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Facile solvothermal preparation of Fe₃O₄-Ag nanocomposite with excellent catalytic performance

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Functional nanocomposites demonstrate excellent comprehensive properties and outstanding characteristics for numerous applications. Magnetic nanocomposites are an important type of composite materials, due to their applications in optics, medicine and catalysis. In this report, a new Fe₃O₄-loaded silver (Fe₃O₄-Ag) nanocomposite has been successfully synthesized *via* a simple solvothermal method and *in situ* growth of silver nanowires. The silver nanowires were prepared *via* the reduction of silver vanadate with the addition of uniformly dispersed Fe₃O₄ nanoparticles. Structural and morphological characterizations of the obtained Fe₃O₄-Ag nanocomposite were carried out using many characterization methods. As a new composite catalyst, the synthesized magnetic Fe₃O₄-Ag nanocomposite displayed a high utilization rate of catalytically active sites in catalytic reaction medium and showed good separation and recovery using an external magnetic field. The facile preparation and good catalytic performance of this Fe₃O₄-Ag nanocomposite material demonstrate its potential applications in catalytic treatment and composite materials.

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1. Introduction

In recent years, the pollution of water by phenol and aniline compounds has attracted widespread attention worldwide. Among these compounds, 4-nitrophenol (4-NP) belongs to a class of highly toxic compounds, which are difficult to degrade and most difficult to treat. At the same time, 2-nitroaniline (2-NA) has strong water solubility, and can easily penetrate into the soil. The polluted soil then causes pollution of surface water, groundwater and river water. Thus, nitroaniline substances have been listed as key environmental pollutants in many countries. In order to efficiently degrade 2-NA and 4-NP, various nanomaterial catalysts, such as CuO,¹ Ag/KCC-1,² and CuPd,³ have been successfully developed. In particular, among these materials, magnetic nanoparticles have shown great potential in the degradation and catalysis of organic pollutants. Fe₃O₄ has aroused great research interest in the field of magnetic materials and its wide applications are well established⁴ in fields such as magnetic resonance imaging (MRI),⁵ spintronics,^{6,7} lithium ion batteries,⁸ catalysis,⁹ targeted drug delivery^{10,11} and environmental remediation.¹²⁻¹⁴ Metal nanoparticles also have many applications, in fields

such as catalysis, electronics, optics, optoelectronics, sensing and data storage. Because of their unique properties,¹⁵⁻²⁶ metal nanoparticles represent an important class of components. Ag nanoparticles exhibit many original chemical, physical and catalytic properties as good metal conductors.²⁷ In recent years, self-assembled Ag composite materials have attracted the increasing attention of a large number of scientific researchers, including those in fields such as high-performance nanofilms,²⁸ biosensors,²⁹ electroless copper plating³⁰ and catalytic materials.³¹⁻³³ Thus, in the present work, Ag nanoparticles were speculated to form a magnetic nanocomposite with Fe₃O₄ particles, which could be expected not only to show good catalytic performance, but also reduce the loss of Ag nanoparticles and increase reusability.

In this work, Fe₃O₄ nanoparticles with uniform and controllable sizes were prepared using a modified solvothermal method. Then a uniform Fe₃O₄-Ag nanocomposite was successfully prepared and characterized by a series of morphological and spectral characterization techniques. The obtained Fe₃O₄-Ag composite was used as a new catalyst for catalytic reduction reactions of 4-NP and 2-NA, demonstrating good catalytic ability and excellent reusability.

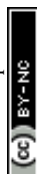
2. Materials and methods

2.1 Materials

Ferric chloride hexahydrate (FeCl₃·6H₂O) and polyethylene glycol (PEG, *M_w* ~6000 g mol⁻¹) were purchased from Aladdin

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situ grown silver nanowires. In the process of oleylamine reducing silver vanadate, Ag nanowire crystals grew *in situ* in the presence of the Fe₃O₄ nanoparticles under strong mechanical agitation. Then, the scheme illustrates the catalytic reaction of the Fe₃O₄-Ag nanocomposite with the substrates 4-NP and 2-NA. More specifically, 4-NP was reacted with fresh NaBH₄ solution to obtain the product 4-aminophenol (abbreviated as 4-AP). Similarly, in the presence of the Fe₃O₄-Ag nanocomposite, the reduction process showed the conversion of the reactant 2-nitroaniline to *o*-phenylenediamine (abbreviated as OPD).⁴⁹

SEM and TEM images in Fig. 2 illustrate the morphology of the initial multifaceted geometry of the Fe₃O₄ nanoparticles and Fe₃O₄-Ag nanocomposite. From Fig. 2a and b, it can be seen that the prepared Fe₃O₄ nanoparticles did not have a smooth spherical surface, but instead exhibited an obvious angular multifaceted geometry. As shown in Fig. 2c and d, organized silver nanowires appeared after reduction by oleylamine.⁵⁰ The obtained silver nanowires seemed uniform with an average diameter of approximately 40–60 nm with Fe₃O₄ nanoparticles anchored on the surface.

In addition, as shown in Fig. 3, the element mapping results show that the prepared Fe₃O₄-Ag nanocomposite was mainly composed of elements including Ag, O, and Fe. These elemental maps further verify that the Fe₃O₄ nanoparticles were surrounded by Ag nanowires and that Ag nanowires were dispersed homogeneously. To gain more information about the microstructure of the Ag nanowire crystals, high-resolution transmission electron microscopic (HRTEM) analysis was necessary. As shown in Fig. 4a, the obtained Ag nanowire crystal showed a clear crystalline structure and distinct lattice fringe. The interplanar spacing was easily resolved with a value of 0.238 nm, corresponding to the Ag (111) crystal plane. In addition, Fig. 4b demonstrates the XRD pattern of the original Fe₃O₄ nanoparticles compared with that of the Fe₃O₄-Ag nanocomposite. It can be seen from the XRD data that the pattern of the Fe₃O₄ nanoparticles



Fig. 3 SEM image (a) and O/Fe/Ag (b, c, and d) elemental mapping of the obtained Fe₃O₄-Ag nanocomposite.

demonstrates characteristic diffraction peaks at 2θ values of 30.1°, 35.4°, 43.0°, 53.4°, 57.1° and 62.2°, which corresponded to the (220), (222), (400), (422), (511) and (440) crystals planes, respectively. These results are in good agreement with the standard card (PDF#88-0315).⁵¹ In previous reports, many similar test results for the diffraction peaks of Fe₃O₄ NPs have been described explicitly.^{52–54} As for the pattern of the Fe₃O₄-Ag nanocomposite, obvious diffraction peaks could be observed and indexed to the (111), (200), (220) and (311) planes of pure face-centered cubic (fcc) silver crystals.⁵⁵

Fig. 5 demonstrates the magnetization hysteresis loops of the as-prepared Fe₃O₄ nanoparticles and Fe₃O₄-Ag nanocomposite at room temperature. The saturation magnetization of the prepared Fe₃O₄ nanoparticles was 83.0 emu g⁻¹, which is more than the values reported for other pure Fe₃O₄ nanocrystals.^{56–59} In addition, the saturation magnetization value of the Fe₃O₄-Ag nanocomposite was 63.2 emu g⁻¹. These results indicate the successful preparation of the Fe₃O₄-Ag nanocomposite with a coating of Ag nanowires. X-ray photoelectron spectroscopy (XPS) was used to study the surface composition and elemental states in the



Fig. 2 TEM and SEM images of the prepared Fe₃O₄ nanoparticles (a and b) and Fe₃O₄-Ag nanocomposite (c and d).



Fig. 4 HRTEM image of Ag nanowires (a) and XRD patterns of the Fe₃O₄-Ag nanocomposite (b).



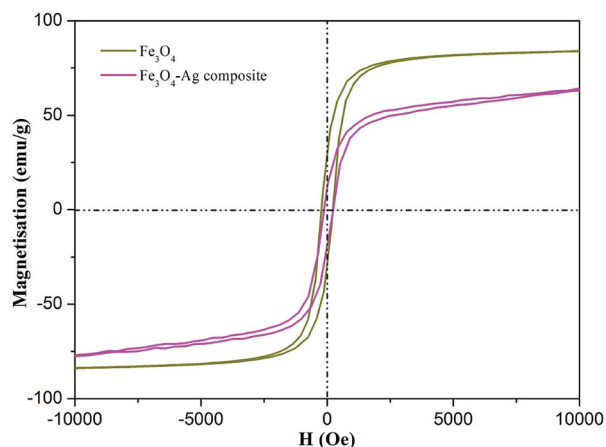


Fig. 5 Magnetization hysteresis loops of as-prepared Fe_3O_4 nanoparticles and Fe_3O_4 -Ag nanocomposite.



Fig. 6 XPS profiles of the Fe_3O_4 -Ag nanocomposite (a) and the deconvolution of Ag peaks (b).

composite.^{60–68} As depicted in Fig. 6a, the wide scan of the XPS spectrum showed characteristic peaks of C1s, O1s, Ag3d and Fe2p from the as-prepared Fe_3O_4 -Ag nanocomposite. From the deconvolution of Ag peaks in Fig. 6b, two peaks appeared in the Ag(3d) binding energy region. One, located at a binding energy of 368.1 eV, is assigned to $\text{Ag}(3d_{5/2})$ and the other located at 374.1 eV corresponds to $\text{Ag}(3d_{3/2})$ with a distance of 6 eV, which may correspond to the $3d_{5/2}$ and $3d_{3/2}$ lines of metallic silver, respectively. Compared with the main Ag^0 (71%) component, the Ag^+ (7%) and Ag^{2+} (22%) content seemed very small, indicating the successful anchoring of the Ag metal onto the obtained composite.

3.2 Catalytic performance

Next, reduction reactions of 4-NP and 2-NA with NaBH_4 solution were used to assess the catalytic activity of the as-obtained Fe_3O_4 -Ag nanocomposite. NaBH_4 provided negative hydrogen ions to attack 4-NP. As shown in Fig. 7, with the process of the classic reduction reaction, the peak of 4-NP at 402 nm decreased gradually, indicating the formation of 4-AP. It can be observed that a period of 70 min was required to finish the reaction. As for the catalytic reduction of 2-NA, after adding the aqueous NaBH_4 solution fully, no significant changes in the 2-NA solution were observed in the characteristic absorption peak at 415 nm, as shown in Fig. 7b. However, after adding the Fe_3O_4 -

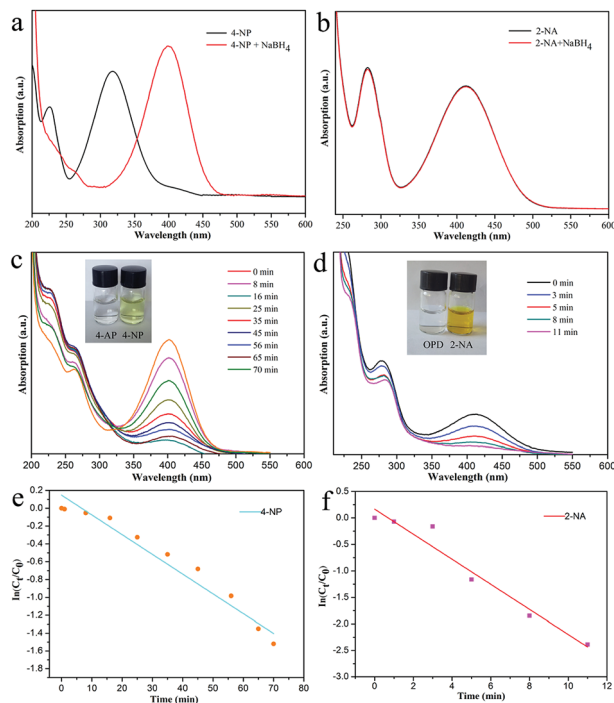


Fig. 7 The spectra of 4-NP and 2-NA with the addition of NaBH_4 (a and b). Curves of the catalytic reduction of 4-NP (c) and 2-NA (d). Plots of $\ln(C_t/C_0)$ versus time for 4-NP (e) and 2-NA (f). Pictures (insets in (c) and (d)) represent the solutions before/after the catalytic reaction.

Ag composite as a catalyst, the absorption peak of 2-NA was significantly reduced, while the system solution changed from bright yellow to pale yellow to colorless simultaneously after only 11 minutes. Fig. 7e and f depict the linear relationship between $\ln(C_t/C_0)$ and time (t), in which C_0 represents the initial concentration, and C_t represents the concentrations of 4-NP at time t . The pseudo-first-order reaction rate constant (k) was fitted and calculated with the values 0.022 and 0.24 min^{-1} for the reduction reactions of 4-NP and 2-NA, respectively.

For heterogeneous materials to be used in practical applications, their level of reusability and activity in cycling experiments are critical factors. Therefore, in this work, the reusability of the as-prepared Fe_3O_4 -Ag catalyst was investigated *via* reduction reactions of 4-NP and 2-NA with fresh NaBH_4 solution. The activity of the catalyst was calculated based on the reduction in the reaction rate relative to the initial rate per use. After the catalytic reductions of 4-NP and 2-NA, the Fe_3O_4 -Ag composite catalyst could be easily recovered magnetically from the mixed solution and washed with deionized water and ethanol for the next cycle test. Fig. 8 shows the reusability test of the Fe_3O_4 -Ag composite over eight successive cycles. The material exhibited a stable catalytic conversion of more than 92%, demonstrating its wide applications in material catalysis and composite design. In addition, as shown in Fig. 9, the morphological and elemental mapping results of the Fe_3O_4 -Ag nanocomposite after eight consecutive cycles demonstrate a stable and high dispersion state, highlighting its great potential and high capacity as a new catalyst.





Fig. 8 Reusability test of the present $\text{Fe}_3\text{O}_4\text{-Ag}$ nanocomposite as a catalyst for the reduction of 4-NP (a) and 2-NA (b) for eight consecutive cycles.

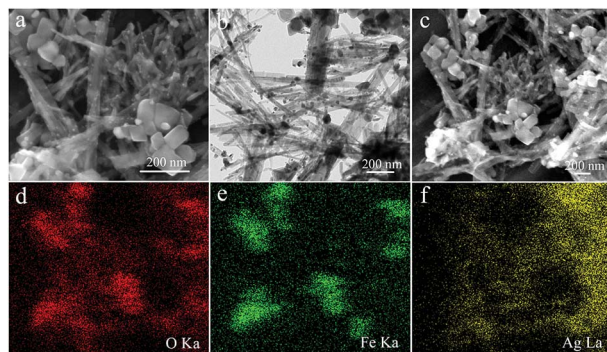


Fig. 9 SEM (a) and TEM (b) images with O/Fe/Ag elemental mapping (d, e, and f) of the $\text{Fe}_3\text{O}_4\text{-Ag}$ nanocomposite after eight consecutive cycles of catalytic reactions.

4. Conclusions

In summary, we have successfully synthesized a $\text{Fe}_3\text{O}_4\text{-Ag}$ nanocomposite material *via* a solvothermal method. A series of characterization techniques revealed that Ag nanowires were successfully anchored onto Fe_3O_4 nanoparticles. The synthesized $\text{Fe}_3\text{O}_4\text{-Ag}$ composite showed excellent catalytic activity for the reduction of 4-NP and 2-NA in the presence of NaBH_4 . It is interesting to note that the as-synthesized $\text{Fe}_3\text{O}_4\text{-Ag}$ composite catalyst demonstrated good reusability even after eight cycles at room temperature. Thus, the as-prepared composite catalyst, with superior catalytic ability, convenient magnetic separation capability and excellent reusability, is expected to have great potential as a magnetic functional composite for various advanced catalytic applications.

Conflicts of interest

There are no conflicts to declare.

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