the reduction of the waste outflow into the environment, reduced CO_2 emissions, less energy consumption as well as the promotion of the circular bio economy. As endorsed by most of the studies, the agro-food wastes as green resources have more advantages in biodegradability, biocompatibility, fluorescence, UV protection, reducing agent, flame retardant, antimicrobial potency, and even medical values than synthetic ones[4–6]. Therefore, the introduction of agro-food wastes in the textile finishing and dyeing industries is a highly attractive option to reduce dyestuffs-related water pollution and should also be popularized more and more to decrease the enormous environmental pollution of agro-food wastes. On this basis, exploring new potential resources of agro-food wastes as colorants

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and mordants with availability in large quantities is a very promising green chemistry concept. Working in this perspective, agro-industrial wastes and by-products as clean, sustainable, and multifunctional textile dye resources not only could be the ideal option but could decrease the cost of wastewater treatment, eliminate the use of toxic chemical materials from the various textile process, and establish a safe technology for agricultural waste management. Herein, the present work provides a new insight into the sustainable dyeing and finishing processes using *S. rebaudiana* leaf and mango seed kernel extracts for the first time.

Stevia rebaudiana belongs to the family of Asteraceae and it is native to humid areas[7]. Its leaves are the source of non-caloric sweet steviol glycosides and are rich in such important phytochemicals as polyphenolic compounds, trace elements, lipids, amino acids, oligosaccharides, hydroxycinnamic acids (caffeic, chlorogenic, etc.), alkaloids, flavonoids, coumarins, and tannins. Among them, the polyphenol content of *S. rebaudiana* extract has been reported to amount to 2–4% of the dried *S. rebaudiana* leaf[8]. It is well known that its leaf extract contains such major polyphenol components as chlorogenic acids, diosmin, ellagic acid, caffeic acid, chlorogenic acid, pyrogallol, rutin, galuteolin, quercitrin, roseoside, catechin, luteolin, sinapic acid, and trans-ferulic acid[8,9]. *S. rebaudiana* leaves constitute a potential resource of bioactive compounds that might be used as a green dye. Accordingly, attention should be given to evaluate the extract from this bio-mass using innovative technologies and valorize it as a new low-cost bio-dyestuff for use by the textile dyeing industry.

Despite the wonderful benefits of natural colorants, they have issues regarding their fixation and fastness ratings. In this perspective, metal salt mordants are usually used to overcome these drawbacks, but they can end up as pollutants in wastewater streams. Recently, a special interest is growing on the utilization of bio-mordants that are obtained from agro-food wastes. Moreover, the application of these natural resources for the textile dyeing process would help to cope with the challenge of waste disposal. Nevertheless, in line with the global demand for exploring bio-mordants based on agro-food wastes, Mango (*Mangifera Indica)* seed kernel can be an ideal option. Mango belongs to the Anacardiaceae family and its processing industry generates a significant quantity of wastes every year. The mango solid wastes do not decompose quickly which can cause such serious environmental problems as greenhouse gas emissions, vegetation damage, asphyxiation, unpleasant odors, and water pollution[10]. However, mango seed kernels are considered prospective resources of high-value polyphenolic compounds with multifunctional properties. Literature sources indicate that mango seed kernel contains such major bioactive compounds as antioxidant minerals (especially potassium, manganese, copper, zinc, and selenium), phenolic acids (especially gallic acid, ellagic, ferulic, coumaric, protocatechuic, and caffeic acids), and phenolic compounds (especially anthocyanins, kaempferol, homomangiferin, isomangiferin, mangiferin, and quercetin)[11–13]. As a result, it can be considered a highly attractive bio-mordant for value-added product development.

The present study attempts to valorize the application of two agro-food wastes i.e., *S. rebaudiana* leaves and mango seed kernel to impart multifunctional properties to wool yarns for the first time. The effects of independent coloration parameters on process performance were also evaluated. The optimized dyeing conditions were determined via the desirability function approach that captures the overall process. Optimization of the dyeing process is essential for avoiding possible qualitative discrepancies among the colored specimens and minimizing materials and energy costs. It was hypothesized that treating of wool with mango seed kernel may be enhanced the dyeing efficiency due to the formation of strong complexes. Furthermore, fastness properties, surface morphology, antioxidant performance, and the antibacterial properties of the dyed wool bio-yarns as well as environmental parameters were investigated. The findings of this work could be considered as an attractive step towards producing a green process for bio textiles, resource optimization, and agro-food wastes management.

Materials and methods

Materials

Merino wool yarn (160 tex/twofold), procured from the local market in Tehran, Iran, was scoured in a bath of 2 gL^{-1} non-ionic detergent at 40 °C for 25 min before it was rinsed, washed, and dried[14]. *S. rebaudiana* leaves were obtained from Arak, Iran. The leaf thus collected was then washed thoroughly with distilled water to remove impurities before they were air-dried in the shadow to be subsequently ground in an electric grinder to powders of uniform granular size. The ripened mango fruit was obtained from the local market of Semnan, Iran for its potential application as a bio-mordant. The mango seed kernels have been separated from the fruits with sharp and blunt knives, were washed with distilled water, and were then air-dried in the shadow, later they were ground into powders using an electric grinder, finely.

Methods

Dye and bio-mordant extraction

Using 30% methanol in distilled water as the solvent, the colorant was extracted from *S. rebaudiana* leaves by employing conventional and ultrasound-assisted methods. When employing the conventional extraction method, the macerated mixture was boiled for 90 min while in the sonicated method, the glass beaker was exposed under the ultrasonic environment at 55 °C for 25 min. Both mixture extracts were then placed into a shaker incubator at 70 °C for 1 h and strained through the filter paper. Finally, the mixtures feed into a rotary evaporator to remove the solvent and condense the extracts for use in the dyeing process.

For the bio-mordant extraction, the mango seed powder was macerated in the diluted methanol solvent (3:2 v/v) under continuous stirring for 24 h. The macerated mixture was exposed to the ultrasonic environment at 60 °C for 40 min, was then cooled to room temperature, strained through the filter paper, centrifuged for 10 min at room temperature, and used for the bio-mordanting process.

Independent variables	Code	Levels		
		−1 (Lower)	0 (Middle)	+ 1 (Upper)
Dye concentration (% owf)	A	70	110	150
Bio-mordant concentration (% owf)	B	0	15	30
Citric acid concentration (% owf)	C	0	5	10

Table 1. Independent variables, their coded and actual levels, dependent variables, and constant variables used in the CCD method. Dependent variable: color strength. Constant variables: Dyeing temperature (boil), dyeing time (60 min), pH = 5.

Runs	Factor 1 A (Dye)	Factor 2 B (Biomordant)	Factor 3 C (Citric Acid)	Response
1	110	15	5	11.66
2	110	30	5	11.18
3	110	15	0	10.63
4	70	0	10	4.97
5	110	15	10	12.46
6	150	0	10	11.05
7	70	30	0	6.41
8	110	15	5	11.66
9	110	0	5	9.09
10	110	15	5	11.25
11	70	15	5	7.24
12	150	15	5	12.32
13	150	0	0	10.28
14	150	30	10	12.94
15	110	15	5	10.87
16	110	15	5	12.52
17	70	30	10	8.98
18	150	30	0	10.52
19	110	15	5	12.66
20	70	0	0	4.09

Table 2. Design matrix for dyeing with *S. rebaudiana* leaf extract according to CCD with experimental response values.

Quantification of dye extract

Since flavonoids can form stable complexes with metal ions, the total flavonoid content of the dye extract was measured according to Garcia-Mier and Jimenez-Garcia[15]. The flavonoid content was further quantified according to the standard curve of rutin and reported as mg of rutin equivalent per gram of dry sample. The effectiveness of the extraction process in the two sonicated and conventional methods was evaluated based on extraction yield as estimated by mass balance (Eq. 1)[16]:

$$Extraction\,Yeild = \frac{M_{i-}\,M_f}{M_i} \times 100 \tag{1}$$

where M_i and M_f, respectively, are the initial and final dry weight of the colorant measured before and after the extraction process.

Dyeing procedure

Based on study reviews, preliminary experiments were performed to select dyeing variables (namely, dye, bio-mordant, and citric acid concentrations) and their levels with color strength identified as the response variable. The main dyeing factors as independent variables and their levels are presented in Table 1.

The dyeing process was accomplished as per the recommended instructions of experimental central composite design (CCD) with 6 center points (Table 2). A temperature gradient was proposed for the dyeing process to facilitate color adsorption and diffusion. For this purpose, the soaked specimens were loaded into a dye bath at 25 °C and it was then placed into a thermostatic water bath at boiling temperature for 60 min. Finally, the dyed specimens were thoroughly washed with tap water and air-dried.

Color measurement
The chromatic factors of the specimens were evaluated using an X-Rite Sp64 Portable Sphere Spectrophotometer (ASTM E308, ASTM E1331) with an illuminant D 65 and a 10° standard observer. The Kubelka-Munk's equation was employed to calculate color strength at maximum absorbance using (Eq. 2).

$$\left(\frac{K}{S}\right)_{\lambda} = \frac{(1 - R_{\lambda})^2}{2R_{\lambda}} \tag{2}$$

Fastness properties
The colorfastness of the dyed specimens was investigated according to ISO 105-C06 (A1S): 2010 and ISO 105-B02: 2013 methods.

Scanning electron microscope
The surface morphological changes were evaluated using images captured by a scanning electron microscope (Philips XL-30).

Functional finishing
The radical scavenging activity of the pristine specimens and those dyed with the *S. rebaudiana* extract were investigated using a stable α, α- diphenyl- β- picrylhydrazyl (DPPH) free radical scavenging assay in accordance with a previously reported method[17]. Antioxidant activity evaluated as a function of radical scavenging activity was determined as follows:

$$Antioxidant\ activity\ (\%) = \frac{A_{p-}\ A_d}{A_p} \times\ 100 \tag{3}$$

Where A_p *and* A_d, respectively, are the absorbance values of the dark incubated DPPH/methanol solutions measured at 517 nm for the pristine yarns and those dyed under optimized conditions.

The antibacterial activity of the dyed specimens was quantitatively studied according to the AATCC 100–2004 method. *Staphylococcus aureus* ATCC 6538 (a Gram-positive bacterium) was used as a test strain, due to its popularity as an infectious organism.

Environmental parameters
The total polyphenol content was estimated by the Folin-Ciocalteu method according to the calibration curve of gallic acid[9,18]. Here the following equation was used for determining total polyphenols removal:

$$TP_{removal}\,(\%) = \frac{TP_{b-}\ TP_a}{TP_b} \times\ 100 \tag{4}$$

where TP_b and TP_a are the total polyphenols of the dye baths before and after the dyeing process, respectively.

The biological oxygen demand in the dye bathes before and after the dyeing process was tested by a BOD tester (Aqua-BD 600, Germany) and its removal can be calculated from the following equation:

$$BOD_{removal}\,(\%) = \frac{BOD_{b-}\ BOD_a}{BOD_b} \times\ 100 \tag{5}$$

The chemical oxygen demand in the dye baths before and after the dyeing process was estimated by a COD tester (Aqua-AL 100, Germany). Equation (6) expresses the COD removal.

$$COD_{removal}\,(\%) = \frac{COD_{b-}\ COD_a}{COD_b} \times\ 100 \tag{6}$$

Where COD_b and COD_a are the COD of dye baths before and after the dyeing process, respectively.

Statistical analysis
Statistical analysis was carried out using Design Expert V 12.0.3.0. Software. RSM-based central composite design (CCD) was employed to design the trials and study the effects of process parameters on the response. ANOVA test was used to evaluate the obtained experimental findings and determine the coefficient of the established model. The evolution and fitting quality of the model were done by the regression coefficients. A confidence level of 95% was chosen to decide on the model terms and their interactions.

Results and discussion

Extraction and quantification of the bio-phenolic compounds

There are, generally, a number of extraction methods for natural dyes, such as microwave-assisted, ultrasound-assisted, pressurized liquid, supercritical fluids, and conventional extraction, available for extracting color and bioactive compounds from plant dye sources. Ultrasound-assisted extraction (UAE) is a green and environmentally friendly method for extracting bioactive compounds from natural dyes. It offers such advantages as energy saving, fast extraction, low extraction costs, low process temperatures, and reducing the amount of water needed in the process[19,20]. This technique is essentially based on the generation of microbubbles that surround particles

Extraction Method	Extraction Conditions		Total Flavonoids Content (Mean ± SD)	Extraction Yeild (%)
	T(°C)	t(min)		
Convention	98	90	0.172 ± 0.012	16
Sonication	55	25	0.253 ± 0.008	29

Table 3. Effect of dye extraction method on the extraction performance.

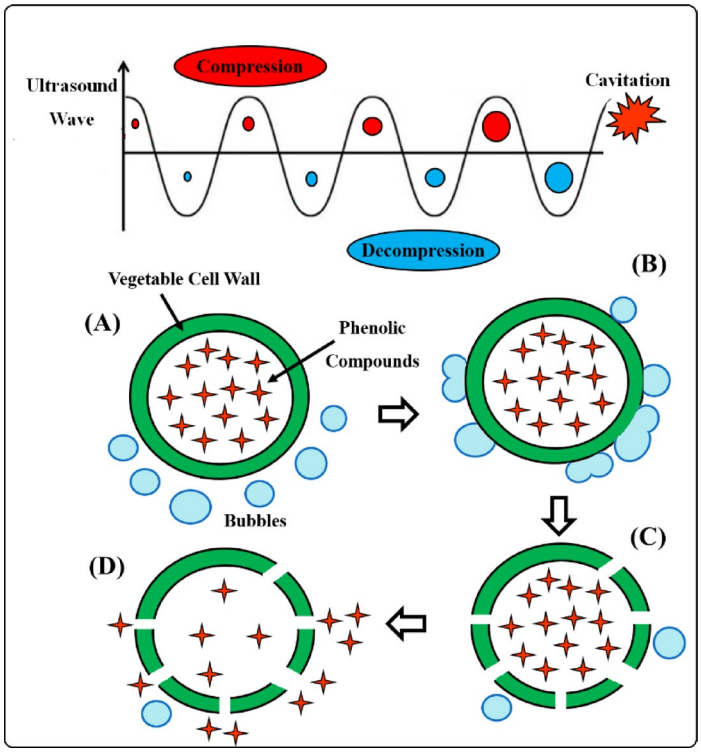

Fig. 1. Ultrasound-assisted extraction of natural dyes (**A**) Vegetable cell wall and bubbles, (**B**) Bubble collapse, (**C**) Cell wall disruption, and (**D**) Solvent diffusion and phenolic compounds release.

in the liquid phase and grow and collapse under high pressure to generate shock waves and microjets near the solid surface. These processes ultimately promote mass and heat transfer inside the specimen[20].

In the present work, conventional and ultrasound-assisted methods were employed to extract the bioactive compounds from *S. rebaudiana* leaves. The two extraction methods were investigated and compared based on estimations of total flavonoid content and extraction efficiency values obtained for each method. The results for both approaches are reported in Table 3.

As seen in Table 3, a total flavonoid content of 0.253 mg equivalent of rutin/g of dry sample and an extraction efficiency of 29% was recorded for the ultrasound extraction method, while the same parameters were measured at 0.172 mg equivalent of rutin/g of dry sample and 16% respectively, for the conventional method. Clearly, the sonicated method proved more effective in extracting the bioactive compounds from *S. rebaudiana* leaves. This can be explained by the fact that the bigger color particles break up as a result of the acoustic cavitation process applied and that the following effective solid-liquid interaction under the mass transfer kinetics leads to a higher mass (color) being extracted into the solvent, whereby the extraction performance is improved. It has also been documented in the literature that numerous extraction mechanisms such as fragmentation, erosion, sonoporation, shear force, and swelling index in the cellular matrix of the sample can be obtained as a result of collapsing cavitation bubbles and ultrasound wave propagation (Fig. 1)[21,22].

Another important point to note in Table 3 is that the advantages of the sonicated method in addition to its enhanced performance are the significantly reduced extraction time, temperature, and energy used. This method may, therefore, be recommended for dye extraction due to the associated time and energy efficiency that make it an eco-friendly and sufficient option for textile dyeing applications[20–22]. Flavonoids such as quercetin, luteolin, apigenin-7-O-glucoside, centaureidin, eupatorin, nepetin, hispidulin, pectolinaringenin, chrysosplenetin, eupatilin, apigetrin, methoxylated flavonoid casticin, and glycoside flavonoids have been reported to be present in *S. rebaudiana* species[23].

Factors	Sum of Squares	Degrees of Freedom	Mean Square	F- Value	*p*- Value
A	64.62	1	64.62	238.71	< 0.0001
A^2	8.16	1	8.16	30.15	0.0003
B	11.13	1	11.13	41.12	< 0.0001
B^2	5.14	1	5.14	19.00	0.0014
C	7.17	1	7.17	26.50	0.0004
C^2	0.0049	1	0.0049	0.0182	0.8955
AB	2.20	1	2.20	8.15	0.0171
AC	0.0084	1	0.0084	0.0312	0.8633
BC	1.39	1	1.39	5.15	0.0466

Table 4. ANOVA results of the effects of process variables on response value.

Factor	Coefficient Estimate	df	Standard Error	95% CI Low	95% CI High	VIF
Intercept	11.67	1	0.1597	11.32	12.02	
A: Dye	2.54	1	0.1506	2.21	2.87	1.0000
B: Biomordant	1.06	1	0.1506	0.7270	1.38	1.0000
C: Citric acid	0.8470	1	0.1506	0.5190	1.18	1.0000
AB	−0.5250	1	0.1683	−0.8918	−0.1582	1.0000
BC	0.4175	1	0.1683	0.0507	0.7843	1.0000
A^2	−1.71	1	0.2662	−2.29	−1.13	1.56
B^2	−1.35	1	0.2662	−1.93	−0.7720	1.56

Table 5. The coefficient estimates and their standard errors in the predicted quadratic reduced model.

Source	Sum of Squares	Degrees of Freedom	Mean Square	F-Value	Probability > F
Model	124.07	7	17.72	78.19	< 0.0001
Residual	2.72	12	0.2267	-	-
Lack of fit	0.2611	7	0.0373	0.0758	0.9981
Pure error	2.46	5	0.4918	-	-
Total	126.79	19	-	-	-

Table 6. ANOVA for the reduced quadratic model for wool dyeing with *S. rebaudiana* leaf extract.

S. rebaudiana extract dyeing process optimization

It is well established that dyeing conditions play important roles in dyeing performance. Hence, these conditions should be efficiently optimized to avoid qualitative discrepancies among the colored specimens and to minimize the number of experimental trials and the associated costs. For this purpose, the Response Surface Methodology (RSM) based on CCD was employed to evaluate the effects of dyeing variables and their interactions on the response value, to develop an efficient model of the process, and to optimize the dyeing conditions. The experimental response values were fitted to various models and the quadratic model equation was chosen to describe the dyeing process (Sequential p-value < 0.0001, lack of Fit p-value = 0.9876, adjusted $R^2 = 0.9594$, and predicted $R^2 = 0.9665$).

Table 4 shows the results of ANOVA for evaluating the effects of process variables on the response value.

All the linear terms, the quadratic effects of A^2 and B^2, and the interaction terms of AB and BC are significant as evidenced by their high Fisher's F-value and relevant probability values of less than 0.05. The predicted quadratic reduced model in terms of coded variables and their standard errors May be expressed by Table 5 and Eq. (7):

$$K/S = 11.67 + 2.54\,A + 1.06\,B + 0.8470\,C - 0.5250AB + 0.4175\,BC - 1.71\,A^2 - 1.35\,B^2 \quad (7)$$

where A, B, and C represent dye, bio-mordant, and citric acid concentrations, respectively.

The ANOVA results for the fitted response surface quadratic equation reported in Table 6 verify the adequacy of Eq. (7).

The statistically significant F-value of 78.19 and p-value of less than 0.0001 obtained for the proposed model indicate its validity. Also, the model's lack of fit F-value (0.0758) was found to be much lower than its F-value (78.19), again indicating its high validity. The adequacy of the model may be supported by determination coefficients (R^2, adjusted determination coefficients (R^2adj), and predicted determination coefficients (R^2pred).

In this study, a high R-squared value of 0.9785 indicates that the model was able to explain 97.85% of the total variations. The adjusted R-squared value (0.9660) is also in reasonable agreement with the predicted R-squared value (0.9695), endorsing the effects of dying process factors on the response variable.

Response surface (3D) and counter plots were employed to evaluate the effects of citric acid concentration, dye concentration, and bio-mordant concentration on color strength. The counterplots were obtained by considering two independent parameters while all the others were kept constant. Figure 2 shows the contour and 3-D plots for evaluating the effects of process variables on the response value.

It is seen that, compared to citric acid concentration, dye concentration and bio-mordant concentration have significant effects on the dye uptake of the colored specimens (AB, BC). Indeed, color strength was rapidly

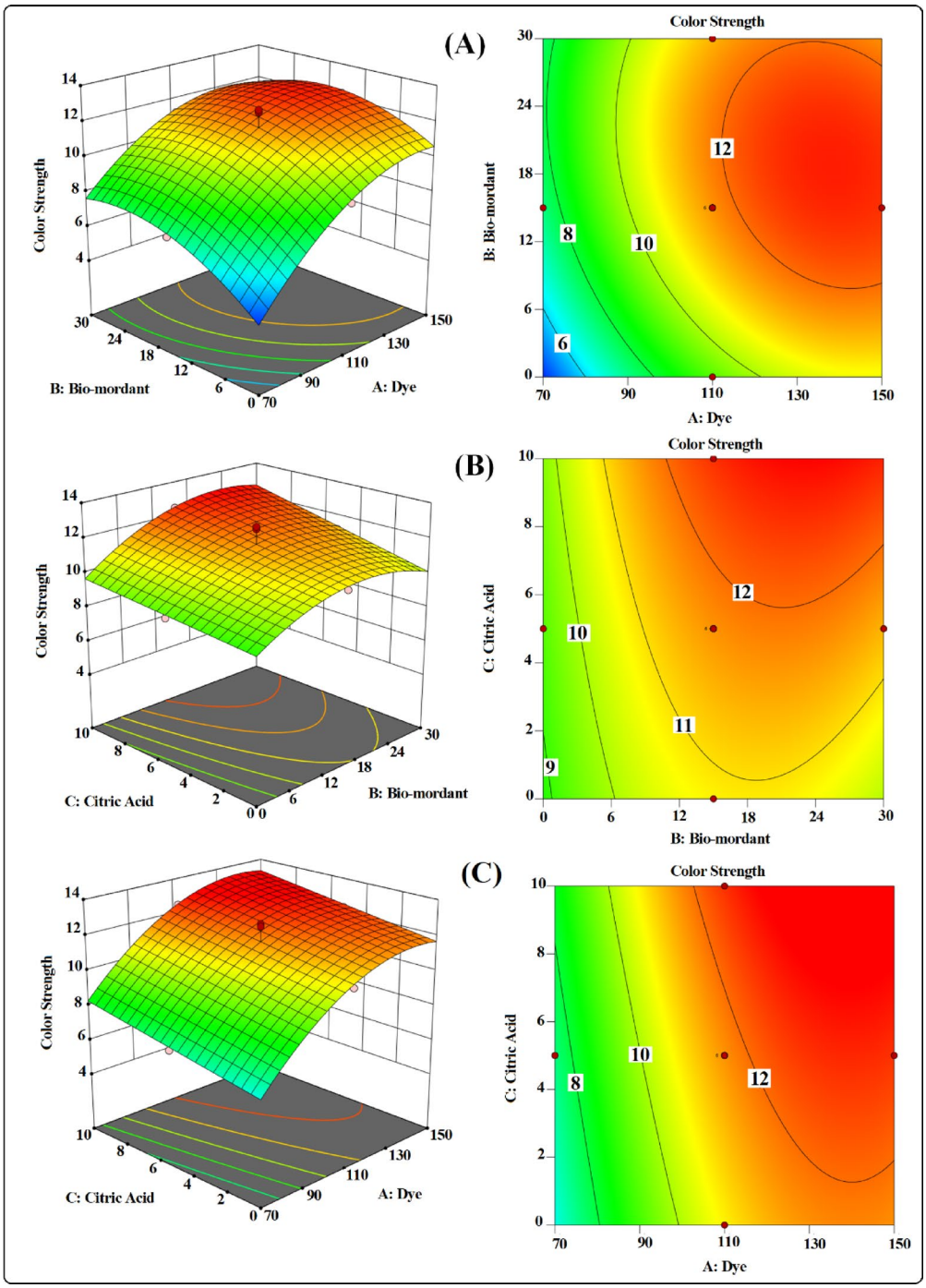

Fig. 2. Counter and 3-D plots showing the combined effects of (**A**) dye concentration and bio-mordant concentration, (**B**) citric acid concentration and bio-mordant concentration, and (**C**) citric acid concentration and dye concentration on color strength.

enhanced with increasing dye concentration by up to about 130% o.w.f whilst it slightly increased with excessively high dye concentrations.

It is also clear that the color strength increases as bio-mordant and citric acid concentrations increase. As mentioned earlier, mango seed kernel contains major polyphenol components[11–13]. They have a strong affinity to woolen polypeptide chains and are able to form stable complexes between wool macromolecules and the color molecules, which subsequently enhances the dye uptake of the specimens. This indicates that mango seed kernel could be considered as a highly attractive bio-mordant for value-added product development, which is obviously a superb advantage.

The improved coloration efficiency with increasing citric acid concentration could also be explained by two events. Citric acid is a cost-effective zero formaldehyde-based tri-carboxylic acid with one hydroxyl group and three carboxylic groups in one molecule. Wool peptide chains have numerous amino groups in their structure which are capable of forming amide linkages with citric acid. In addition, citric acid has the properties to form ester bonds with hydroxyl groups of wool specimens[24]. As a result, citric acid is expected to form cross-linkages with color molecules and the functional groups of wool yarns, and thus could be regarded as an organic mordant to enhance dyeing performance. The other event is the presence of numerous carbonyl and hydroxyl groups in the *S. rebaudiana* extract[8,9] which makes it an anionic dye. Under acidic pH conditions, the number of protonated amino groups of wool specimens is increased, which can be more interesting in the anionic sites of dye molecules: hence higher coloristic intensity is obtained.

Optimization of the process conditions was accomplished using the numerical optimization function of the designed software. The process variables (namely, dye, bio-mordant, and citric acid concentrations) were set to the proposed range whereas color strength was set at its maximum. The optimized dyeing conditions obtained for wool yarn dyeing with the solution of *S. rebaudiana* leaf extract are shown in Fig. 3.

Based on the results obtained, the best dyeing parameters were determined to include dye, bio-mordant, and citric acid concentrations of 128% o.w.f, 24% o.w.f, and 8.5% o.w.f, respectively.

The color space position of the dyed yarns with *S. rebaudiana* leaf extract at various dying conditions are shown in Fig. 4(A).

It indicates that all dyed samples are placed in the yellow–red and yellow-green regions of CIE L*a*b* color space. When the b* values were evaluated, it could be observed that the colors are all yellow but with different green-red shades. The effects of bio-mordant and citric acid concentrations on the a* value was also investigated (Figs. 4(B) and 4(C)). Regardless of bio-mordant and citric acid are applied, solely dyeing wool yarns with the extract of *S. rebaudiana* represents a green-yellow shade. Moreover, increasing the bio-mordant and citric acid concentrations lead to a considerable increase in the a* values and the dyed samples are located in the yellow-red zone of the CIE space.

Surface functionalities analysis of the bio-yarns produced

To study the surface functionalities of wool bio-yarns, the pristine samples and those dyed with *S. rebaudiana* under optimized dyeing conditions were analyzed by FTIR spectroscopy. The results are shown in Fig. 5; Table 7.

The structure of wool yarn as a protein material has C = O, OH, and NH functional groups. It is evident that the main characteristic peaks are found in certain regions of wool wavenumbers. The pristine wool yarns show characteristic bands at 3549 cm^{-1} and 2925 cm^{-1} for the -OH stretch, -NH stretch, and -CH stretching vibrations of wool protein[25,26]. The decreasing shifts at 3515 cm^{-1} and 2922 cm^{-1} observed in the dyed specimen suggested that both inter- and intra-molecular hydrogen bonds may have developed between the functional groups of wool yarns and dye molecules. A strong peak at around 1700 cm^{-1} was observed in the pristine sample representing the asymmetric coupling of –C = O stretching vibrations. Also, a broadband occurs at 1720 cm^{-1} with a shoulder on the low wavenumber side at 1640 cm^{-1} for the dyed specimen. This shoulder may be related to the C = O stretching vibration of the aromatic group which represents the presence of flavonoids in the dyed specimens[27]. The result can be supported by the strong peaks at 1500 cm^{-1}, 1445 cm^{-1}, and 1228 cm^{-1} in the dyed wool sample, which were ascribed to the -C-C- aromatic stretching, the -CH out-of-plane bending vibration, and C-O-C stretching vibrations, respectively[27,28].

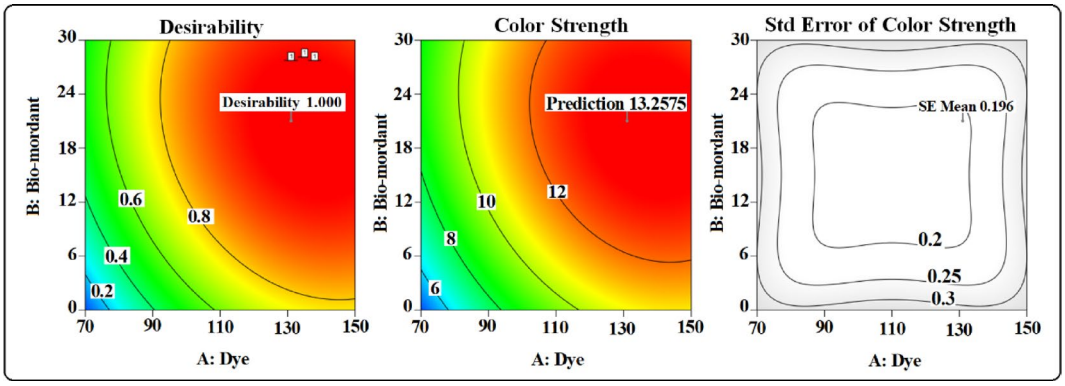

Fig. 3. Optimized conditions of wool dyeing procedure with the solution of *S. rebaudiana* leaf extract.

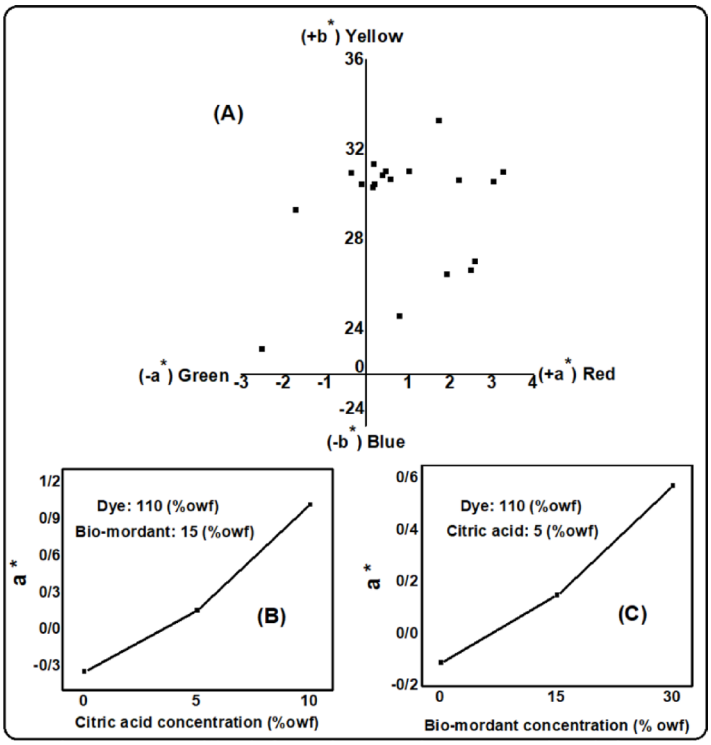

Fig. 4. (**A**) Graphical representations of the color coordinates a* and b*, the effects of (**B**) citric acid concentration, and (**C**) bio-mordant concentration on a* value.

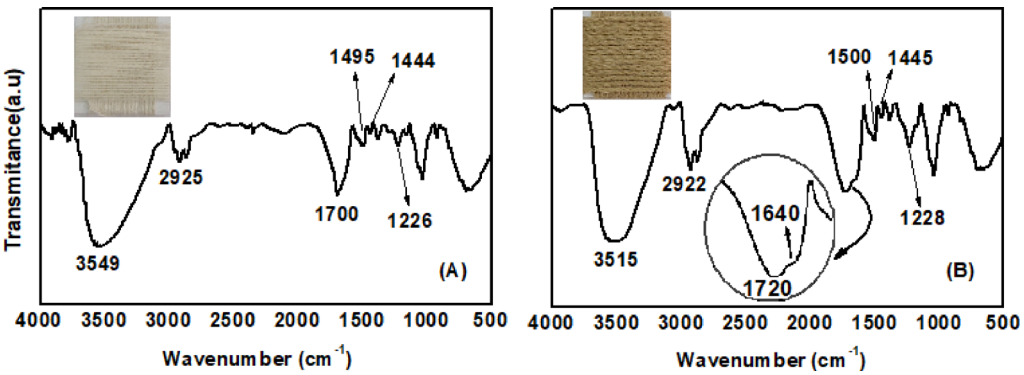

Fig. 5. FTIR spectra of (**A**) pristine wool yarns and (**B**) those dyed under optimized dyeing conditions.

Types of vibrations	Wavenumbers (cm^{-1})	
	Pristine yarns	Dyed yarns
-OH stretching and -NH stretching	3549	3515
-CH stretching	2925	2922
Asymmetric coupling of –C = O stretching	1700	1720 (with a shoulder at 1640)
-C-C aromatic stretching	1495(medium)	1500 (strong)
-CH out-of-plane bending	1444(weak)	1445 (medium)
-C-O-C stretching	1226(medium)	1228(strong)

Table 7. FTIR spectra of the pristine and dyed wool yarns.

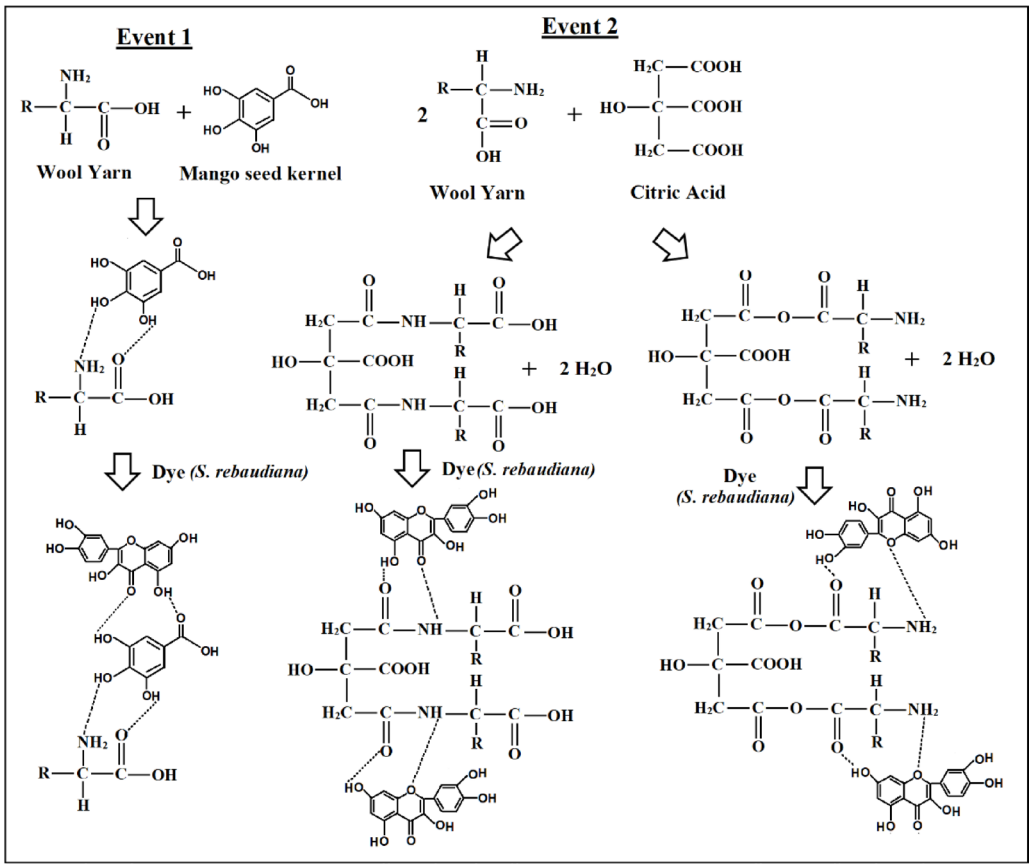

Fig. 6. Schematic mechanism of dyeing wool yarn by mango seed kernel and *S. rebaudiana* extracts.

Specimens	Light Fastness	Wash Fastness			Antioxidant Performance(%) (Mean ± SD)	Antibacterial Activity(%) (Mean ± SD)
		S_c	S_w	C_c		
Pristine	-	-	-	-	5.73 ± 1.13	-
Dyed	5	5	5	4–5	90.47 ± 0.96	84 ± 2

Table 8. Colorfastness properties, antioxidant performance, and antibacterial activity of the bio-yarns produced under optimized dyeing conditions. C_c: Color change S_c: color staining on adjacent cotton sample. S_w: color staining on adjacent wool sample.

Based on the FTIR results, it may, thus, be claimed that the mango seed kernel and *S. rebaudiana* functional groups were chemically absorbed on the wool peptide chains. The proposed mechanism of dyeing wool yarn using bio-materials derived from mango seed kernel and *S. rebaudiana* extracts is shown in Fig. 6.

Two events may take place as a result of dyeing wool yarns with the *S. rebaudiana* extract in the presence of bio-mordant and citric acid. First, mango seed kernel as a bio-mordant is able to form strong complexes with amino and carboxylic groups of wool and phenolic groups of *S. rebaudiana* molecules via hydrogen bonds. As a result, it is capable of producing ternary complexes on one site with the dye molecules and the other site with the peptide chains, as seen in Event 1 (Fig. 6). Second, as already mentioned above, Woolen peptide chains contain numerous amino and hydroxyl functional groups in their structure which are capable of forming amide linkages and ester bonds with citric acid[24]. The stable complexes may, then, be obtained through hydrogen bonds between functional groups present in dye molecules of *S. rebaudiana* extract and the yarn modified with citric acid as well as hydrophobic interactions between their non-polar groups, chains, as seen in Event 2 (Fig. 6). The presence of strong complexes in the dyed specimens can be confirmed by the good colorfastness properties.

Assessment of colorfastness ratings and SEM photographs

The colorfastness properties recorded for the bio-materials produced in the present work are reported in Table 8.

Clearly, a far better light fastness (5) was obtained, which might be ascribed to a free radical scavenging influence of flavonoids in the dye molecules extracted from *S. rebaudiana* leaf[29]. Accordingly, the electron density in the chromophore can be decreased, and then acceptable light fastness is obtained. In addition,

the dyed specimens show very good color change and staining values of 4–5 and 5, respectively. This may be explained by the formation of great and strong complexes between peptide chains, bio-mordant, citric acid, and dye molecules through hydrogen bonds and hydrophobic interactions (Fig. 6).

Scanning electron microscopy (SEM) images were used to confirm the successful wool dyeing process with the *S. rebaudiana* leaf extract (Fig. 7).

It is clearly seen that the surface scales of the plain yarns are relatively compact, smooth, and ordered. The smoothness of the surface, however, decreased as a result of the dyeing process, which might be ascribed to the presence of non-planar particles.

Functional finishing of the bio-yarns produced

The antioxidant activity and antibacterial performance of the bio-materials produced were evaluated. Table 8 presents the antioxidant scavenging activities of pristine wool yarns and those dyed with *S. rebaudiana* extract under optimized dyeing conditions. It is clear that the dyed sample has a higher value of antioxidant activity than does the pristine specimen. Such bio-finishing could be attributed to the presence of phytochemicals, especially, flavonoids and high total polyphenolic contents in mango seed kernel and *S. rebaudiana* extracts[11–13] and their strong scavengers of free radicals[13,30].

Textile materials, especially natural ones, are good media for multiplication and microbial growth. This growth not only creates numerous health-related problems for the wearer but can produce a range of unwanted effects on the textile itself. Nowadays, the potential use of bio-components extracted from agricultural by-products as antibacterial agents for preventing bacterial infections is a major scientific research topic due to environmental aspects. As a result, it is worthwhile to investigate the possible application of the mango seed kernel and *S. rebaudiana* extracts in developing protective textiles against common microorganisms. Along these lines, the antibacterial activity of the bio-materials produced in this study was evaluated in terms of percentage reduction of bacteria, and the results are presented in Table 8.

It is interesting to note that the plain wool specimen exhibits no antibacterial performance; the one dyed with the *S. rebaudiana* extract, however, showed a good antibacterial performance. This might be because of the presence of such bioactive ingredients as tannins, phenols, and flavonoids in the mango seed kernel and *S. rebaudiana* extracts[8,11–13,31]. It is well established that polyphenols are able to act against bacterial/microbial metabolism by employing several mechanisms[32–34]. Furthermore, it is well known that citric acid has both bacteriostatic and bactericidal effects against various pathogenic strains and is widely used as an antibacterial agent[35,36]. These results suggest that the combined effect of the phenolic compounds present in the mango seed kernel and *S. rebaudiana* extracts and citric acid might provide antibacterial activity in the dyed specimens.

Effect of the elaborated dyeing process on the environmental parameters

The assessment of environmental characteristics, viz., total polyphenols, chemical oxygen demand (COD), and biological oxygen demand (BOD) is critical for achieving economic, eco-friendly, and clean production. Working in this perspective, the effect of the elaborated dyeing process on the effluent parameters was evaluated and the results are given in Table 9.

For the conditions used in our experiments, the total polyphenols removal, COD removal, and BOD_5 removal of the elaborated dyeing process were found to be 33.33%, 17.40%, and 11.83%, respectively. It may, therefore, be concluded that the elaborated dyeing process not only could be absorbed polyphenols as the main coloring materials but could significantly decrease the impact on the environment and establish a green process for achieving environmentally-friendly clean production. Furthermore, it was ensured because of the process biodegradability ratio lower than 3 what means that the elaborated dyeing process used in this study is biodegradable.

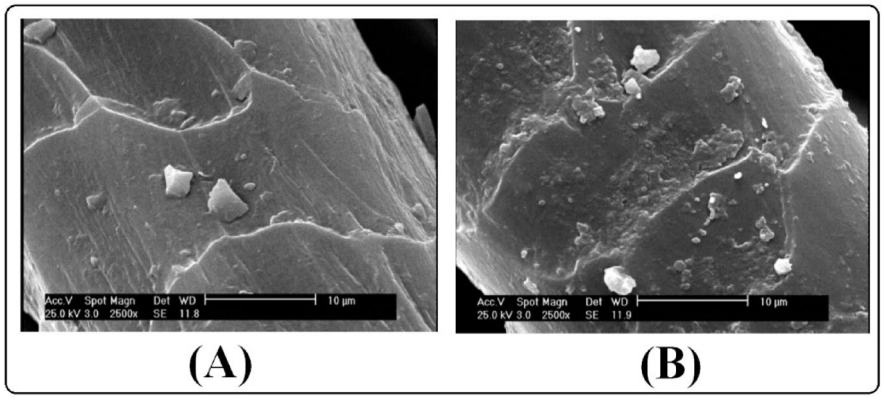

Fig. 7. SEM images of the wool specimens (**A**) pristine yarns (**B**) dyed specimens under optimized dyeing conditions.

Environmental parameters	Before dyeing (Mean ± SD)	After dyeing (Mean ± SD)
Total polyphenols (gL^{-1})	1.26 ± 0.11	0.84 ± 0.08
COD (gL^{-1})	14.31 ± 0.60	11.82 ± 0.41
BOD_5 (gL^{-1})	11.92 ± 0.17	10.51 ± 0.20
COD/BOD_5	1.20	1.12
Polyphenol removal (%)	33.33	
COD removal (%)	17.40	
BOD_5 removal (%)	11.83	

Table 9. Environmental analysis of the elaborated dyeing process.

Conclusions and future perceptions

Decreasing the hazardous waste-related impacts poses an environmental challenge around the world. In this context, one main approach is to explore more sustainable and safer bio-based conservation technologies. This work valorized two agro-food wastes in the wool dyeing process for the first time. Mango seed kernel (MSK) as a bio-mordant was used in conjunction with *S. rebaudiana* leaf extract for sustainably wool dyeing and bio-finishing process. The response surface method was employed to design experiments, investigate the effects of coloration factors, and optimize the dyeing process. The results of extraction yield and total flavonoid contents confirmed the eco-friendly advantages of UAE in terms of extraction time, temperature, and efficiency. Based on the ANOVA results, all the linear terms, the quadratic effects of bio-mordant and dye concentrations, and the interaction terms of biomordant-dye concentrations and biomordant-citric acid concentrations were significant. Results also revealed that mango seed kernel and *S. rebaudiana* extracts used as new resources in wool dyeing could further display a good antioxidant property and antibacterial performance against Gram-positive bacterium. The elaborated dyeing process reduced the pollutant load of the *S. rebaudiana* extract, which is of critical to cleaner production. Moreover, the use of mango seed kernel in the dyeing procedure provides two advantages: one, by eliminating hazardous mordants from the dyeing procedure, the adverse metallic mordant-related impacts will be reduced. Secondly, the dyeing performance, bio-functional finishing activity, colorfastness, color shades, and hues of wool yarns will be improved. Finally, to the advantage of identifying new waste bio-materials, the results of the present work might be considered as a main step towards producing green medical and healthcare textiles and sustainable agro-food wastes management.

Data availability

All data generated or analyzed during this study are included in this article. The data that support the findings of this study are available from the corresponding author upon reasonable request.

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References

1. He, Q. et al. Preparation of wood-based hydrogel membranes for efficient purification of complex wastewater using a reconstitution strategy. *Front. Environ. Sci. Eng.* **18**, 84 (2024).
2. Atinkut, H. B. et al. Cognition of agriculture waste and payments for a circular agriculture model in central China. *Sci. Rep.* **10**, 10826 (2020).
3. Phiri, R., Rangappa, S. M. & Siengchin, S. Agro-waste for renewable and sustainable green production: A review. *J. Clean. Prod.* **434**, 139989 (2024).
4. Hossain, M. A. UV–Visible–NIR camouflage textiles with natural plant based natural dyes on natural fibre against woodland combat background for defence protection. *Sci. Rep.* **13**, 5021 (2023).
5. Habib, N. et al. Green extraction and application of yellow natural Curcumin colorant from Curcuma aromatica rhizomes for silk dyeing. *Sci. Rep.* **14**, 13032 (2024).
6. Shahmoradi Ghaheh, F., Razbin, M., Tehrani, M., Zolfipour Aghdam Vayghan, L. & Sadrjahani, M. Modeling and optimization of dyeing process of polyamide 6 and woolen fabrics with plum-tree leaves using artificial intelligence. *Sci. Rep.* **14**, 15067 (2024).
7. Nikraftar, S., Ebrahimzadegan, R., Majdi, M. & Mirzaghaderi, G. Genome-wide analysis of the C2H2-ZFP gene family in *Stevia rebaudiana* reveals involvement in abiotic stress response. *Sci. Rep.* **14**, 6164 (2024).
8. Myint, K. Z. et al. *Rebaudiana* polyphenols, their antimicrobial and Anti-Inflammatory properties, and inhibitory effect on digestive enzymes. *Molecules* **28** (22), 7572 (2023).
9. Garcia-Mier, L. et al. Polyphenol Content and Antioxidant Activity of S. rebaudiana and Peppermint as a Result of Organic and Conventional Fertilization. *J. Food Qual.* 6620446 (2021). (2021).
10. Gupta, P., Ray, J., Aggarwal, B. K. & Goyal, P. Food processing residue analysis and its functional components as related to human health: recent developments. *Austin J. Nutr. Food Sci.* **3** (3), 1068 (2015).
11. Lebaka, V. R., Wee, Y. J., Ye, W. & Korivi, M. Nutritional composition and bioactive compounds in three different parts of Mango fruit. *Int. J. Environ. Res. Public. Health.* **18** (2), 741 (2021).
12. Mutua, J. K., Imathiu, S. & Owino, W. Evaluation of the proximate composition, antioxidant potential, and antimicrobial activity of Mango seed kernel extracts. *Food Sci. Nutr.* **5** (2), 349–357 (2017).
13. Zahoor, S., Anwar, F., Qadir, R., Soufan, W. & Sakran, M. Physicochemical attributes and antioxidant potential of kernel oils from selected Mango varieties. *ACS Omega*. **8** (25), 22613–22622 (2023).
14. Baseri, S. Agricultural crop of Scrophularia striata as a new dye for ecofriendly dyeing and bioactive finishing of handwoven piles. *Sustainable Chem. Pharm.* **33**, 101088 (2023).
15. Garcia-Mier, L. et al. Elicitor mixtures significantly increase bioactive compounds, antioxidant activity, and quality parameters in sweet bell pepper. *J. Chem.* 269296 (2015). (2015)(3).

16. Tomasi, I. T., Santos, S. C. R., Boaventura, R. A. R. & Botelho, C. M. Optimization of microwave-assisted extraction of phenolic compounds from chestnut processing waste using response surface methodology. *J. Clean. Prod.* **395**, 136452 (2023).
17. Rahman, M. M., Koh, J. & Hong, K. H. Sustainable Chitosan biomordant dyeing and functionalization of cotton fabrics using pomegranate rind and onion Peel extracts. *J. Nat. Fibers*. **21**, 2290856 (2024).
18. Singleton, V. L. & Rossi, J. A. Colorimetry of total phenolics with Phosphomolybdic-Phosphotungstic acid reagents. *Am. J. Enol. Vitic*. **16**, 144–158 (1965).
19. Ismal, O. E., Yildmm, L. & Ozdogan, E. Use of almond shell extracts plus biomordants as effective textile dye. *J. Clean. Prod.* **70**, 61–67 (2014).
20. Sivakumar, V., Verma, V. R., Rao, P. G. & Swaminathan, G. Studies on the use of power ultrasound in solid-liquid Myrobalan extraction process. *J. Clean. Prod.* **15** (18), 1813–1818 (2007).
21. Linares, G. & Rojas, M. L. Ultrasound-Assisted extraction of natural pigments from food processing By-Products: A review. *Front. Nutr.* **9**, 891462 (2022).
22. Medina-Torres, N., Ayora-Talavera, T., Espinosa-Andrews, H. & Sánchez-Contreras, A. Pacheco, N. Ultrasound assisted extraction for the recovery of phenolic compounds from vegetable sources. *Agronomy* **7** (3), 47 (2017).
23. Borgo, J., Laurella, L. C., Martini, F. & Catalán, C. A. N. Sülsen, V. P. *S. rebaudiana* genus: phytochemistry and biological activities update. *Molecules* **26**, 2733 (2021).
24. Mohsin, M. et al. Performance enhancement of wool fabric with environmentally-friendly bio-cross-linker. *J. Clean. Prod.* **68**, 130–134 (2014).
25. Kondo, T., Sawateri, C., Manley, R. S. J. & Gray, D. G. Characterization of hydrogen bonding in cellulose-synthetic polymer blend systems with regioselectively substituted Methylcellulose. *Macromolecules* **27**, 210–215 (1994).
26. Pavia, D., Lampman, G. M. & Kriz, G. S. Introduction to Spectroscopy, Second ed., Washington (1996).
27. Borah, S. et al. Sustainable dyeing of mulberry silk fabric using extracts of green tea (Camellia sinensis): extraction, mordanting, dyed silk fabric properties and silk-dye interaction mechanism. *Ind. Crops Prod.* **205**, 117517 (2023).
28. Liman, M. L. R., Islam, M. T., Repon, M. R., Hossain, M. M. & Sarker, P. Comparative dyeing behavior and UV protective characteristics of cotton fabric treated with polyphenols enriched banana and watermelon Biowaste. *Sustainable Chem. Pharm.* **21**, 100417 (2021).
29. Ghouila, H., Meksi, N., Haddar, W., Mhenni, M. F. & Jannet, H. B. Extraction, identification and dyeing studies of isosalipurposide, a natural chalcone dye from Acacia Cyanophylla flowers on wool. *Ind. Crops Prod.* **35** (1), 31–36 (2012).
30. Yang, B. R. & Liu, P. Z. Composition and health effects of phenolic compounds in hawthorn (*Crataegus spp.*) of different origins. *J. Sci. Food Agric.* **92** (8), 1578–1590 (2012).
31. Myint, K. et al. Polyphenols from *S. rebaudiana rebaudiana (Bertoni)* leaves and their functional properties. *Food Sci.* **85**, 240–248 (2020).
32. Farha, A. K. et al. Tannins as an alternative to antibiotics. *Food Biosci.* **38**, 100751 (2020).
33. Villanueva, X. et al. Effect of chemical modifications of tannins on their antimicrobial and antibiofilm effect against Gram-negative and Gram-positive bacteria. *Front. Microbiol.* **13**, 1–15 (2023).
34. Zhou, H., Li, Q., Zhang, Z., Wang, X. & Niu, H. Recent advances in superhydrophobic and antibacterial Cellulose-Based fibers and fabrics: Bio-inspiration, strategies, and applications. *Adv. Fiber Mater.* **5**, 1555–1591 (2023).
35. El-Fawal, G. Preparation, characterization and antibacterial activity of biodegradable films prepared from Carrageenan. *J. Food Sci. Technol.* **51**, –2234 (2014).
36. Raja, A. S. M. & Thilagavath, G. A. Pre-treatment process for natural dyeing of wool to impart durable antimicrobial efficacy. *Text. Color. Finish.* **23**, 69–75 (2011).

Author contributions

Somayeh Baseri: Writing – original draft, Writing – review & editing, Visualization, alidation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declarations

Competing interests

The authors declare no competing interests.

Additional information

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