Journal of Molecular Structure 1127 (2017) 74–80

Contents lists available at ScienceDirect

Journal of Molecular Structure

journal homepage: http://www.elsevier.com/locate/molstruc

Synthesis, structural characterization and antibacterial activity of cotton fabric modified with a hydrogel containing barium hexaferrite nanoparticles

Desislava Staneva [a,*], Tatyana Koutzarova [b], Benedicte Vertruyen [c], Evgenia Vasileva-Tonkova [d], Ivo Grabchev [e,**]

[a] University of Chemical Technology and Metallurgy, 1756 Sofia, Bulgaria
[b] Institute of Electronics, Bulgarian Academy of Sciences, 1784 Sofia, Bulgaria
[c] LSIC, Chemistry Department B6, University of Liege, Sart Tilman, B-4000 Liege, Belgium
[d] Institute of Microbiology, Bulgarian Academy of Sciences, 1113 Sofia, Bulgaria
[e] Sofia University "St. Kliment Ohridski", Faculty of Medicine, 1407 Sofia, Bulgaria

ARTICLE INFO

Article history:
Received 30 May 2016
Received in revised form
19 July 2016
Accepted 20 July 2016
Available online 21 July 2016

Keywords:
Nanoparticles
Ultrasound
Hydrogel
SEM
XRD
Antibacterial

ABSTRACT

Barium hexaferrite nanoparticles were synthesized by co-precipitation of Ba^{2+} and Fe^{3+} cations with NaOH under of high-power ultrasound. The nanoparticles were dispersed in an aqueous solution of the hydrogel precursors. This solution was used to impregnate the cotton fabric dyed with a photoinitiator. The composite material $BaFe_{12}O_{19}$ nanoparticles-hydrogel-cotton fabric was prepared by surface initiate photopolymerization under visible light.

The modification of the cotton fabric and uniform distribution of the nanoparticles in the structure of the hydrogel were analyzed by scanning electron microscopy (SEM), IR spectroscopy, X-ray diffraction analysis (XRD), fluorescence and colourimetric analyses. The antibacterial efficacy of the material was evaluated against Gram-negative *Escherichia coli* and *Pseudomonas aeruginosa*.

1. Introduction

In the last years, the resistance of pathogenic bacteria to almost all commercially available antibiotics has led to an increasing interest in finding new antimicrobial agents and identifying new strategies for the treatment of infectious diseases [1,2]. Recently, novel antimicrobial agents have been developed using nanoscale materials [3–5]. Nanoparticles have received great attention due to their unique physical, chemical, and effective biological properties in various fields, including medicine [6,7]. Antimicrobial and *anti*-biofilm effect of different types of nanoparticles, especially metals or metal oxides of zinc, copper, silver, gold, and iron have been reported [8,9]. Magnetic nanoparticles have been widely used in biomedical research because of their biocompatibility and magnetic properties [10,11]. Nanoparticles have been developed as contrast agents for magnetic resonance imaging, hyperthermia agents and as carriers for targeted drug delivery to treat several types of cancer [12–14]. The magnetic nanoparticles as delivery nanosystems are considered effective new tools to tackle the current challenges in treating infections [11].

Barium hexaferrite is one of the most important hard magnetic materials used for permanent magnets, magnetic recording medium and in the creation of microwave components and devices such as circulators, microwaves absorbers, etc. [15,16]. Its widespread use is associated with its properties - high magnetocrystalline anisotropy, high Curie temperature, relatively high saturation magnetization, excellent chemical stability and corrosion resistance [16].

Incorporation or immobilization of magnetic nanoparticles on textile materials is a promising way to obtain materials with magnetic and microwave absorption properties. The possible application of these materials are as flexible magnetic elements in catalysis, for advanced recording, as a part of intelligent clothing for

* Corresponding author.
** Corresponding author.
E-mail addresses: grabcheva@mail.bg (D. Staneva), i.grabchev@chem.uni-sofia.bg (I. Grabchev).

http://dx.doi.org/10.1016/j.molstruc.2016.07.087
0022-2860/

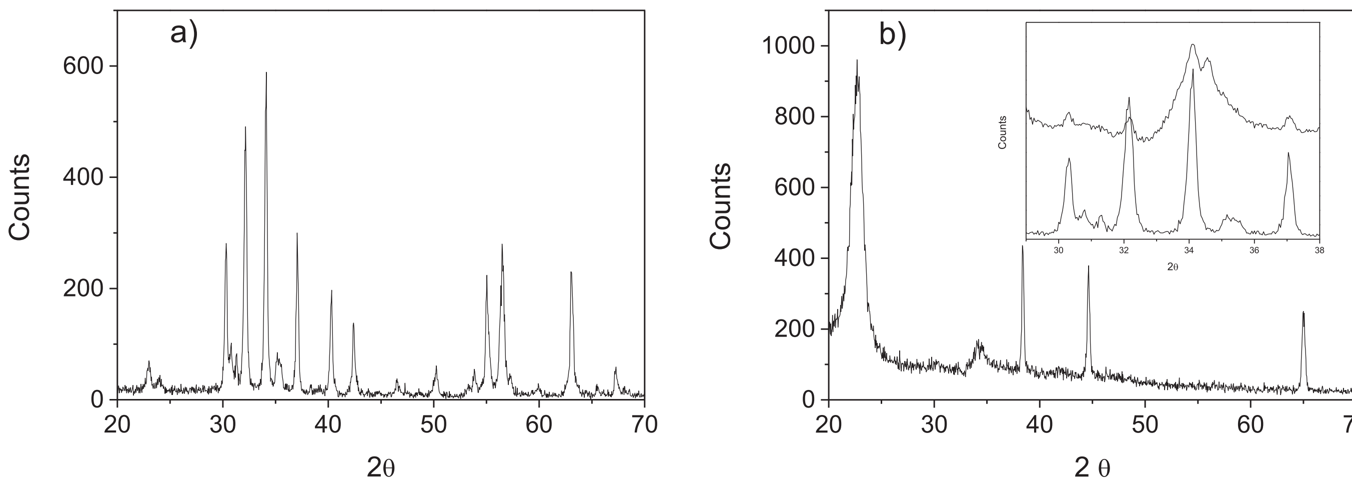

Fig. 1. XRD spectra of (a) nanosized powder of $BaFe_{12}O_{19}$ and (b) modified cotton fabric.

monitoring and protecting in medicine, sport activities and industry [17,18].

Cotton fabric was coated with barium hexaferrite ($BaFe_{12}O_{19}$) doped in a polyaniline film in order to improve the multifunctional and intelligent fabric with magnetic and electroconductive properties. 30 wt % of nanopowders is necessary to obtain fabric with magnetic properties and only 5 wt % of nanopowders for the highest conductivity [19]. M. Grosu et al. [20] have also found the magnetic properties of the woven fabric to depend on the mass percentage of magnetic power in the coating solution. Magnetite nanoparticles (Fe_3O_4) have been used to improve the antibiofilm properties of textile dressing, tested *in vitro* against monospecific Candida albicans biofilms [21]. However, limited information is available for using barium hexaferrite nanomaterials in textiles for antimicrobial purposes.

The aim of this study is to obtain a cotton fabric with uniformly dispersed barium hexaferrite nanoparticles on the fibre surface by means of an acrylamide hydrogel and to assess their antimicrobial activity. FTIR, XRD, SEM, EDX, fluorescence spectroscopy and colourimetry have been used for the characterization of the barium hexaferrite nanoparticles and composite material.

2. Experimental

2.1. Materials

A bleached and unmercerized, plain-woven 100% cotton fabric with a surface weight of 140 g m^{-2} was used throughout the work. Acryl amide (AAm), *N,N′*-methylenebisacrylamide (bis-AAm) and N-methyldiethanolamine (MDEA), $Ba(NO_3)_2$ and $Fe(NO_3)_3 \cdot 9H_2O$ were used as obtained from Aldrich without further purification. Precosolve was obtained from Schill&Seilacher (Germany). 4,4′-bis(1,3,5-triazin-2-ylamino)-stilbene-2,2′-disulfonic acid were synthesized according to a method described previously [22]. All solutions were made with distilled water.

The light sources for photopolymerization were two energy saving lamps:

1. HL 8325, 25 w, 1230 Lumen, 6400 K, Horoz Electric
2. VO 11211, 11 w, 550 Lumen, 6400 K, Vitoone

2.2. Preparation of $BaFe_{12}O_{19}$ nanoparticles

$BaFe_{12}O_{19}$ nanoparticles were synthesized by co-precipitation of Ba^{2+} and Fe^{3+} cations with NaOH under high-power ultrasound. $Ba(NO_3)_2$ and $Fe(NO_3)_3 \cdot 9H_2O$ were dissolved in distilled water at a ratio Ba:Fe = 1:10, which was suitable to obtain monophasic powder from $BaFe_{12}O_{19}$. The ratio was not stoichiometric Ba:Fe = 1:10, because of the high solubility constant of the precipitate of $Ba(OH)_2$ - $K_s = 10^{-3.6}$ at 25 °C, which determined its instability in an aqueous solution [23–25].

The solution was homogenized with a magnetic stirrer (*Boeco MMS-3000)* for 2 h. An ultrasonic probe (VCX750 Ultrasonic Processor at 750 W) was placed in the solution and concentrated NaOH was adding dropwise until complete co-precipitation of cations. The resulting precipitate was centrifuged and dried at 100 °C. The precursor thus obtained was ground, placed in a corundum boat and subjected to high temperature treatment to obtain single-phase nano-sized $BaFe_{12}O_{19}$. The optimal parameters of the process were pH = 11.5 and 120 min ultrasound treatment with pulse duration 2 s to every 4 s and amplitude - 40%. In this way the temperature for the preparation of a single-phase powder of nanosized $BaFe_{12}O_{19}$ could be lowered by 100 °C as compared to a previously known technology for the synthesis of this material. The high-temperature synthesis was performed at 800 °C for 4 h in a tube furnace *Carbolite*.

2.3. $BaFe_2O_{19}$ nanoparticles-hydrogel-cotton fabric preparation

The procedure for cotton surface functionalization involved dyeing with the modified photoinitiator eosin Y (MEY) (step 1) and subsequent hydrogel formation with dispersed magnetic nanoparticles by surface initiated photopolymerization under visible light (step 2). The MEY was used as photo-sensitizer and methyldiethanolamine (MDEA) as co-initiator. The synthesis of MEY and dyeing of cotton fabric was previously reported [26]. The concentration of the MEY was 1.0% owf.

The deposition of nanoparticles and hydrogel formation was performed via the following procedure:

$BaFe_{12}O_{19}$ (barium hexaferrite) nanoparticles (6 wt % to AAm and bis-AAm) were added to an aqueous solution of surfactant