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Combined antimicrobial finishing and pigment printing of cotton/polyester blends

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ABSTRACT

This study demonstrates the possibility of enhancing the antibacterial functionality and pigment printing properties of cotton/polyester blends (50/50 and 35/65) in one step. Inclusion of chitosan (10 g/kg), choline chloride (15 g/kg), triclosan derivative (20 g/kg), hyperbranched poly amide-amine/silver or zinc oxide nanoparticles (HBPAA/Ag-NP's hybrid or HBPAA/ZnO-NP's hybrid – 20 g/kg) into a pigment print formulation followed by printing and microwave curing at 386 W for 5 min results in an improvement in antibacterial activity and pigment printability. It was further noted that, in all cases, the G+ve (*S. aureus*) bacteria is more susceptible to the action of the immobilized antibacterial agents than the G−ve bacteria (*E. coli*). The functionalized pigment prints exhibited very sufficient antibacterial activity even after 20 washing cycles. Modes of interactions were proposed, and surface modification was also confirmed by SEM and EDX analysis.

1. Introduction

Among the various functional finishes, high attention has given to the textile materials that are bioactive, to cope with the growing awareness of health and quality of hygiene, without adversely affecting their traditional characteristics using different antimicrobial agents. The available antimicrobial agents differ in their chemical structure, antibacterial activity, application method, mode of interaction as well as environmental impact (Gao & Cranston, 2008; Simoncic & Tomsic, 2010).

The most promising antimicrobial agents currently available for textile application include: inorganic compounds such as metal salts, nano-sized metals and metal oxides (Dastjerdi & Montazer, 2010; Gao & Cranston, 2008; Gowri, Almeida, Amorim, Carneiro, Souto, & Esteves, 2010; Ibrahim, Abo-Shosha, Gaffar, Elshafei, & Abdel-Fattah, 2006; Ibrahim, Mahrous, El-Gamal, Gouda, & Husseiny, 2010; Ibrahim, Eid, Hashem, Refai, & El-Hossamy, 2010; Ibrahim, El-Gamal, Gouda, & Mahrous, 2010; Ibrahim, Eid, & El-Batal, 2012; Ibrahim, Eid, Youssef, El-Sayed, & Salah, 2012; Ibrahim, Amr, Eid, Mohamed, & Fahmy, 2012; Ibrahim, Refaie, & Ahmed, 2010; Mahapatra & Karak, 2008) halogenated phenols (Hashem, Ibrahim, El-Sayed, El-Husseiny, & Elanany, 2009; Hashem, Ibrahim, El-Shafei, Refaie, & Hauser, 2009; Ibrahim, Hashem, El-Sayed, El-Husseiny, & Elanany, 2010; Orhan, Kut, & Gunesoglu, 2009), N-halamines (Gouda & Ibrahim, 2008; Ibrahim, Aly, & Gouda, 2008; Liu & Sun, 2006), chitosan (Lim & Hudson, 2003; Shin, Yoo, & Jang, 2001), Neem oil (Ibrahim, Eid, & El-Zariy, 2011; Joshi, Ali, & Rajendran, 2007), antibiotics (Ibrahim, Eid, et al., 2010), quaternary ammonium compounds (Gao & Cranston, 2008), and immobilized enzymes (Ibrahim, Gouda, El-Shafei, & Abdel-Fattah, 2007).

In recent years, the use of nanotechnology in textile field has increased rapidly to develop efficient, non-toxic, durable and cost effective functionalized textiles with multifunctional properties and increased potential applications (Dastjerdi & Montazer, 2010; Gowri, Almeida, Amorim, Carneiro, Souto, & Esteves, 2010; Hebeish & Ibrahim, 2007; Liu & Dong, 2002; Wong, Yuen, Leung, Ku, & Lam, 2006). The high durability of the imparted functional properties reflects the positive impacts of the large surface area along with the high surface energy of nanomaterials thereby ensuring better affinity for treated fabrics as well as higher durability of textile functions compared with the conventional materials (Becheri, Durr, Nostro, & Baglionl, 2008; Lee, Yeo, & Jeong, 2003).

On the other hand, it is well known that pigment printing is the most economical printing process and can be applied to all substrates. To attain an eco-friendly pigment prints, solvent-free thickening agents and low or zero-formaldehyde binding agents have been used along with other proper additives, e.g. crosslinker, softener, active ingredients, catalyst, etc. (Ibrahim, El-Zairy, Zaky, & Borham, 2005).

The main goal of the current research is to develop a new and effective printing method for enhancing the antibacterial activity

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of pigment printed cellulose-containing fabrics via incorporation of certain antibacterial agents such as chitosan, choline chloride, Invasan®, HBPAA/Ag-NP's and HBPAA/ZnO-NP's hybrids in the print paste formulations.

2. Experimental

2.1. Materials

Plain weave mill-scoured and bleached cotton/polyester (50/50, 118 g/m^2) and (35/65, 123 g/m^2) blends were used throughout the study.

Printofix® Binder MTB-01 liquid (acrylate based copolymer, anionic, Clariant); GB Binder®FMD (based on polyacrylate, anionic, BASF/GB Chem, Egypt) Printofix® Thickener 160 EG liquid (synthetic thickening agent based on ammonium polyacrylate, Clariant); GB Resin®CPN (based on hydroxymethylated 4,5-dihydroxyethylene urea, BASF/GB Chem, Egypt); Durex® Silicone-1020 (based on modified polysiloxane microemulsion, Texchem, Egypt); Printofix®Red H3BD pigment, Printofix®Green HXP (Clariant); Imperon® Royal Blue SP pigment (DyStar), Unisperse® Yellow MR pigment (Ciba), and Invasan® (Triclosan derivative, Huntsman, USA) were of commercial grade.

All of other chemicals namely, chitosan (degree of deacetylation of >85%, Sigma), Silver nitrate ($AgNO_3$, Sigma), zinc acetate ($Zn–(CH_3COO)_2{\cdot}2H_2O$, Aldrich); choline chloride (Aldrich), acetic acid, sodium hydroxide and ammonium persulphate [$(NH_4)_2S_2O_8$] were reagent grade.

2.2. Methods

2.2.1. Synthesis of the hyperbranched poly amide-amine (HBPAA)

HBPAA was synthesized using Ibrahim et al. method (Ibrahim, Mahrous, et al., 2010).

2.2.2. Synthesis of HBPAA/Ag-nanoparticles hybrid (HBPAA/Ag-NP's)

Synthesis of HBPAA/Ag-NP's hybrid was similar to that reported in the literature (Ibrahim, Eid, Youssef, et al., 2012).

2.2.3. Synthesis of HBPAA/ZnO-NP's hybrid

The HBPAA (3.1 g) was dissolved in distilled water (80 ml)/ethyl alcohol (20 ml) mixture at ambient temperature. To the solution the pre-dissolved 1.24 g Zn–acetate was added drop wise with continuous stirring. After 30 min of reaction the pre-dissolved 0.45 g NaOH was added drop wise with continuous stirring to the mother liquor and the reaction mixture was kept under stirring for further 30 min.

2.2.4. Pigment printing

Guide formulation for solvent-free pigment printing of cotton cellulose/polyester using flat screen technique with two Storkes follows:

Printing paste components	g/kg
Pigment colorant	20
Synthetic thickener agent	20
Binding agent	100
Crosslinker	20
Silicone softener	10
$(NH_4)_2S_2O_8$ catalyst	2
Bio-active agent:	
Chitosan	10
Choline chloride	20
HBPAA/ZnO-NP's hybrid	20
HBPAA/Ag-NP's hybrid	20
Or Invasan®	20
Acetic acid	1
Water	817 or 807
Total	1000

Printed fabric samples were then simultaneously dried and fixed in a commercial microwave oven at output power of 386 W/5 min.

2.3. Measurements

Nitrogen content (%N) was determined according to the Kjeldahl method.

The color strength (K/S) values of the obtained pigment prints were determined from the reflectance measurements using the Kubelka Munk equation (Judd & Wyszeck, 1975).

$K/S = (1R)^2/2R$, where K/S is the ratio of absorption and scattering coefficient, and R is the reflectance at the wave length of maximum absorbance of the used pigment colorants.

Fastness properties to washing, rubbing and light of the produced pigment prints were determined according to AATCC Test Methods (61-1972), (8-1972), and (16A-1972), respectively.

Antibacterial activity assessment against G+ve (*S. aureus*) and G−ve bacteria (*E. coli*) was evaluated qualitatively according to AATCC Test Method (147-1988), and expressed as zone of growth inhibition (mm). Durability of the imparted antibacterial activity to washing was evaluated according to ASTM Standard Test Method (D 737-96).

The morphology and particle size of the prepared HBPAA/Ag-NP's and HBPAA/ZnO-NP's hybrids were determined by transmission electron microscope (TEM) using JEOL, JEM 2100F electron microscope at 200 kV.

The surface morphology of HBPAA/Ag-NP's and HBPAA/ZnO-NP's-loaded pigment prints (SEM) were observed with SEM Model Quanta 250 FEG (field emission gun) attached with EDX unit (energy disperse X-ray analysis) with accelerating voltage 30 kV (FEI Co., Netherland).

3. Results and discussion

In this study we focus on improving the antibacterial activity of pigment printed cellulose-containing fabrics via individual inclusion of certain bio-active ingredients namely HBPAA/Ag-NP's hybrid, HBPAA/ZnO-NP's hybrid, chitosan, choline chloride and triclosan derivative in the print paste formulation along with other ingredients. Discussion of the obtained results follows.

3.1. Effect of hybrid type and concentration

The formation of Ag-NP's as well as ZnO-NP's in the HBPAA matrix are confirmed by TEM images of the prepared hybrids (Fig. 1a and b, respectively). Both the TEM images reveal that the prepared hybrids are well dispersed and almost spherical in shape with particles size in the range of 6–18 and 32–35 nm, respectively.

As far as the changes in %N, K/S and ZI values of the obtained pigment prints as a function of the prepared hybrid and its concentration, the data in Table 1 reveal that: (i) inclusion of any of the prepared hybrid in the pigment – printing paste results in an increase in the %N as well as in the K/S values along with an outstanding enhancement in the antibacterial efficacy against G+ve (*S. aureus*) and G−ve (*E. coli*) bacteria, regardless of the used substrate, (ii) the improvement in both the %N, K/S and ZI values follows the decreasing order HBPAA/ZnO-NP's > HBPAA/Ag-NP's > none and (iii) the higher the hybrid concentration, the darker the shade and the better the imparted antibacterial activity.

The enhancement in the aforementioned properties reflects the positive impacts of including the nominated hybrids in the printing