



A facile strategy for the preparation of a porous flower-like $\text{Fe}_3\text{O}_4/\text{Cu}_2\text{O}/\text{Ag}$ nanocomposite with unexpected and recyclable photocatalytic activity under visible light irradiation

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ABSTRACT

In this work, a novel magnetic and ordered $\text{Fe}_3\text{O}_4/\text{Cu}_2\text{O}/\text{Ag}$ ternary composite was successfully synthesized via one-pot method in the presence of Fe_3O_4 nanoparticles. The glycol-ethanol mixed system was used as both solvent and reductant for the simultaneous formation of Cu_2O and Ag. The porous and hollow flower-like $\text{Fe}_3\text{O}_4/\text{Cu}_2\text{O}/\text{Ag}$ composite was self-assembled by many nanosheets with an average thickness of 10 nm and exhibited unexpected photocatalytic activity for the degradation of methyl orange (MO) under visible light. The degradation rate of MO in aqueous solution ($1 \times 10^{-4} \text{ mol L}^{-1}$) reached to 97.7% within 40 min. The (111) facet of Cu_2O exhibited higher activity for the absorption of MO. Furthermore, ternary composite could be easily recycled by applying an external magnetic field without significant decrease of the catalytic activity even after running 5 times, which presented higher stability and efficiency than $\text{Fe}_3\text{O}_4/\text{Cu}_2\text{O}$. This work would provide a new sight for the construction of visible light-responsive photocatalysts with high performance.

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1. Introduction

Photocatalysts have attracted significant attention in contaminant degradation due to their ability to vanish pollutants under environment-friendly light [1,2], such as sunlight. To date, developing photocatalysts which can absorb visible light is still the research hotspot [3].

Cuprous oxide (Cu_2O) is a P-type oxide semiconductor with a direct band-gap of 2.17 eV, which has been found in application of solar energy conversion, micro/nanoelectronics, magnetic storage devices and biosensors [4]. Moreover, Cu_2O can be easily generally deactivated by photocorrosion, which greatly limits its practical application. In previous reports, many research efforts have been focused on combination with noble metal to enhance the photocatalytic activity of Cu_2O . Zhu et al. [5] prepared the octahedron shaped Cu_2O crystal and $\text{Cu}_2\text{O}-\text{Au}$ heterostructures with various morphologies by using organic solvent hydrazine hydrate, and the resultant nanocomposites exhibit fascinating photocatalytic degradation of methylene blue. Kong et al. [6] used heterogeneous nucleation method synthesized nearly monodisperse octahedral $\text{Au}/\text{Cu}_2\text{O}$ nanocrystals with single crystalline sheets, which

present excellent photocatalytic activity toward MO degradation. Inspired by the above mentioned results, we successfully synthesized a $\text{Fe}_3\text{O}_4/\text{Cu}_2\text{O}/\text{Ag}$ ternary composite. The whole process did not involve any toxic reagents or surfactants, and could be finished via a one-pot synthesis method in the presence of Fe_3O_4 . Furthermore, the photocatalytic performances of the samples were evaluated under visible light ($\lambda > 420 \text{ nm}$) irradiation.

2. Experimental details

Fe_3O_4 nanoparticles were synthesized by a green, modified coprecipitation method according to our previous report [7]. The preparation process of $\text{Fe}_3\text{O}_4/\text{Cu}_2\text{O}/\text{Ag}$ composites are given as follows: 0.2416 g of $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ and 0.0566 g of AgNO_3 were dissolved in a 25 mL of ethylene glycol and ethanol mixed solution with volume ratio of 3:2, and stirred for 20 min. Then 25 mg of Fe_3O_4 nanoparticles were added and sonicated for 30 min. The mixed solution was sealed in a Teflon lined stainless-steel autoclave and was heated at 160 °C for 3 h, 5 h and 7 h (the obtained final products were named S-1, S-2 and S-3 in turn.) in the electric oven, respectively, cooled down to room temperature. The products were centrifuged and washed with deionized water and ethanol three times and finally dried at 60 °C for 24 h in vacuum oven. The synthetic method for $\text{Fe}_3\text{O}_4/\text{Cu}_2\text{O}$ composite is similar to the $\text{Fe}_3\text{O}_4/\text{Cu}_2\text{O}/\text{Ag}$ composite, but without the addition of AgNO_3 .

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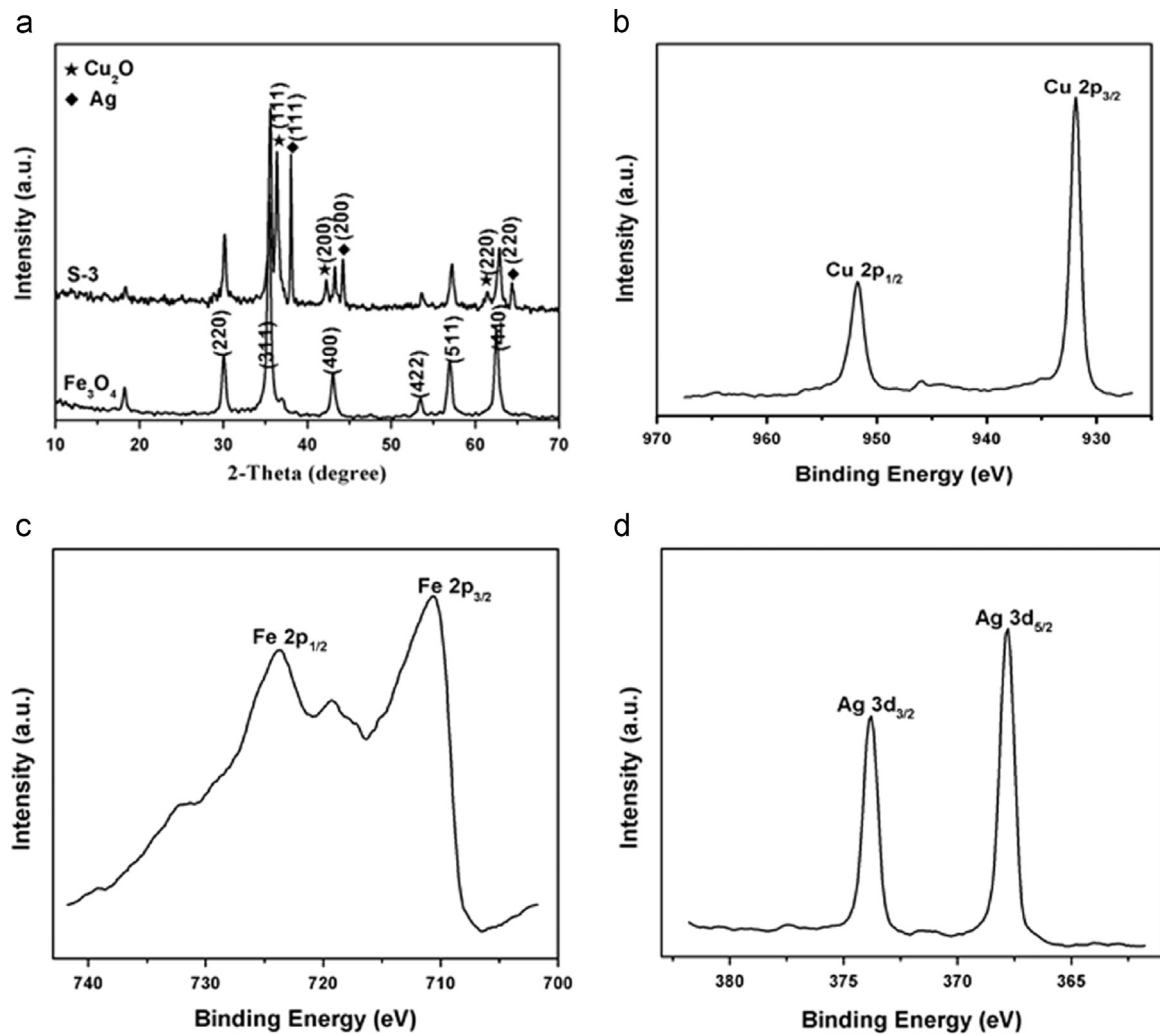


Fig. 1. (a) XRD patterns of Fe₃O₄ nanoparticles and S-3. XPS spectrum of S-3: (b) Cu 2p, (c) Fe 2p and (d) Ag 3d.

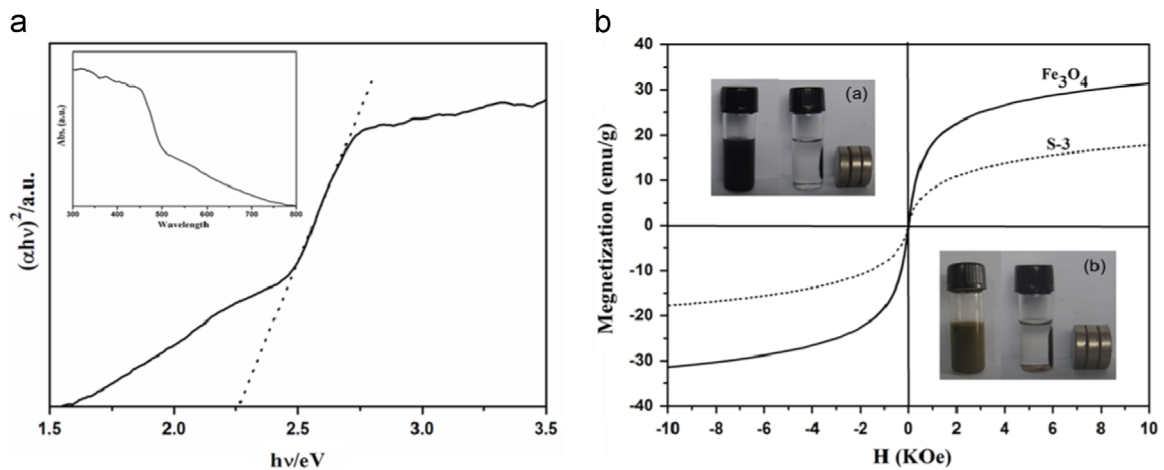


Fig. 2. (a) UV-vis DRS spectrum (inset) and $(\alpha h\nu)^2$ versus $h\nu$ plot of S-3. (b) Room temperature magnetization of Fe₃O₄ nanoparticles and S-3.

3. Results and discussion

Fig. 1a shows the XRD patterns of the as-prepared Fe₃O₄ nanoparticles and S-3. Six major reflections located at about 30.1°, 35.4°, 43.1°, 53.5°, 57.1° and 62.6° can be assigned to diffraction of Fe₃O₄ with cubic-phase from the (220), (311), (400), (422), (511) and (440) planes (JCPDS card no. 01-074-1910), respectively. No

other peaks are observed, indicating the pure crystalline phase of Fe₃O₄. The new peaks of S-3 at 2θ values of 36.4°, 42.4° and 61.4° noted as “★” can be perfectly indexed to the crystal planes of (111), (200) and (220) of the cubic phase Cu₂O (JCPDS card no. 01-078-2076), respectively. There also present three diffraction peaks at 38.1°, 44.3° and 64.2°, noted as “◆”, which are assigned to the (111), (200) and (220) planes of a cubic phase Ag nanocrystals

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