Journal of Environmental Chemical Engineering 5 (2017) 5786–5792

Contents lists available at ScienceDirect

Journal of Environmental Chemical Engineering

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

Research paper

An antibacterial study of a new magnetite silver nanocomposite

Seyed Amir Hossein Jalali[a,b,*], Alireza Allafchian[c], Hamid Bahramian[c], Razieh Amiri[c]

[a] Department of Natural Resources, Isfahan University of Technology, Isfahan 84156-83111, Iran
[b] Research Institute for Biotechnology and Bioengineering, Isfahan University of Technology, Isfahan 84156-83111, Iran
[c] Research Institute for Nanotechnology and Advanced Materials, Isfahan University of Technology, Isfahan 84156-83111, Iran

ARTICLE INFO

Keywords:
Silver
Poly(ethylene terephthalate)
Antibacterial
Nanocomposite

ABSTRACT

A new silver magnetite nanocomposite $Co_{0.7}Zn_{0.3}Fe_2O_4$/Poly(ethylene terephthalate)/Silver ($Co_{0.7}Zn_{0.3}Fe_2O_4$/PET/Ag) was synthesized for its antimicrobial activity investigation against gram-negative and −positive bacteria. At first, $Co_{0.7}Zn_{0.3}Fe_2O_4$ nanoparticles were prepared and covered by the PET polymer; then silver nanoparticles were decorated on the surface of the PET shell. Subsequently, some spectroscopic techniques such as FT–IR, XRD, TEM and SEM were applied to characterize them; and the VSM method was used to determine the level of the magnetic power of these nanocomposites. The FTIR spectra of synthesized nanocomposites indicated that $Co_{0.7}Zn_{0.3}Fe_2O_4$ nanocomposite was firstly composed and functional groups appearance including carbonyl and ester groups' emphasis to covering core magnetic with PET. The comparison of XRD pattern of synthesized nanocomposites showed that coating $Co_{0.7}Zn_{0.3}Fe_2O_4$ nanoparticles with PET and Ag nanoparticles, respectively, had no effect on the spinel structure of the magnetic core. In addition, the Ag nanoparticles structure in the target nanocomposite structure had an *fcc* symmetry. The SEM images have presented a cloudy and spongy porous cluster for fabricated nanomaterials; and the spherical shapes of core magnetite and Ag nanoparticles; and PET layer were approved by TEM pictures. The bactericidal potential of the synthesized nanocomposites was evaluated by using the disk diffusion process and measuring the minimum inhibitory concentration (MIC) and the minimum bactericidal concentration (MBC) quantities against the two different groups of bacteria. The results showed that $Co_{0.7}Zn_{0.3}Fe_2O_4$/PET/Ag nanocomposites had a high antibacterial potential. In addition, by applying the magnetic field, they could be separated from the media after the disinfection procedure.

1. Introduction

Waterborne diseases including gastrointestinal diseases, diarrhea and systematic illnesses caused by different agents are a global health threat leading to mortality and the high costs of their prevention and treatment. A wide variety of organisms including viruses, bacteria, protozoa and helminths can contaminate water. Gram negative bacteria (the main cause) including *Salmonella typhimurium*, *Escherichia coli*, *Vibrio cholerae*, *Legionella* and *Campylobacter jejuni,* and some gram positive bacteria including *Bacillus cereus* and *Staphylococcus aureus* have been reported as causative agents of water contamination from different countries. Therefore, improving the new methods and materials for water disinfection seems to be necessary [1–3].

Most researchers have recognized the high antimicrobial activity of silver nanoparticles (Ag NPs) among other metal nanoparticles. The Ag NPs aggregate in the media with a high electrolyte potential, reducing the antibacterial property [4]; some supporting materials such as zeolite, titania, carbon, ferrites and polymers are applied to the solution of these nanoparticles with nanocomposite formation [5–8]. Polyethylene, polydopamine, polyimide, poly(vinyl alcohol) and polypyrrole polymers have been successfully applied as the supporting compounds [9–13].

One of the practical means to prevent Ag NPs releasing is loading Ag NPs as the second layer of a three-part nanocomposite to form core/shell/Ag NPs composites. Since polymers have such beneficial mechanical properties as low cost, high transparency, high process ability, industrial applications, and moderate recyclability, they are used as the shell of nanocomposite structures like poly acrylonitrile co maleic anhydride (PAMA), polyaniline (PANI), polydopamine (PDA) and polyimide (PI) [14–18]. Poly(ethylene terephthalate) (PET) is one of the aromatic polyesters extensively used as a kind of polyester resin in the conventional industry. This polymer holds a good potential for industrial application, including industrial fibers, films, bottles, and engineering plastics [19,20]. In this regard, many studies have been conducted to find the commercial applications of aromatic polyesters and their composites, such as a high performance polymer [21,22].

* Corresponding author at: Research Institute for Biotechnology and Bioengineering, Department of Natural Resources, Isfahan University of Technology, Isfahan 84156–83111, Iran.
E-mail addresses: sahjalali@cc.iut.ac.ir (S.A.H. Jalali), Allafchian@cc.iut.ac.ir (A. Allafchian), hamidbahramian68@gmail.com (H. Bahramian), razieh.amiri@gmail.com (R. Amiri).

http://dx.doi.org/10.1016/j.jece.2017.11.016
Received 22 July 2017; Received in revised form 31 October 2017; Accepted 3 November 2017
Available online 04 November 2017

Generally, Fe_3O_4 magnetite nanoparticles are used as the core in nanocomposite structures because they can be easily removed from the media by applying an external magnetic field after water treatment [23–26]. Therefore, one of the best options for nanocomposite selection with a core/shell/Ag NPs structure can be Fe_3O_4/polymer/Ag NPs. But, one of the important problems in using the ferrite core is corrosion, especially in water disinfection. The application of nano alloy ferrites (XFe_2O_4) is the solution to decrease the corrosion of nanoferrite, especially because it can be a more economically appropriate candidate than Fe_3O_4 for industrial applications [27,28]. Also, in order to increase the magnetic properties of ferrite alloy nanocomposites, such as saturation magnetization (M_s), remanence magnetization (M_r) and coercivity (H_c), nano alloy ferrites with $X_{a\%}Y_{b\%}Fe_2O_4$ structure have been utilized. Wang et al. synthesized $Ni_{0.8}Zn_{0.2}Fe_2O_4$, reporting the specific saturation magnetization, remanence magnetization and coercivity of 63 emu/g, 36 emu/g and 2750 G, respectively [29]. Also, Khairy, by preparing the Polyaniline–$Zn_{0.2}Mn_{0.8}Fe_2O_4$ composite, indicated that the adsorption efficiency of the materials in removing a toxic dye from wastewater was increased [30]. Sendhilnathan et al. also showed that the magnetic parameters M_s, M_r and H_c of $Co_{a\%}Zn_{b\%}Fe_2O_4$ were increased with decreasing the zinc percent of the nanocomposite [31].

This paper was intended to provide a complete overview of synthesis, characterization, and antibacterial effect of the silver magnetic core–shell nanocomposite, $Co_{0.7}Zn_{0.3}Fe_2O_4$/PET/Ag NPs. The structure of the produced magnetic core-shell nanocomposite was characterized by utilizing FT–IR, XRD, SEM and TEM techniques. In addition, the magnetic properties of the nanocomposites were assessed through VSM testing; also, the antibacterial activity of it was studied by using the disk diffusion method against some gram positive and gram negative bacteria. It was demonstrated that the antibacterial properties of the produced magnetic core-shell nanocomposite were enhanced, as compared to those of silver nanoparticles. These nanocomposites could also be easily removed from the medium by utilizing an external magnetic field, ultimately resulting in an easier and greater disinfection process.

2. Experimental

2.1. Materials and methods

The citric acid, dimethyl sulfoxide (DMSO) and other used materials such as NaOH, NH_3, $Zn(NO_3)_2$, $Co(NO_3)_2$, $Fe(NO_3)_3$, $Na(BH_4)$, $Ni(NO_3)_2$ and $AgNO_3$ were prepared from Merck Co. (Germany). Deionized and doubly distilled water were used as a solvent in this study. The FT–IR Jasco Japan spectrophotometer was used for scanning the Fourier transform infrared (FT–IR) spectrum. 1 to 2% of each sample was mixed with KBr and press powdered to take the transparent pellet. The pellet was placed in the spectrometer to obtain a spectrum in 400–4000 cm^{-1}. The X–ray diffraction (XRD) profiles of the produced samples were achieved by using a Philips X–ray diffractometer. These patterns were gained by applying Cu K_α radiation (λ¼ 0.154 nm), from 10° to 80°, for the 2θ. The Hitachi Japan S4160 field emission scanning electron microscope was used for scanning the electron microscopy (FE–SEM) images of the prepared samples. The transmission electron microscopy (TEM) was obtained by applying a Philips CM10-HT 100KV microscope for the investigation of the morphology and the size of the as–synthesized particles. By using a vibrating sample magnetometer (VSM) from Meghnatis Daghigh Daneshpajouh Company (Iran), the magnetic properties of the prepared nanocomposites were scanned. Single–point Brunauer–Emmett–Teller (Chemisorption analyzer (TPR, TPD, TPO) and BET surface area were considered; Toseye Hesgarsazan Asia Co., Iran) method was used for calculating the surface area of the synthesized nanomaterials.

2.2. Synthesis of $Co_{0.7}Zn_{0.3}Fe_2O_4$

At first, 4.04 g of ferric nitrate, 0.8 g of cobalt nitrate, 0.44 g of zinc nitrate, and 1.23 g of citric acid were dissolved separately in 20 mL of distilled water. The solutions were then mixed and by adding 1 mL ammonia solution; the pH of the new solution was adjusted to 9.0. Then, the solution was stirred by a magnetic stirrer for 2 h and placed in an oven at 100 °C for 24 h. Finally, the produced material was baked in the furnace (Yaran Behgozin Parsa Co., model: YTF 1450–30 × 8, Iran) at 400 °C for 2 h, under argon atmosphere.

2.3. Preparation of Ag nanoparticles

There are different procedures for the synthesis of Ag nanoparticles [32–35]. In this research, the one described method in Ref. [34] was used for the aim of the study. To summarize it, 15 mL of the silver nitrate solution 0.1 M was added drop wise to 15 mL of sodium borohydrate 0.1 M in which the sodium:silver molar ratio was 1:1. After 2 h, the silver nanoparticles were synthesized and washed with distilled water several times to eliminate any excess protecting agent. The final product was dried in the oven at 50 °C for 2 h.

2.4. Synthesis of $Co_{0.7}Zn_{0.3}Fe_2O_4$/PET nanocomposite

0.205 g of poly(ethylene terephthalate) (PET) polymer was dissolved in 2 mL of DMSO. Then, 1.0 g of $Co_{0.7}Zn_{0.3}Fe_2O_4$ was added to the PET solution and put in an ultrasonic bath for 1 h. After the evaporation of DMSO from the samples, they were transferred in a porcelain mortar and powdered.

2.5. Preparation of $Co_{0.7}Zn_{0.3}Fe_2O_4$/PET/Ag nanocomposite

0.205 g of PET was dissolved in 2 mL of distilled DMSO; then 1.0 g of the synthesized $Co_{0.7}Zn_{0.3}Fe_2O_4$ was added to it. After that, this prepared sample was mixed with 0.12 g of Ag nanoparticles by a mechanical stirrer for 2 h; it was then sonnicated for 15 min. After 5 min, the gained precipitate was filtered, washed two times with distilled water, and dried under the atmosphere temperature.

2.6. Antimicrobial tests of the nanocomposite

The standard method Kirby–Bauer disk diffusion was applied for the evaluation of the microbicidal activity of the synthesized nanocomposites [36]. The details of this method have been described in a previous study [37]. To put it briefly, *Salmonella typhimurium* (ATCC14028) and *Escherichia coli* (ATCC35218) bacteria, for gram negative assessment, and *Bacillus cereus* (ATCC14579) and *Staphylococcus aureus* (ATCC29213) bacteria, for gram positive assessment, were selected and suspended in the phosphate buffer saline (PBS). The turbidity of the bacterial suspensions was attuned to the 0.5 McFarland standard, and sterile swabs were dipped into the inoculum tubes. 0.1 mg from $AgNO_3$, Ag NPs, $Co_{0.7}Zn_{0.3}Fe_2O_4$NPs, $Co_{0.7}Zn_{0.3}Fe_2O_4$/PET and $Co_{0.7}Zn_{0.3}Fe_2O_4$/PET/Ag nanocomposite suspensions was dissolved in 1.0 mL distilled water, separately. Then, after the 10 min sonication of them, the four standard disks were prepared, and the distilled water was used as a negative control. At the end, the impregnated–disks were utilized to evaluate the antibacterial activity on Mueller–Hinton agar (Merck, Germany) plates. The disks were placed on the surface of the agar using the sterile forceps. Then, the plates were incubated at 37 °C for 24 h to check the antimicrobial activity. The antibacterial activity assays were carried out triplicate (the zone of inhibition) and quantified in the millimeter unit.

The content of bacteria colonies was adjusted to 10^6 CFU (colony forming units)/mL for MIC experiments. The concentrations of the $Co_{0.7}Zn_{0.3}Fe_2O_4$/PET/Ag nanocomposite was diluted in a twofold manner, with 1000, 500, 250, 125, 62.5 and 31 μg/mL in the Phosphate Buffer Saline solution. Then, an identified value of any nanocomposite solutions was added to 180 μL of the nutrient broth in tubes containing diluted bacterial suspensions (10^6 CFU/mL) and incubated at 37 °C for