Carbohydrate Polymers 106 (2014) 319–325

Contents lists available at ScienceDirect

Carbohydrate Polymers

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

Antimicrobial fabrication of cotton fabric and leather using green-synthesized nanosilver

Palanivel Velmurugan[a,1], Min Cho[a,1], Sang-Myeong Lee[a], Jung-Hee Park[a], Sunyoung Bae[b], Byung-Taek Oh[a,*]

[a] Division of Biotechnology, Advanced Institute of Environment and Bioscience, College of Environmental and Bioresource Sciences, Chonbuk National University, Iksan, Jeonbuk 570-752, South Korea
[b] Department of Chemistry, Seoul Women's University, Seoul 139-774, South Korea

ARTICLE INFO

Article history:
Received 24 December 2013
Received in revised form 28 January 2014
Accepted 5 February 2014
Available online 15 February 2014

Keywords:
Antibacterial activity
Flower
Silver nanoparticles
Phytosynthesis
Fabrics

ABSTRACT

This study aims to investigate the green synthesis of silver nanoparticles (AgNPs) by *Erigeron annuus* (L.) pers flower extract as reducing and capping agent, and evaluation of their antibacterial activities for the first time. The obtained product was confirmed by UV–Vis spectrum, high resolution-transmission electron microscopy, energy-dispersive X-ray spectroscopy, Fourier transform infrared spectroscopy, and X-ray diffraction studies. The optimum AgNPs production was achieved at pH 7, metal silver (Ag^+ ion) concentration of 2.0 mM, flower extract concentration 4%, and time 335 min. In addition, the antibacterial activity of cotton fabrics and tanned leather loaded with AgNPs, commercial AgNPs, flower extract, Ag^+ ion and blend of flower extract with AgNPs were evaluated against Gram-positive odor causing bacteria *Brevibacterium linens* and *Staphylococcus epidermidis*. The results showed maximum zone of inhibition (ZOI) by the cotton fabrics embedded with blend of flower extract and AgNPs against *B. linens*. The structure and morphology of cotton fabric and leather samples embedded with AgNPs, Ag^+ ion and blend of flower extract with AgNPs were examined under field emission scanning electron microscope.

1. Introduction

Over the past few decades, substantial research attempt was made to fabricate antibacterial coatings on the surfaces of varied objects, such as garments, medical devices and food pack, to prevent infection and spoilage (Ravindra, Murali Mohan, Narayana Reddy, & Mohana Raju, 2010). Several attempts have been made to develop economical, non-toxic, and value effective antimicrobial finishing textiles for applications in medical, healthcare, pharmaceutical, hygienic products, and protective textile (Ul-Islam, Shahid, & Mohammad, 2013). Metal nanoparticles synthesized by physical, chemical and biological routes were extensively studied as a result of their wide applications (Mochochoko, Oluwafemi, Jumbam, & Songca, 2013; Isaacet al., 2013). Synthesis of various metal nanoparticles through chemical and physical routes is found to show certain toxicological effects in the medical research field (Kanmani & Lim, 2013); additionally it is quite expensive and as well unsafe. To overcome this limitation, we have fabricated AgNPsin an eco-friendly, inexpensive, easily available substrate, and safe method of synthesis from a phyto source (flower). The substrate originated from a natural source, particularly textile and leather, acts as an excellent harbor for microbes, as they will provide ideal conditions like moisture, temperature, oxygen, and nutrient required for its growth (Ul-Islam et al., 2013). Microbial pathogens have lethal effect on all forms of life. The odor in feet, shoes and/or socks is due to breakdown of amino acids present in our sweat and skin by *Brevibacterium linens* and *Staphylococcus epidermidis*. *B. linens* can breakdown an amino acid methionine present in sweat into methanethiol a gas. *S. epidermidis* creates body odor by breaking sweat into isovaleric acid (3-methyl butanoic acid) from the amino acid leucine (Ara et al., 2006; Kanlayavattanakul & Lourith, 2011), which causes unpleasant smell in feet, shoes and/or socks. To beat this difficulty, it is truly fascinating to possess antimicrobial properties in textile and leather product. Many chemicals and its strategies of imparting antimicrobial property into textile materials are of not eco-friendly, toxic to humans, and also bacteria will develop resistance over the chemical or/antibiotics (Rajendran, Radhai, Kotresh, & Csiszar, 2013; Ul-Islam et al., 2013). Thus, the people are now increasing awareness on antibiotic textile and leather finishing with eco-friendly and biodegradability. In view of these ecological and environmental concerns, green source (plants and its parts) become a key resources to obtain

* Corresponding author. Tel.: +82 63 850 0838; fax: +82 63 850 0834.
E-mail address: btoh@jbnu.ac.kr (B.-T. Oh).
[1] These authors contributed equally to this work.

http://dx.doi.org/10.1016/j.carbpol.2014.02.021
0144-8617/

eco-friendly nanoparticles to fabricate bioactive fabric and leather products.

To combat these adversities, we have been made to fabricate AgNPs using *Erigeron annuus* (L.) pers flower extract as a reducing and capping agent for fabric and leather finishing. To brief, genus *E. annuus* belonging to the family Asteraceae (tribe Astereae), involves about 150 species occurring within the hemisphere zone, mainly in North America. Some of them were introduced to Europe. *E. annuus* (daisy fleabane) is an annual plant and reaches a height of up to 150 cm posses erect, branched stem completing inflorescences. The central disk florets are numerous, very small, and yellow; they are surrounded by 50–120 white ray florets. Both kinds of florets can be self-fertile. They often settle on same places like roadsides and wastelands. Various parts of *E. annuus* have been employed in Chinese folk drugs for the treatment of dyspepsia, enteritis, epidemic hepatitis, and haematuria. As the constituents of the aerial part of *E. annuus* contain γ-pyranone derivatives, flavonoids, and phenolic acids and their derivatives, sesquiterpenoids and cyclopentenone derivatives have been reported (Nazaruk & Kalemba, 2009). Human beings are often infected by microorganisms such as bacterium, mold, yeast, and virus in living environment. In this study, the synthesis and application of AgNPs were systematically optimized, characterized and their antibacterial properties of cotton fabric and tanned leather samples loaded with various obtained materials also evaluated against the bacteria responsible for causing odor in sweat, shoe and shocks.

2. Experimental

2.1. Flower material

The *E. annuus* flowers were collected from the road side of Chonbuk National University, Iksan, South Korea and washed thoroughly with copious amount of RO in nanopure purified water (conductivity = 18 μΩ/m, TOC < 3 ppb, Barnstead, Waltham, MA, USA). Such flowers (100 g) were added to 250 ml distilled water and crushed by a juicer. The extract was filtered through a Whatman filter paper and stored at 4 °C for further experiments.

2.2. Chemicals and media

Silver nitrate ($AgNO_3$) (99.9%) acquired from DaeJung chemicals, South Korea, and commercial silver nano powder (99.5%, <100 nm) purchased from Sigma–Aldrich (St. Louis, MO), were used for the synthesis of AgNPs, and Brain Heart Infusion broth (BHI) and Mueller-Hinton agar (MHA) were purchased from MB Cell, South Korea for antibacterial study. All chemicals were used as supplied. Nanopure purified water was used throughout this investigation.

2.3. Microorganisms

The antimicrobial activity of cotton fabric and leather samples loaded with various materials was evaluated against two stains *B. linens* (KACC-14346) and *S. epidermidis* (KACC-13234) purchased from KACC (Korean Agricultural Culture Collection). The microbial cultures were maintained in nutrient agar media and stored in 4 °C for further use.

2.4. Synthesis and optimization of silver nanoparticles

For the entire study, silver nitrate ($AgNO_3$) was used as a source for silver in nanopure water. For the AgNPs synthesis, 5 ml of *E. annuus* flower extract was added to 45 ml of 1 mM aqueous $AgNO_3$ solution in a 250 ml Erlenmeyer flask. To obtain optimum AgNPs production, various parameters like pH (4, 5, 6, 7, 8, 9, and 10), flower extract (1, 2, 3, 4, 5, 6, 7, 8, 9 and 10%), Ag^+ ion concentrations (0.25, 0.5, 1.0, 2.0, 3.0,4.0 and 5.0 mM), and time (15, 30, 45, 60, 95, 110, 125, 140, 175 and 190 min), were studied. The reaction mixtures were filtered through 0.22 μm Steritop Millipore filters and centrifuged at 12,000 rpm for 15 min for AgNPs isolation. The resulting pellets were redispersed in nanopure water to eliminate any uncoordinated molecules. The process was repeated several times in order to ensure better separation of free entities from the metal NPs. The obtained NPs were stored freeze-dried to obtain a powder. To check the stability of synthesized AgNPs, it was exposed to ambient condition for several months and scanned for the wavelength. Furthermore, the overall study and production was carried out in the optimized parameters. All experiments were carried out in triplicates.

2.5. Preparation of leather and fabrics samples

The leather sample was purchased from the local market in South Korea. The obtained leather was punched with help of 13 mm cork borer in order to get spherical shape and slice to reduce the thickness by removing the smooth surface, using a sharp knife with care. To remove the impurities, the leather was immersed in boiling water (50 ml) with 2 ml/l of non-ionic detergent for 1 h, followed by hot and cold water washing (to avoid break down of the emulsion and precipitation of the impurities onto the leather), and air dried at room temperature. Cotton fabric was obtained from the local market in Iksan, South Korea. To remove the impurities, 3 g of cotton fabric treated in the same condition was used for leather. Later the dried cotton fabrics were punched to get spherical holes. These samples were used for further study. Before embedding the particles, we ensured that the fabric and leather samples were free from any chemicals obtained by EDX spectra.

2.6. Embedding silver nanoparticles onto cotton fabrics and tanned leather

Different materials are used for embedding onto cotton fabric and tanned leather like synthesized AgNPs (0.01 g/10 ml), commercial AgNPs (0.01 g/10 ml), Ag^+ ion (0.01 g/10 ml), flower extract (5 ml), and flower extract with AgNPs (5 ml: 0.01 g/5 ml). The commercial grade AgNPs were kept in glassware cleaning ultra sonicator water bath with maintained agitation for 20 min at 100 Hz (Elmasonic S30H. Ultrasonic bath, Germany). For the successive embedding of each materials on to cotton fabric and leather samples, the samples were immersed in separate 50 ml screw cap tubes with the above said materials and kept in a ultrasonicator with manual agitation for 20 min at 70 °C at 100 Hz, respectively. The samples were then pressed with tissue paper. The same conditions were followed for control samples without the implant material. To remove excess particles or materials, the dried samples were subjected to several washing with detergent water and the same drying conditions were repeated. The cotton fabric samples were dried at 120 °C for 2 min, and leather samples were dried at 80 °C for 15 min (in order to avoid damage occur faster at higher temperatures) for curing, respectively. Later, the antibacterial efficacy was evaluated in triplicates for each prepared fabric and leather samples and average values were presented.

2.7. Characterization of AgNPs and coated cotton fabric and leather samples

The optical absorption spectra of the AgNPs were observed by UV-1800, UV–Vis (Shimadzu, Japan) spectrophotometer. The morphology of AgNPs were observed using HR-TEM (JeolJEM100SX). The elemental composition of the AgNPs was analyzed using energy dispersive spectroscopy (EDX) (SEM–EDX; JEOL-6 4000, Japan).