Bull. Mater. Sci., Vol. 40, No. 1, February 2017, pp. 147–155. © Indian Academy of Sciences.
DOI 10.1007/s12034-016-1357-x

Enhanced antibacterial performance of Fe_3O_4–Ag and $MnFe_2O_4$–Ag nanocomposites

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MS received 6 May 2016; accepted 13 June 2016

Abstract. **In this work, we have described the antibacterial activities of Fe_3O_4 nanoparticles with different organic parts, including Humic acid (HA), Nicotinic acid (Nico) and Histidine (His), and the antibacterial activity of $MnFe_2O_4$ nanoparticles coated with PANI and SiO_2 against different bacteria and some standard antibacterial drugs. The present study revealed that the newly fabricated various Fe_3O_4 and $MnFe_2O_4$ nanocomposites, when combined with some different organic parts, are superiour antibacterial agents. Also, the synthesized nanocomposites can be easily separated from aqueous solution by magnetic filtration without any contamination of the medium.**

Keywords. **Antibacterial property; magnetic nanoparticles; silver.**

1. Introduction

For over two decades, developments of organic parts integrated with magnetic nanoparticles have been reported because of their promising properties that are difficult to obtain separately with the individual components [1,2]. Many researchers have been trying to fabricate new composite materials by modifying the magnetic surface with organic molecules or inorganic parts, such as SiO_2, PANI, APTES, caffeic acid, folic acid, etc., since such new hybrid nanostructures have significant properties due to the their unique multifunctional applications in industrial and biomedical fields [3–9].

Antibacterial materials are of great importance and necessity in our daily life. Many researchers are trying to develop and design these kinds of materials, which can be used very efficiently because of low cost for better antibacterial performance. For instance, Wang *et al* [10] prepared and developed an antibacterial and high light transmittance bulk materials to incorporate the hydrophobic and hydrophilic antibiotics [10]. In another work, Majumdar *et al* [11] developed an antibacterial material using polyester–silver nanocomposite fibres. Furthermore, Yadav *et al* [12] synthesized nickel-doped TiO_2 nanoparticles and reported visible light photocatalytic antibacterial activity against Gram-positive and Gram-negative bacteria. The nanomaterials, including silver, copper and some other metal or metal-based materials, are also efficient bacteriostatic agents [13]. Silver nanoparticles are reported to bear antimicrobial property against an array of pathogenic microorganisms but in order to improve the antibacterial activity of Ag NPs, some supports are needed. The combination of surface properties of Ag NPs and magnetic properties of magnetite leads to the potential applications of these magnetic nanocomposites in biological targeting, biological separation, high-density magnetic recording, catalysis and targeting therapy [14]. In addition, the introduction of silver nanoparticles into Fe_3O_4 enhances the biological activity of Ag NPs, but many studies have shown that the recovery of silver-based antibacterial agents is a general problem since they have a potential toxicity for both human and aqua systems and thus removing them from the disinfected media is efficiently required. The striking feature of Fe_3O_4 nanocomposites combined with silver nanoparticles is that they can be easily separated using an external magnet, providing a simple separation of the nanocomposite [2,15]. Finally, using these materials for killing bacteria under a magnetic field, we can expect a short sample preparation time and an easy cleanup procedure.

Here, we combined the collective advantages of magnetite that is coated with some organic molecules like Humic acid (HA), Nicotinic acid (Nico) and Histidine (His) with Ag nanoparticles and $MnFe_2O_4$ coated with PANI and SiO_2 with Ag NPs to produce multifunctional materials showing novel, good antibacterial activity and magnetic separability. Five magnetic nanocomposites, i.e., Fe_3O_4@HA@Ag, Fe_3O_4@His@Ag, Fe_3O_4@Nico@Ag, $MnFe_2O_4$@PANI@Ag and $MnFe_2O_4$@SiO_2@Ag, have been reported for the first time for their antibacterial activity against Gram-positive and Gram-negative bacteria in this study.

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2. Experimental

2.1 *Synthesis of magnetic nanocomposites*

The detailed experimental procedure for all products is given in our previous reports [16–21] except for the Fe_3O_4@HA@Ag magnetic nanocomposite, which is newly synthesized.

2.1a *Preparation of Fe_3O_4@HA magnetic nanocomposites*: Fe_3O_4@HA nanocomposite was prepared by a simple reflux method. For synthesis of Fe_3O_4@HA nanocomposite, $FeCl_3{\cdot}6H_2O$ and $FeCl_2{\cdot}4H_2O$ salts were taken in 1:2 ratio and dissolved in 50 ml of distilled water in a three-neck round-bottom flask and their homogeneous solution was prepared using magnetic stirring. Then 0.2 g of HA was added to this mixture under constant stirring. After that concentrated NH_3 solution was added drop-wise to adjust the pH level of the solution up to 10, at which the precipitation of all ferrites takes place. Then the flask was transferred to the heating mantle, where it was refluxed under Argon at 80°C for 2 h. The synthesized Fe_3O_4@HA nanocompistes were separated using a permanent magnet and washed with distilled water several times to remove impurities. Finally a black powder product was obtained, which was dried at 80°C for 4 h.

2.1b *Preparation of Fe_3O_4@HA@Ag magnetic nanocomposites*: The prepared Fe_3O_4@HA nanocomposite (150 mg) was dispersed in 50 ml of deionized water. Then it was sonicated for 1 h, followed by the addition of 30 ml of $AgNO_3$ solution (0.2 mmol l^{-1}). The solution was vigorously stirred for 1 h, then 0.6 g of $NaBH_4$ was quickly added and the mixture was allowed to react for 2 h under rapid stirring. The product was separated magnetically and washed several times with deionized water to eliminate impurities.

2.2 *Antibacterial study*

2.2a *Bacterial strains*: Several doses of Fe_3O_4, Fe_3O_4@HA, Fe_3O_4@HA@Ag, Fe_3O_4@His, Fe_3O_4@His@Ag, Fe_3O_4@Nico,

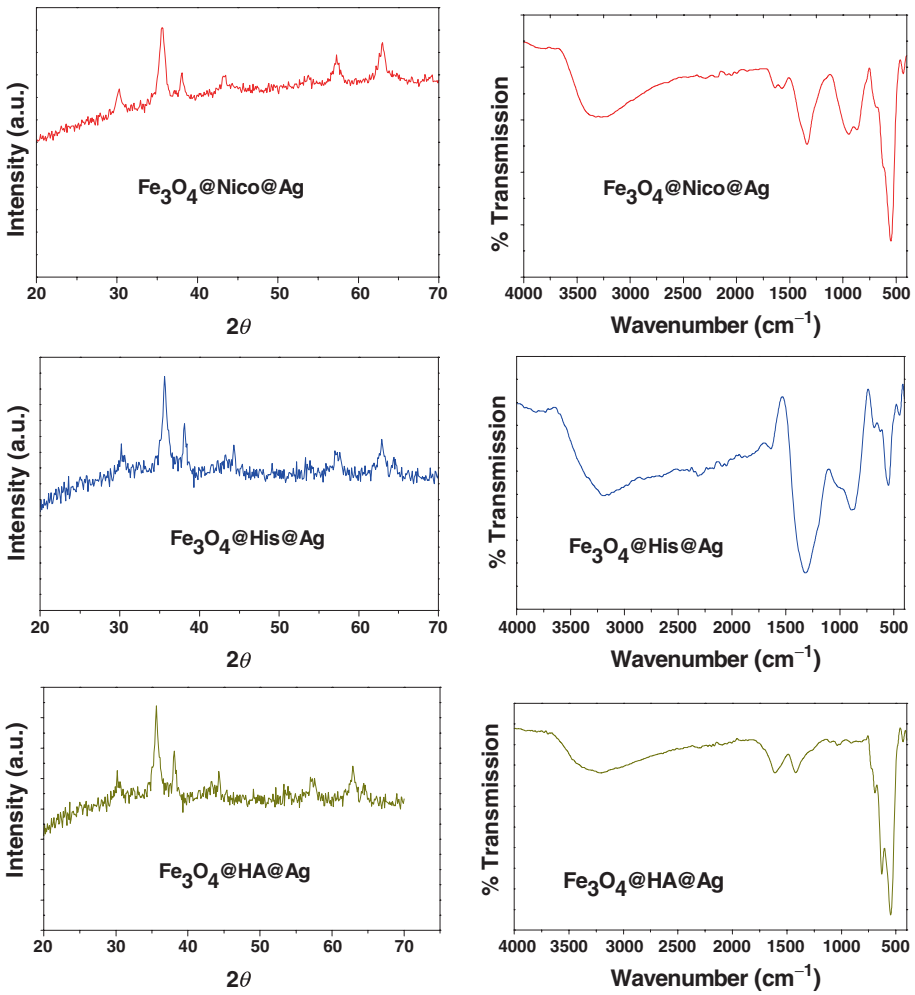

Figure 1. XRD and FT-IR results of Fe_3O_4@Nico@Ag, Fe_3O_4@His@Ag and Fe_3O_4@HA@Ag magnetic nanocomposites.

Fe_3O_4@Nico@Ag, $MnFe_2O_4$, $MnFe_2O_4$@PANI, $MnFe_2O_4$@PANI@Ag, $MnFe_2O_4$@SiO_2 and $MnFe_2O_4$@SiO_2@Ag in sterile distilled water were prepared. All these agents were examined against both Gram-positive bacteria such as *Staphylococcus aureus* (ATCC®25923™), *Bacillus subtilis* (ATCC®31785™) and *Enterococcus faecalis* (ATCC® 29212™) and Gram-negative bacteria such as *Escherichia coli* (ATCC®25922™), *Pseudomonas aeruginosa* (ATCC ®27853™) and *Burkholderia cepacia* (ATCC®25416™). All strains were stored at −80°C in stocks containing glycerol (2.5 M).

2.2b *Agar well diffusion assay*: In this study, four concentrations including 5, 10, 20 and 40 mg ml^{-1} of Fe_3O_4, Fe_3O_4@HA, Fe_3O_4@HA@Ag, Fe_3O_4@His, Fe_3O_4@His @Ag, Fe_3O_4@Nico, Fe_3O_4@Nico@Ag, $MnFe_2O_4$, Mn Fe_2O_4@PANI, $MnFe_2O_4$@PANI@Ag, $MnFe_2O_4$@SiO_2 and $MnFe_2O_4$@SiO_2@Ag were prepared in sterile water and dispersed by sonication. All the magnetic materials have been screened for antimicrobial activity using the agar well diffusion method by measuring the inhibition zone in mm. The assay was performed in triplicate based on the previous studies work of Sangnoi *et al* [22] with modifications.

Bacteria were first grown in Mueller Hinton agar (MHA; Merck, Germany), and incubated at 37°C for 24 h. Each bacterial strain in sterile saline was adjusted to 0.5 McFarland scale, which corresponds to1.5×10^8 CFU ml^{-1}. The agar well diffusion assay was applied to determine the difference in the antibacterial activity of sample $MnFe_2O_4$, $MnFe_2O_4$@PANI, $MnFe_2O_4$@PANI@Ag, $MnFe_2O_4$@SiO_2 and $MnFe_2O_4$@SiO_2@Ag [23,24]. Briefly, bacterial suspensions were adjusted to 10^7–10^8 CFU ml^{-1} (McFarland turbidity standard no. 0.5) and spread on the MHA plates using a sterile triangular loop. This suspension was spread on MHA medium plates (thickness of approximately 4 mm). Following inoculation, a sterile glass borer was used to punch 5-mm wells into the surface of the agar. These wells were filled with 50 µl aqueous solution of the magnetic materials. Known standard antibiotic was filled into the well as a control at the centre of the plate. All plates were incubated overnight at 37°C. After incubation, microbial growth was determined by measuring the diameter of zone of inhibition. For each bacterial strain, controls were the selected standard antibiotics

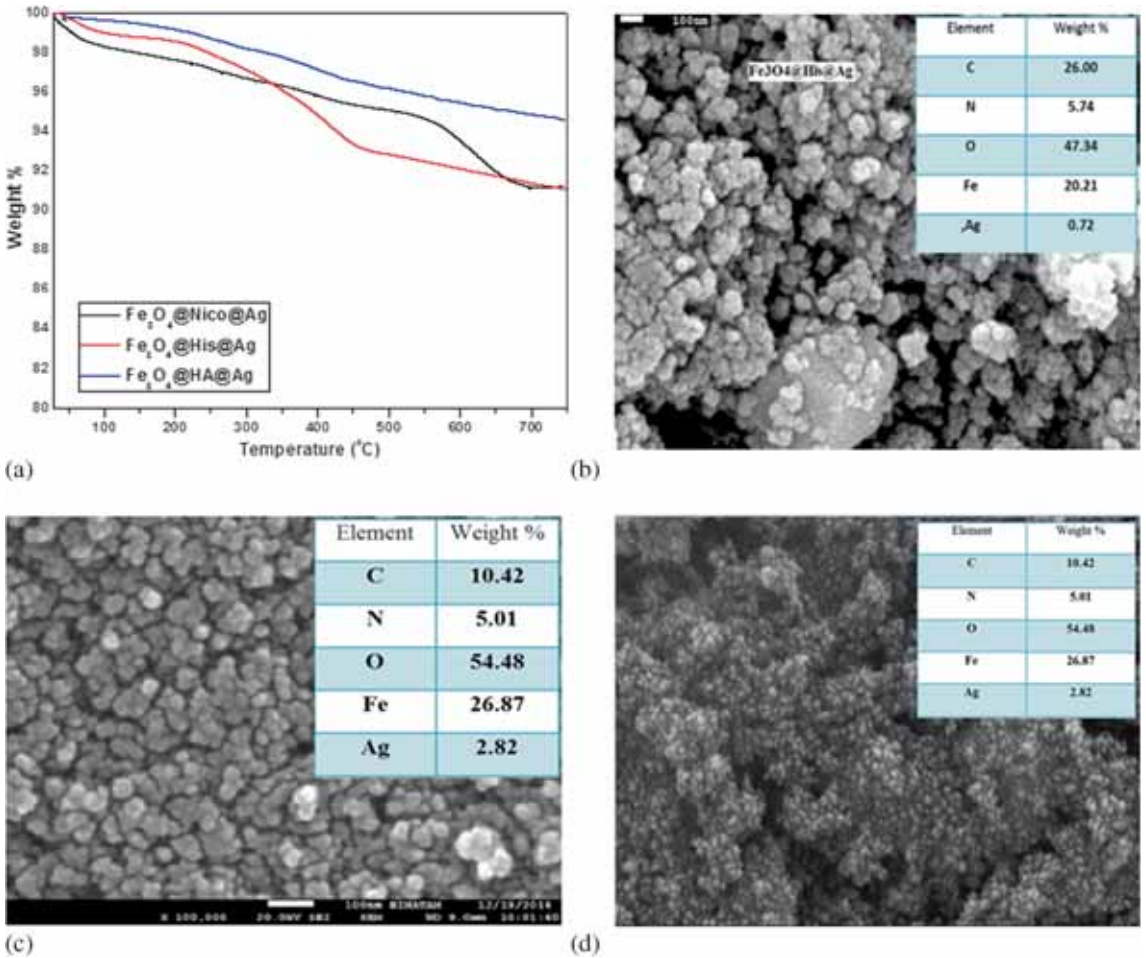

Figure 2. (**a**) TGA results of each magnetic nanocomposites and SEM result of (**b**) Fe_3O_4@Nico@Ag, (**c**) Fe_3O_4@His@Ag and (**d**) Fe_3O_4@HA@Ag magnetic nanocomposites.

such as streptomycin and amoxicillin. In this study, the selection of the standard antibiotics and their concentrations and the evaluation of the antimicrobial activity of the tested materials were based on the CLSI (Clinical and Laboratory Standards Institute) guidelines. The experiment was done three times and the clinical mean values are presented [25].

3. Results and discussion

3.1 *Characterization of nanocomposites*

3.1a *Characterization of Fe_3O_4@Nico@Ag, Fe_3O_4@His @Ag and Fe_3O_4@HA@Ag nanocomposites*: Complete XRD, FT-IR, TGA and SEM characterizations of Fe_3O_4 @Nico@Ag, Fe_3O_4@His@Ag and Fe_3O_4@HA@Ag magnetic nanocomposite were given in our previous studies [10–13]. Briefly, in figure 1, the XRD result of each magnetic nanocomposites confirmed the presence of both Fe_3O_4 ((2 2 0), (3 1 1), (4 0 0), (4 2 2), (5 1 1), (4 4 0)) and Ag ((1 1 1) and (2 0 0)) NPs [10–13], and FT-IR results confirmed the successful modification of the Fe_3O_4 nanoparticles with the organic parts [16–19]. In figure 2a, TGA analysis of each nanocomposites shows the complete degradation of organic part [16–19], SEM analysis of each nanocomposites confirmed the homogeneity of the product (the presence of very tiny nanoparticles) and EDAX analysis indicated the presence of Fe, Ag, C, O and N as the major elements in the product and absence of any impurity (figure 2b–d) [16–19].

3.1b *Characterization of $MnFe_2O_4$@PANI@Ag and $MnFe_2O_4$@SiO_2@Ag nanocomposites*: Similarly full XRD, FT-IR and SEM characterization of magnetic nanocomposite $MnFe_2O_4$@PANI@Ag and $MnFe_2O_4$@SiO_2@Ag were given in our reported study [20,21]. In short, the XRD result of each magnetic nanocomposites confirmed the presence of both $MnFe_2O_4$ ((2 2 0), (3 1 1), (4 0 0), (5 1 1) and (4 4 0) and Ag NPs ((1 1 0), (2 0 0), (2 2 0) [20,21]. FT-IR results confirmed the successful modification of the PANI and SiO_2 with $MnFe_2O_4$ in figure 3 [20,21]. SEM characterization of $MnFe_2O_4$@PANI@Ag and $MnFe_2O_4$@SiO_2@Ag nanocomposites confirmed the homogeneity of the product and EDAX analysis confirmed the presence of Mn, Ag, C, O and N and Mn, Fe, Ag, Si, and O for $MnFe_2O_4$@PANI@Ag and $MnFe_2O_4$@SiO_2@Ag, respectively, as the major elements in the product and the absence of any impurity in figure 4 [20,21].

3.2 *Results and discussion*

3.2a *Antibacterial performance of antimicrobial activities of various Fe_3O_4 nanocomposites with different coated organic parts including HA, His and Nico*: Due to the

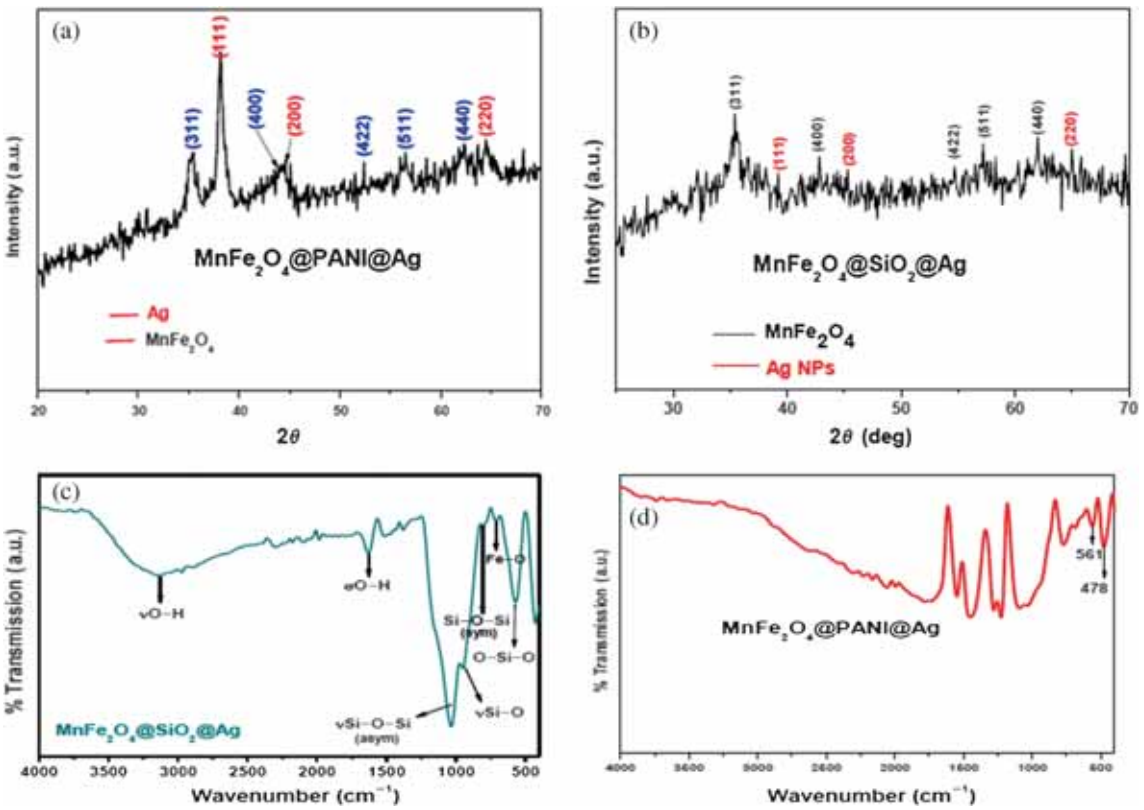

Figure 3. XRD and FT-IR results of $MnFe_2O_4$@PANI@Ag and $MnFe_2O_4$@SiO_2@Ag magnetic nanocomposites.

catalytic activity, optical and electronic properties, antimicrobial activity and magnetic properties, metal nanoparticles have been studied extensively [26]. Despite the fact that there have been many reports in novel studies allocated to studying nano-silver antibacterial activity, the study of silver-coated magnetic nanocomposite still requires investigation. Thus, the antibacterial effect of these nanocatalysts with different concentrations was investigated against both Gram-positive strains such as *S. aureus*, *B. subtilis*, and *E. faecalis* and Gram-negative strains such as *P. aeruginosa*, *E. coli*, and *B. cepacia* using the agar diffusion method. The diameter of inhibition zones (in millimetres) around the wells are shown in table 1.

The antibacterial activities of amoxicillin and streptomycin showed bactericidal effect clearly against the test strains as positive controls. Fe_3O_4 alone had no effect

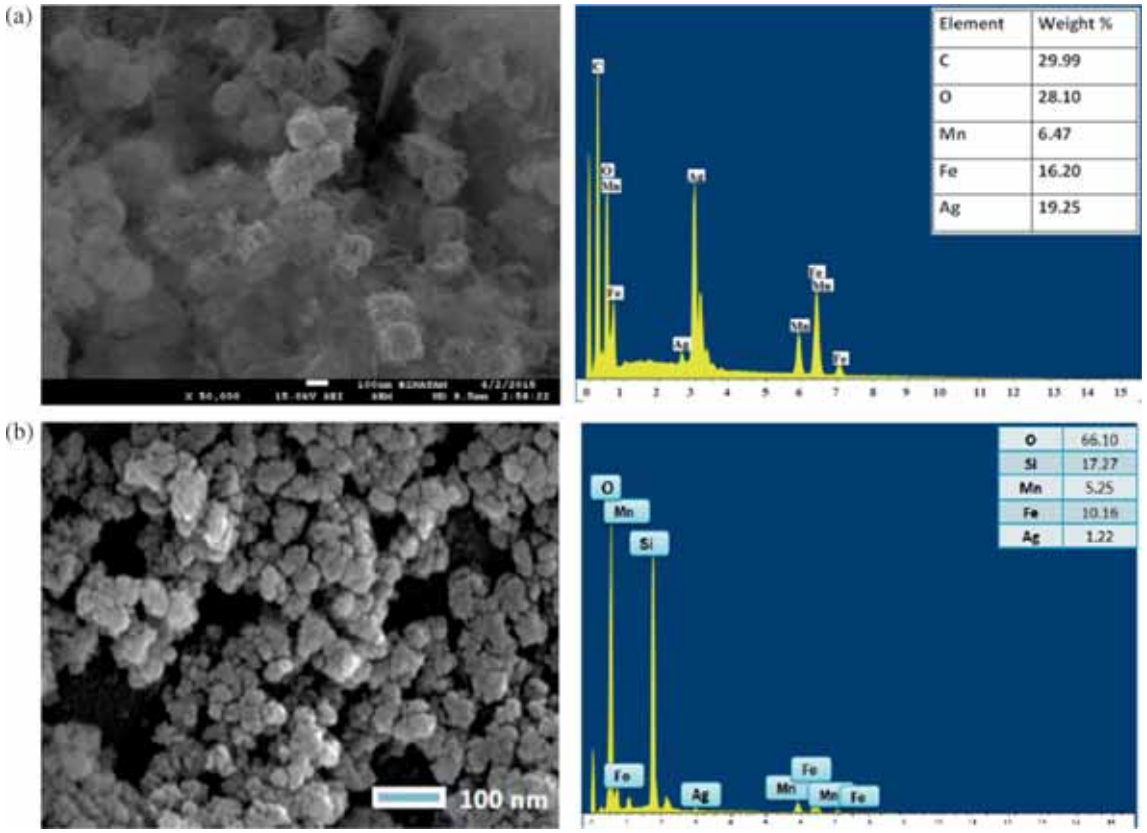

Figure 4. SEM and EDAX results of (**a**) $MnFe_2O_4$@PANI@Ag and (**b**) $MnFe_2O_4$@SiO_2@Ag magnetic nanocomposites.

Table 1. Antimicrobial activities of various Fe_3O_4 nanocomposites with different organic parts including HA, His and Nico coated.

	Microorganisms																							
	Gram-positive												Gram-negative											
Fe_3O_4 nanocomposites	*S. aureus*				*B. subtilis*				*E. faecalis*				*P. aeruginosa*				*E. coli*				*B. cepacia*			
C (mg ml^{-1})	5	10	20	40	5	10	20	40	5	10	20	40	5	10	20	40	5	10	20	40	5	10	20	40
	Zone of inhibition (mm)																							
Fe_3O_4	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
Fe_3O_4@HA	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
Fe_3O_4@HA@Ag	8	9	9	10	8	9	9	11	12	14	15	17	—	9	9	12	—	—	—	10	8	9	9	12
Fe_3O_4@His	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
Fe_3O_4@His@Ag	10	10	11	12	9	10	12	13	16	17	19	20	11	12	13	15	10	10	14	14	10	13	14	16
Fe_3O_4@Nico	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—
Fe_3O_4@Nico@Ag	10	10	11	12	11	10	10	14	12	10	14	15	12	12	15	16	10	10	14	14	13	12	14	16
Amoxicillin (25 mg)	32				27				40				0				30				35			
Streptomycin (10 mg)	22				18				20				20				25				30			

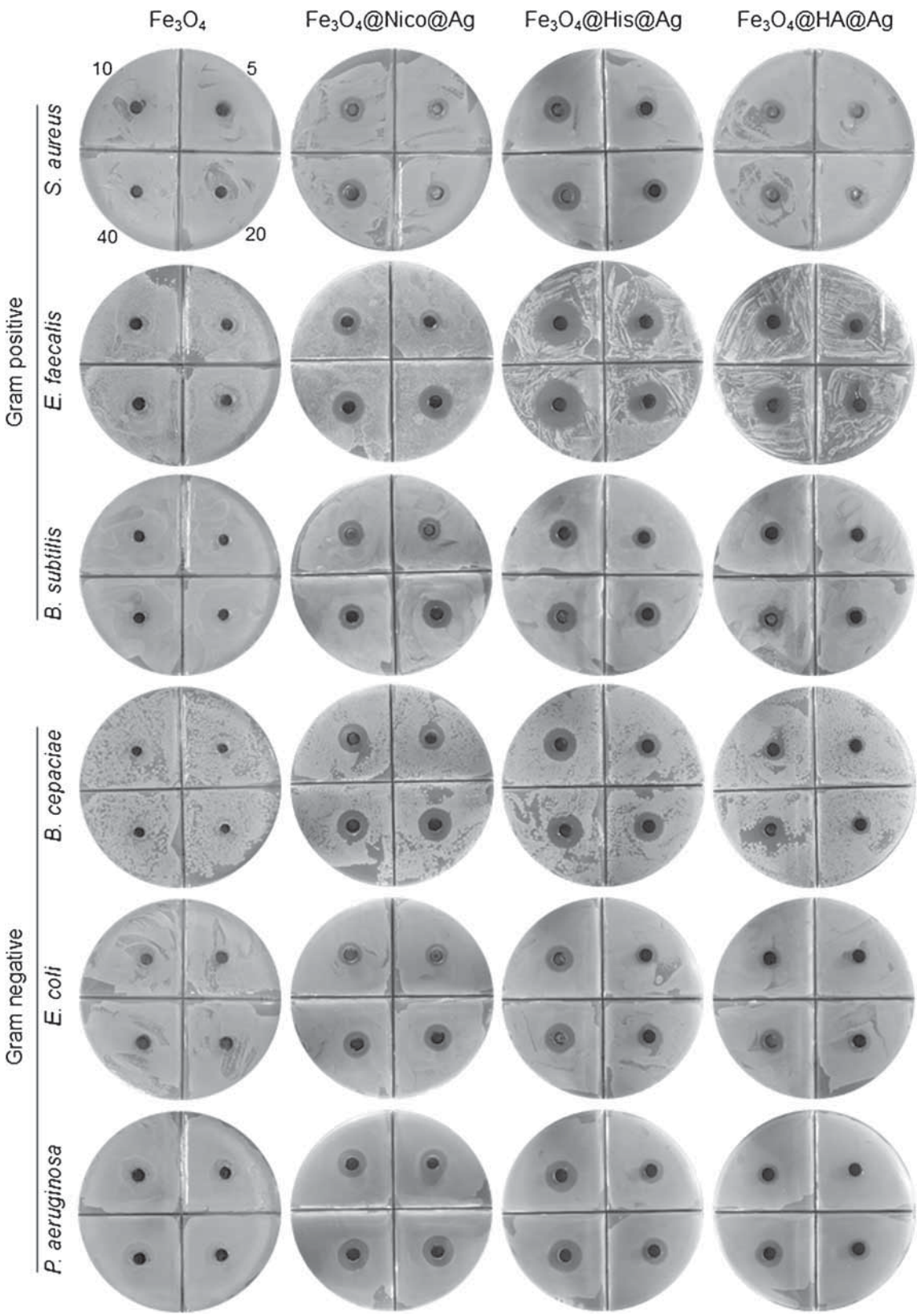

Figure 5. Antimicrobial effects of Fe_3O_4@Nico@Ag, Fe_3O_4@His@Ag and Fe_3O_4@HA@Ag against Fe_3O_4 with different concentrations (5, 10, 20 and 40 mg ml^{-1}) on both Gram-positive and -negative bacteria.

Table 2. Antimicrobial activities of $MnFe_2O_4$@PANI, $MnFe_2O_4$@PANI@Ag and $MnFe_2O_4$@SiO_2@Ag.

	Microorganisms																							
	Gram-positive												Gram-negative											
Nanocatalyst	*S. aureus*				*B. subtilis*				*E. faecalis*				*P. aeruginosa*				*E. coli*				*B. cepaciae*			
C (mg ml^{-1})	5	10	20	40	5	10	20	40	5	10	20	40	5	10	20	40	5	10	20	40	5	10	20	40
	Zone of inhibition (mm)																							
$MnFe_2O_4$	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
$MnFe_2O_4$@SiO_2	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
$MnFe_2O_4$@PANI	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
$MnFe_2O_4$@SiO_2@Ag	10	11	16	15	10	11	13	13	11	12	13	13	11	13	17	15	8	10	12	11	12	13	15	13
$MnFe_2O_4$@PANI@Ag	10	11	13	14	13	11	14	12	12	13	12	14	7	10	13	11	11	12	12	13	12	14	12	15
Amoxicillin (25 mg)	32				27				40				0				30				35			
Streptomycin (10 mg)	22				18				20				20				25				30			

on tested bacteria and also Fe_3O_4@HA, Fe_3O_4@His and Fe_3O_4@Nico showed no antibacterial effects. On the other hand, the effects of Fe_3O_4@HA@Ag, Fe_3O_4@His@Ag and Fe_3O_4@Nico@Ag on the antibacterial activity were observed and the inhibition zones in diameter are presented in figure 5 and table 1. Generally, when the concentration of nanocatalysts including Ag NPs was increased, the antibacterial effects were increased. According to the comparison of the highest concentration of the nanocatalysts (40 mg ml^{-1}), the obtained biggest diameter of inhibition zone (table 1) for Fe_3O_4@HA@Ag was 17 mm against *E. faecalis*, for Fe_3O_4@His@Ag it was 20 mm against *E. faecalis* and for Fe_3O_4@Nico@Ag it was 16 mm against both *P. aeruginosa* and *B. cepacia.*

The lowest concentration of Fe_3O_4@HA@Ag had no antibacterial effect against *P. aeruginosa*; likewise, the concentration of 5, 10 and 20 mg ml^{-1} had no effects against *E. coli*. However, good effect was seen at 40 mg ml^{-1} as compared with amoxicillin antibiotic against *P. aeruginosa*. Fe_3O_4@His@Ag with all its concentrations showed the highest antibacterial effects as strong as that of streptomycin against *E. faecalis* strain among the others. The zone of inhibition was 20 mm, the same for streptomycin and half of the amoxicillin. Fe_3O_4@His@Ag against *E. faecalis* had a good antibacterial activity, the same as that of streptomycin, at the highest concentration of 40 mg ml^{-1}. Fe_3O_4@Nico@Ag with all its concentrations showed the highest inhibition zones against *P. aeruginosa* as 12, 12, 15 and 16 mm, correspondingly, as compared with amoxicillin. This antibiotic had no effect on this strain; however, streptomycin had.

According to obtained results, it is necessary to emphasize that the tested Fe_3O_4 nanocomposites including Ag NPs have shown generally bacteriostatic effects but sometimes bactericidal because Ag NPs have been known to have inhibitory and bactericidal effects [26,27]. The different susceptibilities of Gram-negative and Gram-positive strains towards these examined antibacterial agents possibly depend on their cell wall structure. The mechanism of the bactericidal effect of Ag may be due to its attaching to the surface of the cell membrane and disrupting the permeability and respiration. Various concentrations required for growth inhibition or killing bacteria were determined by the biological properties of individual bacterial species.

3.2b *Antibacterial performance of antimicrobial activities of various $MnFe_2O_4$@PANI@Ag and $MnFe_2O_4$@SiO_2@Ag nanocomposites*: Additionally, $MnFe_2O_4$, which is also a superparamagnetic nanoparticle, with some modified nanocomposites such as $MnFe_2O_4$@PANI, $MnFe_2O_4$@PANI@Ag, $MnFe_2O_4$@SiO_2 and $MnFe_2O_4$@SiO_2@Ag, was also investigated in this study in terms of bactericidal and bacteriostatic effects against both Gram-positive and Gram-negative bacteria determined by the same agar well diffusion method and the zone of inhibition evaluated as presented in table 2. Amoxicillin and streptomycin are the broad-spectrum antibiotics that are used as positive control. $MnFe_2O_4$ nanoparticle alone had no effect on tested bacterial strains and the coated $MnFe_2O_4$@PANI and $MnFe_2O_4$@SiO_2 nanocomposites also showed no antibacterial effects. However when Ag nanoparticles were added to these composites, antibacterial activity was observed positively and the zone of inhibition around the wells on agar was obtained (figure 6).

As observed, $MnFe_2O_4$@SiO_2@Ag nanocomposite had no bactericidal effects on Gram-positive bacteria whereas it possessed bacteriostatic effects as effective as half of that of the antibiotics. There have been four different concentrations of $MnFe_2O_4$@SiO_2@Ag used: 5, 10, 20 and 40 mg ml^{-1}. Generally the highest antibacterial effects were obtained at a concentration of 20 mg ml^{-1} on Gram-positive strains. On the other hand, $MnFe_2O_4$@SiO_2@Ag showed bactericidal effects only on *P. aeruginosa* as Gram-negative bacterium. This bacterium is resistant to amoxicillin but sensitive to streptomycin. The fact remains that $MnFe_2O_4$@SiO_2@Ag exhibited the highest zone of inhibition on *P. aeruginosa* of 17 mm, then *B. cepaciae* of 15 mm and *E. coli* of 12 mm at a concentration of 20 mg ml^{-1}. We could say that this superparamagnetic nanocomposite was not sensitive against *S. aureus*, *B. subtilis*, *E. faecalis*, *E. coli* and *B. cepaciae* but showed intermediate susceptibility.

For $MnFe_2O_4$@PANI@Ag, nearly the same antibacterial effects on both Gram-positive and Gram-negative bacteia are demonstrated. For this nanocomposite, it was observed

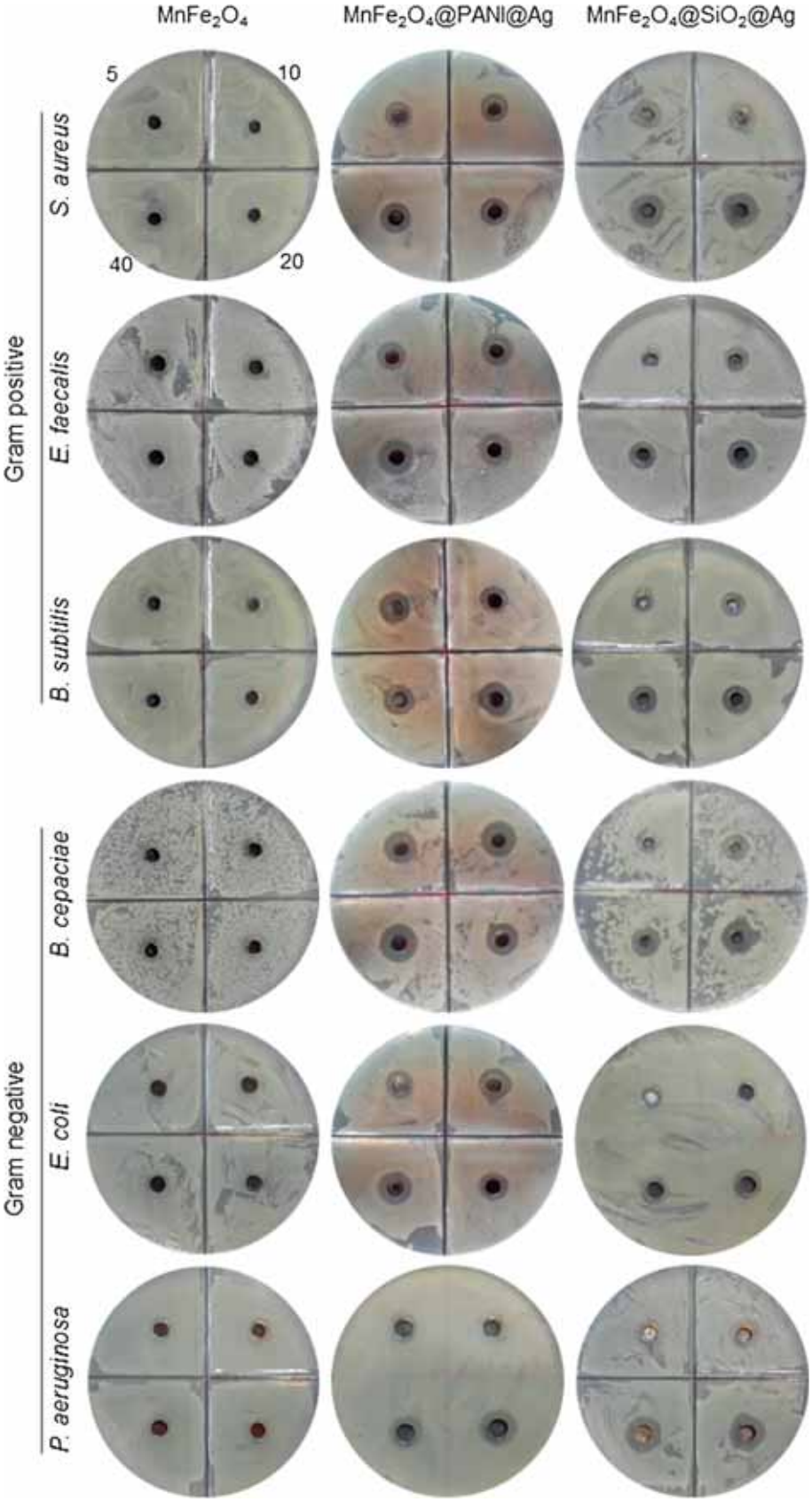

Figure 6. Antimicrobial effects of $MnFe_2O_4$@PANI@Ag and $MnFe_2O_4$@SiO_2@Ag against $MnFe_2O_4$ with different concentrations (5, 10, 20 and 40 mg ml^{-1}) on both Gram-positive and -negative bacteria ($MnFe_2O_4$@PANI and $MnFe_2O_4$@SiO_2 are not shown here).

that when the concentration was increased, the antibacterial activity was also increased. For *P. aeruginosa*, the same situation was notable. Although $MnFe_2O_4$@PANI@Ag had a bacteriostatic effects on both Gram-positive and Gram-negative bacteria, it showed bactericidal effect only on *P. aeruginosa*.

According to the antimicrobial properties of the composites with obtained inhibition zones, the results demonstrated that the magnetic $MnFe_2O_4$, $MnFe_2O_4$@PANI and $MnFe_2O_4$@SiO_2 were not active, while the composite binding with Ag nanoparticle was active in antimicrobial property with inhibition zone diameters. The bacteriostatic persistence evaluation showed that this kind of silver-loaded magnetic composite particles had high stability and persistent bacteriostatic activities.

4. Conclusion

Newly synthesized various Ag-containing Fe_3O_4 and $MnFe_2O_4$ nanocomposites can be considered as antibacterial materials with a broad spectrum antibacterial ability to both Gram-positive and Gram-negative bacteria. The obtained results revealed that these nanocomposites have higher antibacterial performance than that of Fe_3O_4, $MnFe_2O_4$, Fe_3O_4@HA, Fe_3O_4@Nico, Fe_3O_4@His, $MnFe_2O_4$@PANI and $MnFe_2O_4$@SiO_2 nanocomposites and some of the standard antibacterial drugs. Also these magnetic nanocomposites can be reused because of their magnetic property. These results suggest that the unique nanocomposites we have prepared here may serve as an effective, sustainable and recyclable antibacterial agent. This method allows promise for future production technologies and also provides an interesting tool for materials science and a number of novel applications in biological sciences.

Acknowledgement

We would like to thank Fatih University (P50031504_B (10790)).

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