Journal of Cleaner Production 80 (2014) 204–210

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro

Natural dye extracted from Chinese gall – the application of color and antibacterial activity to wool fabric

Bin Zhang [a,*], Lu Wang [a], Liangfei Luo [a], Martin W. King [a,b]

[a] Key Laboratory of Textile Science and Technology of Ministry of Education, College of Textiles, Donghua University, Shanghai 201620, China
[b] College of Textiles, North Carolina State University, Raleigh, NC 27695, USA

ARTICLE INFO

Article history:
Received 24 May 2013
Received in revised form
24 May 2014
Accepted 31 May 2014
Available online 11 June 2014

Keywords:
Wool fabric
Chinese gall
Natural dye
Dyeing parameters
Colorfastness
Antibacterial activity

ABSTRACT

A natural dye has been extracted from Chinese gall and its dyeability of wool fabric has been studied together with its colorfastness and antibacterial performance. A fractional factorial experimental design using Taguchi's orthogonal array was undertaken to determine the optimum extraction conditions of a 90 °C temperature, a 1:30 liquor ratio and two extraction cycles each 60 min long. The extracted dye liquor was then used in a second orthogonal design of experiment to determine the optimum pre-mordant dyeing conditions on wool fabric. Under these conditions of 200% dye liquor concentration, 2.5% mordant concentration, pH 8 and 98 °C, the colorfastness of the optimum dyed fabric to washing, crocking and perspiration were found to be all acceptable with at least a Grey Scale rating of 3. Antibacterial activity of this dyed wool was confirmed by exposing the fabric to *Staphylococcus aureus* and *Escherichia coli*. The fabric dyed with Chinese gall extract reduced the number of viable organisms by 99.90% and 96.55% respectively. The market evaluation and cost of Chinese gall have been analyzed. The economic benefit for Chinese gall was acceptable and can be used in dyeing and antibacterial finishing of textile materials.

1. Introduction

Since some synthetic dyes are associated with environmental pollution and are harmful to people's health, more attention is being paid to environmental protection, sustainable development and natural lifestyle. Interest in the potential use of natural dyes has been growing, given they have high compatibility with the environment, minimal toxicity and low allergic reaction, and they are available in a range of natural shades as compared with synthetic dyes. Natural dyes can be obtained from plants, animals and minerals (Shahid et al., 2013). It is reported that many natural dyes can not only dye unique and natural shades, but can also provide functions to fabrics such as antibacterial activity (Prusty et al., 2010; Khan et al., 2011; Yusuf et al., 2012), ultraviolet protection (Feng and Zhang, 2007; Hou et al., 2013) and insect repellency (Shahid et al., 2013). These natural dyes have been successfully applied to natural fiber fabrics such as cotton (Ali et al., 2009), wool (Khan et al., 2011; Yusuf et al., 2012; Hou et al., 2013), silk (Prusty et al., 2010; Baliarsingh et al., 2012) and flax (Sarkar and Seal, 2003). However, limited availability and high-cost restricted the industrialization of many natural dyestuffs.

Chinese gall is a traditional Chinese herbal medicine originating from the abnormal growth of the sumac gallnuts (*Rhus semialata*) plant in response to secretions of the parasitic aphid (*Pemphigidae family*) (Tian et al., 2009). Images of Chinese gall growing on this plant and its nuts are shown in Figs. 1 and 2. Chinese gall is well known for having abundant gallotannins, a type of hydrolysable tannin, which can represent up to 70% (w/w) of this plant material. Gallotannins are polymers formed when gallic acid (Fig. 3), a polyphenol monomer, esterifies and binds with the hydroxyl group of a polyol carbohydrate such as glucose. This structure contains several gallic acid units, as well as depside moieties (Fig. 4) which are known to have antibiotic, antioxidant and anti-inflammatory activity (Tian et al., 2009; Sun, 1992). When the glucose core is esterified with five or fewer galloyl groups, the resulting compounds are defined as gallotannin precursors. Chinese gall has a wide antibacterial spectrum. For example, in vitro experiments indicate that it has bacteriostatic or bactericidal activity against *Staphylococcus aureus*, *Streptococcus*, *Pneumococcus* and *Salmonella typhi*, *Bacillus dysentery*, *Bacillus anthracis*, *Diphtheria*, and *Pseudomonas aeruginos* (Tian et al., 2009). It is proposed that Chinese gall extract might be able to dye wool fabrics and can also provide

* Corresponding author. Tel.: +86 21 67792694; fax: +86 21 67792627.
E-mail address: zhangbin@dhu.edu.cn (B. Zhang).

http://dx.doi.org/10.1016/j.jclepro.2014.05.100
0959-6526/

Fig. 1. Chinese gall growing on *Rhus semialata* plant.

Fig. 2. Chinese gallnuts.

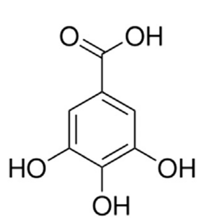

Fig. 3. Chemical formula of gallic acid.

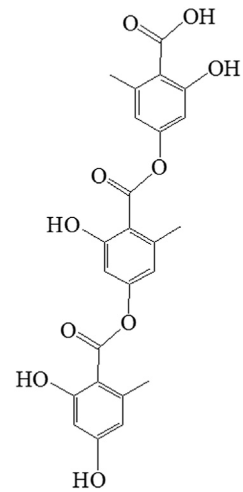

Fig. 4. Chemical formula of depside moiety.

antibacterial activity. There has been no research in this area, though extensive R&D in the area of natural dye applications is underway worldwide such as India, Japan, Korea, China, Thailand, Pakistan, Turkey, Austria, Egypt France, UK and USA (Shahid et al., 2013). The objective of the current study was to investigate the dyeability and antibacterial activity of Chinese gall extract on wool fabrics as well as optimize the conditions for its extraction.

2. Experimental

2.1. Materials and chemicals

In this study, Chinese gallnuts were harvested in late summer and early autumn. The dried product was obtained from Yuanye Biological Technology Co., Ltd., Shanghai, China. The characteristics of a single jersey weft knitted scoured wool fabric, supplied by Heilan Group, Jiangsu Province, are presented in Table 1. The mordant used in this study was aluminum potassium sulfate. Other chemicals, such as anhydrous sodium carbonate, sodium hydroxide, sodium chloride, disodium hydrogen phosphate and sodium dihydrogen phosphate were used to control the pH value and as auxiliaries for the dyeing experiments. They were supplied as laboratory reagent grade chemicals by Shanghai Chemical Agents Company.

2.2. Experimental design

2.2.1. The extraction conditions of Chinese Gall colorant

In order to optimize the extraction conditions for the Chinese gall colorant, we undertook a fractional factorial orthogonal design of experiment with temperature, duration, number of extraction cycles and liquor ratio as the four independent factors which controlled the level of extraction (Taguchi and Konishi, 1987). The levels of each factor in the Taguchi's orthogonal array design of experiment are shown in Table 2. The orthogonal array of 9 experiments L_9 (3^4) is presented in Table 3. They were performed in a random order.

Table 1
Characteristics of wool fabric used in the study.

Wale count (wales/cm)	9.0	Single yarn count (Nm)	60
Course count (courses/cm)	11.0	Mass per unit area (g/m^2)	132
Stitch density (stitches/cm^2)	99.0	Original color	Natural