Color Evaluation of Wool Fabric Dyed With *Rhizoma coptidis* Extract

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ABSTRACT: Berberine is the only cationic colorant of natural plant dyes, which lies in the roots of *Rhizoma coptidis* and stems of phellodendron. In this study, wool fabric was dyed with the extracts of *R. coptidis*. Color evaluation was characterized with ΔE, L^*, a^*, b^*, c^*, H^0, K/S. Effects of mordant, extraction concentration, pH value of dye bath, and treatment temperature on color values were studied. Results indicated that wool fabrics dyed with mordant, or at higher temperature, or in alkali solution possessed deeper shades and darker colors. And the wool fabric showed good antibacterial property after dyeing with *R. coptidis* extracts.
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Key words: *Rhizoma coptidis*; berberine; wool fabric; color evaluation

INTRODUCTION

Until the 20th century, natural dyes were the only source of color available. Therefore, they were widely used and traded.[1,2] Unfortunately, the natural dyes are rarely used in modern dyeing, except by specialist companies and craft dyers. Nowadays, most of the colors used in commercial textile dyeing are synthetic. They are synthesized, by various means, from byproducts of fossil fuels, e.g., aniline and other aromatic derivatives. However, oil supplies are a limited, nonrenewable resource. Currently, there is a move to find renewable resources to reduce the depletion of global supplies of fossil fuels,[1,3] which has initiated research into natural products from plants, including coloring matters such as indigo and curcuma.[4–6] The revival interest in the use of natural dyes is also a result of the stringent environmental standards imposed by many countries in a response to the toxic and allergic reactions associated with synthetic dyes.[7]

In this work, we tried to develop the dyeing function of *Rhizoma coptidis*. Berberine is the only cationic colorant of natural plant dyes, which lies in the roots of *R. coptidis* and stems of phellodendron.[8]

As famous Chinese herb, *R. coptidis* originally rooted in Shen Nong Ben Cao Jing (Divine Husbandman's Classic of the Materia Medica) in the second century. Its English name is coptis or coptis root. The major constituents of *R. coptidis* are berberine (Scheme 1) and related protoberberine alkaloids. Berberine occurs in the range of 4–8%, followed by palmatine, coptisine, berberastine among others.[9]

Numerous reports support the antimicrobial activity of *R. coptidis*. *In vitro* studies have shown that the crude drug and its active constituent, berberine, have a broad spectrum of antibacterial action.[10,11] Both inhibit the *in vitro* growth of staphylococci, streptococci, pneumococci, vibrio cholerae, bacillus anthracis, and bacillus dysenteriae. Berberine was also active *in vitro* against *Entamoeba histolytica*, *Giardia lamblia*, and *Trichomonas vaginalis*. Consequently, it is necessary to ascertain whether *R. coptidis* can produce the source of the yellow dye and keep its antimicrobial activity. Since natural dyes usually work best with natural fibers such as wool, cotton, silk, and jute,[12] and this article pays more attention to protein fiber, wool was selected and dyed with extract of *R. coptidis*.

EXPERIMENTAL

Material

Wool fabric (twill, greige, and weight, 200 g/m^2) was procured commercially. *R. coptidis*, shaped like a cockspur, 5–6-cm long, brownish yellow, densely covered with numerous nodes and in transverse section, the central pith deeper in color, was purchased from medicine market.

Extraction of *R. coptidis*

R. coptidis (10 g) was minced and dipped in distilled water for 30 min, then was boiled in distilled water for

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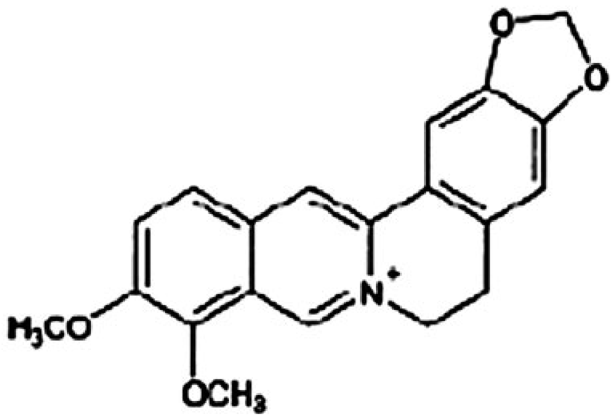

Scheme 1 Chemical structure of berberine.

1 h. The content was cooled at room temperature and filtrated. The *R. coptidis* residue was again boiled in distilled water and filtrated. Finally, the filtrate from the two extractions was mixed and the mixed filtrate concentrated to 200 mL, and this was taken as the original extract and used for dyeing. The pH value of the original extract was 6.5.

Dyeing process

In a dye bath containing *R. coptidis* extract and 1 g/L sodium chloride, wool fabric was dyed in a normal temperature sample dyeing machine, keeping material-to-liquor ratio at 1 : 20. First, the wool fabric was wet out with hot water and then added to the dye bath at room temperature. The temperature was raised to 50°C and maintained for 30 min. Finally, the dyed fabric was removed, rinsed with deionized water, and washed using a nonionic detergent and dried. This was considered as direct dyeing.

Dyeing with mordant Fe^{2+} was done as follows. $FeSO_4$ concentration was 7% on weight of the fabric, keeping material-to-liquor ratio at 1 : 20. Fabric was introduced into the dyeing solution at room temperature, and then the process of direct dyeing was replicated.

In addition, to investigate the effect of concentration, temperature, and pH value on color character, the dyeing treatment in *R. coptidis* extract without salt addition was carried out at the following conditions.[1] Wool fabric was dyed at different concentration of extract to get the optimum recipe of the finish. Extract concentration was varied as follows: 100 mL original extract was taken as dye bath 100% concentration, for 75% concentration 75 mL herb extract was diluted with 25 mL water. By this method, a series of solution with different concentrations were obtained. Other conditions were the same as that of direct dyeing.[2] Maintaining other conditions, the pH of dye bath was varied from 2 to 10 by adding buffer solution (acetic acid + sodium carbonate). Keeping other conditions the same as that of direct dyeing, the treatment temperature was raised to 75°C and 95°C respectively, to get proper color value.

Fastness test

Color fastness expresses the resistance of a material to change in any of its color characteristics, to transfer of its colorant(s) to adjacent materials, or both, as a result of the exposure of the material to any environment during process and testing(including rubbing test and washing test). Color staining is the unintended pickup of colorant by a substrate due to direct contact with dyed or pigmented material, from which colorant transfers by sublimation or mechanical action.

Wash-fastness test was carried according to GB/T3921.1–1997, with a soap solution 4 g/L (liquor ratio 50:1) for 30 min at 40°C. Rub-fastness was measured according to GB/T3920–1997 using Y571B rub-fastness tester. The samples were assessed against the standard gray scale for color change and staining of adjacent undyed fabric. A rating scale consists of pairs of standard gray chips, the pairs representing progressive differences in color or contrast corresponding to numerical colorfastness grades, the numerical value that is assigned to the change in color of a test specimen as compared to an original specimen. The rating scale was 1 (poor) to 5 (excellent). Color-fastness grade 5 is represented on the scale by two reference chips mounted side by side, and the color difference of the pair is 0.0 ± 0.2, color-fastness grade 1 represents the color difference of the pair is 13.6 ± 1.0.

Color values such as L^*, a^*, b^*, c^*, H^0, and K/S were measured for dyed fabric by using DatacolorSF600, Computer Color Matching System (Data Color International), using illuminant D_{65} and 10° standard observer. ΔE, L^*, a^*, b^*, c^*, and H^0 represents color difference, lightness, redness-greenness of color, yellowness-blueness of color, saturation of color, and hue value, respectively. K/S value is relative color strength and determined using the Kubelka–Munk equation:

$$K/S = (1 - R)^2/2R \qquad (1)$$

where K refers to coefficient of absorption, S is coefficient of scatter, and R is fractional reflectance.

Antibacterial test

The antimicrobial activity of dyed wool specimens was tested using modified Quinn's method.[13] *Escherichia coli*, a gram-negative bacterium, and *Candida albicans*, a common fungi, were selected due to their popularity of being selected as a test organism. *E. coli* and *C. albicans* were supplied by Chinese Collection for Type Cultures in Wuhan University. Nutrient agar

TABLE I
Color Values for Wool Fabric After Direct Dyeing and Dyeing with Mordant

	Direct	Mordant
ΔE	77.405	62.645
L^*	68.991	53.97
a^*	6.941	8.233
b^*	72.967	47.576
c^*	72.735	47.749
H^0	84.887	80.71

medium (g/L: peptone 1.0; glucose 4.0; agar 20; distilled water 100 mL) was prepared and autoclaved at 121°C for 30 min, then cooled to 50°C, and 15 mL of the nutrient medium was added into sterilized plates (9 cm in diameter). Fabric (2.5 cm^2; dyed and undyed) inoculated with the desired microbe was introduced into agar plates and incubated at 37°C overnight (24 h). To evaluate antimicrobial activity of *R. coptidis* on wool fabric, we compared the number of colonies (after incubation) in the treated fabric with that of the untreated fabrics.

RESULTS AND DISCUSSION

Direct dyeing and dyeing with mordant

Color values for wool fabric after direct and mordant dyeing

For wool fabric dyed with mordant, treatment with Fe^{2+} produced a noticeable color change, as indicated in Table I.

It is noted that after mordanting the yellowness and vividness of wool fabric decreased obviously. The minor decrease of H^0 further confirmed propensity of color change, from yellowness propensity to redness. Lower ΔE value was mainly attributable to sharp decrease of saturation of color. It is clear from these data that ferrous sulfate mordanted substrate yielded darker and less bright shade, as is concluded from color measurements (Table I) for the corresponding dyed fabrics. These changes of color values may be due to the impact of ferrous sulfate on pigment chemical structure.

Fastness of direct dyeing and dyeing with mordant

Table II shows the wash-fastness and rub-fastness ratings for *R. coptidis* dye on wool fabrics. Direct dyeing showed good rub-fastness whereas wash-fastness was poor. Unexpectedly mordant gave no improvement in the wash-fastness and rub-fastness, except rating for fading. The chemistry of bonding of dyes to fiber is complex. It involves direct bonding, H-bonds, and hydrophobic interactions. In many ways, mordants help binding of dyes to fabric by forming a chemical bridge from dye to fiber, thus improving the staining ability of a dye along with increase in its fastness properties. However, in this study, the chances of electrostatic repulsion between Fe^{2+} and N^+ site of berberine[14] made the complex of dye with iron metal unstable and may result in the decrease of fastness. Anyway, the true reason needs further experiment to determine.

TABLE II
Wash-fastness and Rub-fastness Ratings for Direct and Mordant Dyeing

Dye	Wash-fastness		Dry rub-fastness		Wet rub-fastness	
	Fading	Staining	Fading	Staining	Fading	Straining
Direct	1–2	3	4	4	4–5	3
Mordant	2	2	3	2–3	4	2

Effects of extraction concentration on color values

The ΔE, L^*, a^*, b^*, c^*, H^0 color values for the wool fabric treated in different concentration are reported in Table III.

It is observed that color values of dyed wool fabric had little change when the extract concentration varied from 50 to 100%, i.e. when concentration of dye bath reached 50%, color value basically kept stable. Certainly some minor differences were observed. The lightness value $L^*$and hue value H^0 decreased with the increase in concentration of extract, as shown in Figure 1.

In general, when dyed in lower concentration bath, the wool fabric had more bright and less red character (higher L^* and lower a^* value). The driving force for transfer of a dye molecule from one phase to the other (dye bath to fiber or fiber to dye bath) is the concentration gradient of dye in the two phases. With the increase of the concentration of dye bath, more dye transferred to fabric and thus the apparent depth of color increased.

Effects of pH value on color values

Table IV shows that the pH values of the dye bath have considerable effect on the color of the dyed wool

TABLE III
Color Values for Wool Fabric Dyed with Different Concentration

Concentration (%)	ΔE	L^*	a^*	b^*	c^*	H^0
100	57.329	61.885	18.337	62.184	64.832	73.570
75	57.883	63.465	17.553	63.881	66.248	74.635
50	57.946	64.064	18.259	63.969	66.524	74.069
25	54.338	69.053	14.479	63.143	64.784	77.085
0	0	86.390	0.170	13.773	13.774	90.706

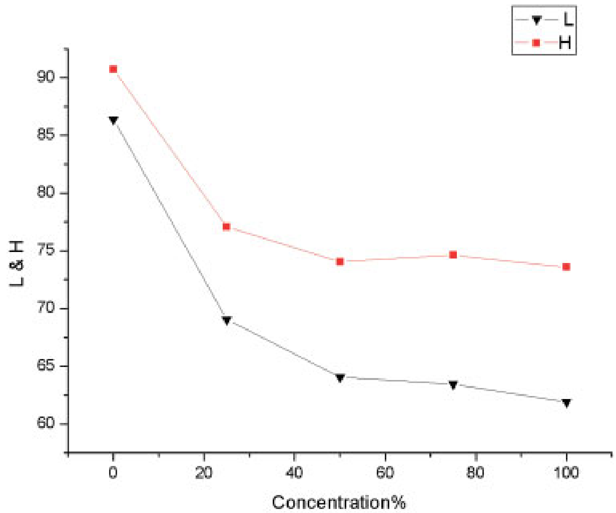

Figure 1 Lightness and hue for wool fabric dyed with different concentration. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

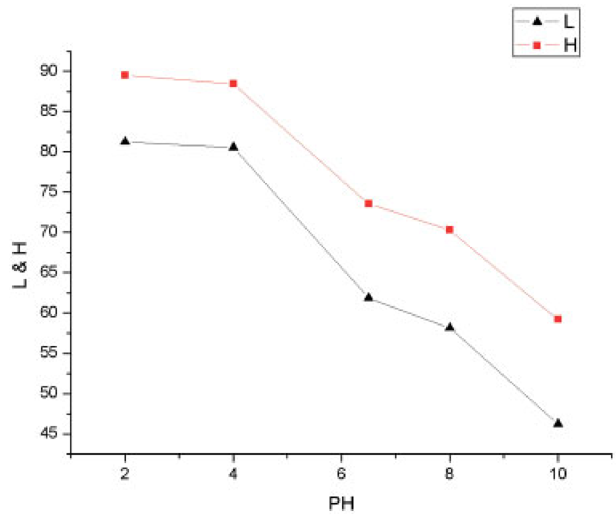

Figure 2 Lightness and hue for wool fabric dyed with different pH. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

fabrics. Redness-greenness (a^*) declined sharply as pH value decreased, and lightness and hue value increased obviously, as observed clearly in Figure 2. Wool fabrics dyed in alkali bath possessed greener and more yellow character and bigger chromatic value compared with that in acidic bath. And saturation (c^*) reached maximum value in original dyeing solution.

The effect of dye bath pH can be attributed to the correlation between dye structure and wool fibers. Since the berberine is a water soluble dye containing cationic quaternary ammonium salt, it would interact ionically with the carboxyl end groups of wool fibers at alkaline pH via ion exchange reaction. The number of available anionic sites on fibers in alkaline conditions is relatively larger than that in acidic conditions, and thus the exhaustion of the cationic berberine was increased in alkaline dyeing solutions. But it is noticeable that wool fiber has not strong resistance to alkali, thus pH of dye bath is not suitable too high.

Effects of treatment temperature on color values

The effect of temperature on the dyeability of wool fabrics was conducted at different temperatures (50, 70, 90°C). As shown in Table V, it is clear the color value decreased with increase in the temperature from 50 to 90°C. When dyed at lower temperature, wool fabric color was bright gay (brighter and more vivid). With the rise in temperature, the conglomeration of colorant molecules declined, and the diffusion of berberine into the fibers could occur very easily and rapidly at a higher temperature, which brought the change of lightness and saturation and color difference and produced color variety.

K/S determination for wool fabrics after dyeing

Absorption spectra were recorded to obtain the K/S values for dyed wool fabrics. The wavelengths of maximum absorption and K/S values identified for direct dyeing and mordant dye and dyes under different conditions are given in Table VI.

There was no apparent shift in absorption spectra when dyed in conditions listed in Table VI.

As expected, color strength increased as treatment temperature and concentration of dye bath increased. Especially with increasing temperature, the affinity of the dyes to the fiber substrate increased clearly. And when dyed under 90°C, wool fabric obtained maxi-

TABLE IV
Color Values for Wool Fabric Dyed With Dyebath of Different pH

pH	ΔE	L^*	a^*	b^*	c^*	H^0
2	35.800	81.231	0.405	49.195	49.196	89.528
4	30.119	80.527	1.197	43.285	43.301	88.416
6.5	57.329	61.885	18.337	62.184	64.832	73.570
8	57.768	58.154	21.261	59.385	63.077	70.301
10	61.308	46.243	29.431	49.419	57.519	59.225

TABLE V
Color Values for Wool Fabric Dyed Under Different Temperature

Temperature (°C)	ΔE	L^*	a^*	b^*	c^*	H^0
50	57.329	61.885	18.337	62.184	64.832	73.570
70	55.617	56.150	17.528	57.080	59.710	72.930
90	53.370	53.417	14.798	53.100	55.124	74.428

mum K/S value. The maximum change in K/S value was observed when dyed in acidic medium. It was observed that at pH 4, there was a hypsochromic shift and K/S value reached minimum value. Then brighter and more vivid wool fabric of yellow color can be obtained by dyeing with *R. coptidis* extract in acidic medium. The higher pH conditions led to better dyeability since berberine was more attractive to the negatively charged carboxylic group of wool fiber under basic conditions.

Antibacterial property

As famous traditional Chinese medicine, *R. coptidis* has broad-spectrum antibacterial abilities that have gained plentiful validation both in traditional Chinese medicine and clinical medicine. As expected, *R. coptidis* imparted good antibacterial activity on the dyed wool fabric. The results are shown in Table VII.

Indeed, the dyed sample with berberine displayed very effective antimicrobial activity and the inhibition rate against to *E. coli* was higher than that against *C. albicans*. This corresponding result can be explained as follows: the structure of the quaternary ammonium salt in the berberine molecules could destroy the negatively charged cell membrane of bacteria by disturbing charge balances of the cell membrane. A common dyeing process provides wool fabric with color and antimicrobial properties that will have tremendous application in apparels and medical textiles.

CONCLUSIONS

Wool fabrics dyed with extraction of *R. coptidis* under different conditions can show various yellow colors, which may result in the difference of dyed samples' antimicrobial function. A positive correlation was observed between the extraction concentration, pH value of dye bath, treatment temperature, and K/S value. Among all factors, the effect of pH value on color characteristic was most noticeable. However, results from fastness testing indicated that both direct and mordant dyeing imparted low wash fastness, which meant the compatibility between berberine and mordant Fe^{2+} was poor and mordant was unsuitable in such dyeing, otherwise dyeing method needs improvement. Wool fabric treated with *R. coptidis* extract also showed excellent antibacterial property. Above all, *R. coptidis* as natural dye with medical function is deserved of further study.

TABLE VI
Absorption Spectral Data for Dyeings Generated on Wool Fabric

Dyeing conditions	λ_{max}	K/S
Dye type		
Direct	440	13.471
Mordant	440	11.845
Extraction concentration (%)		
100	440	13.471
75	430	12.971
50	440	12.466
25	430	8.370
Treatment temperature (°C)		
90	430	17.379
70	430	16.512
50	440	13.471
pH value		
2	430	2.074
4	420	1.718
6.5	440	13.471
8	430	15.992
10	430	14.315

TABLE VII
Results of Antibacterial Testing

Sample	*E. coli*	*C. albicans*
Control fabric	53	417
Treated fabric	0	125

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References

1. Kerry, G.; David, T. Plant Growth Regul 2001, 34, 57.
2. Bechtold, T.; Turcanu, A.; Ganglberger, E.; Geissler, S. J Cleaner Product 2003, 11, 499.
3. Borland, V. Am Text Indust 2000, 29, 66.
4. Sarkar, R.; Purushottam, D.; Chauhan, P. Indian J Fiber Text Res 2003, 28, 322.
5. Sarkar, A.; Seal, C. Cloth Text Res J 2003, 21, 162.
6. Ajoy, K. BMC Dermatol 2004, 4, 15. Available at http://www.biomedcentral.com/1471–5945/4/15.
7. Kamel, M. M.; El-Shishtawy, R. M.; Yussef, B. M.; Mashaly, H. Dyes Pigments 2005, 65, 103.
8. Chang, H.; But, P. Pharmacology and Applications of Chinese Materia Medica; World Scientific: Singapore; 1987.
9. Kaneda, Y.; Torii, M.; Tanaka, T.; Aikawa, M. Ann Trop Med Parasitol 1991, 85, 417.
10. Zhang, H. Identifying and Application of Chinese Local Medicine Material; Guangdong Tour, China; 2000.
11. Nam, C.; Kim, S.; Sim, Y. Chang, I. Pharmacol Appl Skin Physiol 2003, 16, 84.
12. Vankar, P. S. Resonance 2000, 10, 73.
13. Zou, C.; et al. Dyeing Finish 1994, 22, 33.
14. Gulrajani, M. L.; Gupta, D.; Maulik, S. R. Indian J Fibre Text Res 1999, 24, 223.