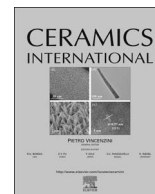




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Scalable and facile preparation of optical-magnetic dual function 3D Ni@graphene-ZnO for high efficiency removal of hexavalent chromium

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ABSTRACT

For the first time, ZnO nanoparticles decorated three-dimensional (3D) Ni@graphene (Ni@GE) composites with uniform size and magnetic separation properties were synthesized by a simple and scalable one-step sol-gel method. The photocatalysts were characterized by X-ray diffraction, fourier-transform infrared spectra, field-emission scanning electron microscope, transmission electron microscopy, UV-vis diffusive reflectance spectra and photo luminescence. The results indicated the ZnO nanoparticles loaded on the outsides of the Ni@GE were no significant numbers of vacancies or apparent aggregation. Meanwhile, the introduction of 3D structured graphene material resulted in the better charge separation efficiency compared with that of the pure ZnO. More significantly, the as-prepared Ni@graphene-ZnO showed superior photocatalytic activity and stability for Cr(VI) reduction to Cr(III).

1. Introduction

Hexavalent chromium (Cr(VI)) and trivalent chromium (Cr(III)) are the most common forms of chromium [1]. Cr(VI) is highly toxic to plants and animals due to its ability to oxidize biomolecules and notably DNA. However, Cr(III) is relatively nontoxic and an essential nutrient in the human diet to maintain the effective glucose, lipid and protein metabolism [2,3]. Therefore, the conversion of Cr(VI) to Cr(III) is an important subject in the field of inorganic pollutants elimination [4]. As a green and sustainable technology, semiconductor-based photocatalysis has received much attention in the last few decades due to a wide variety of applications (e.g., the photocatalytic reduction of Cr(VI)) [5]. ZnO is an n-type semiconductor with a band gap energy of 3.37 eV and has been used as one of the promising photocatalyst due to its low cost, high solar absorbency power and nontoxic nature [6]. Several ZnO-based materials have been developed for photocatalytic reduction of Cr(VI) [7–10], but the preparation methods and recollection of the photocatalysts were relatively complex and difficult, limiting their large scale applications.

In recent years, graphene has attracted considerable interest owing to its high conductivity, fast mass and electron transport kinetics, and large specific surface area [4,11,12]. Our previous studies have already demonstrated that the combination of graphene with semiconductor-based materials such as CoFe₂O₄ [13] and Fe₃O₄@CuO [14] not only

solved the nanoparticles agglomeration problem and showed high catalytic activity, but also implemented easily the composite separation via an external magnetic field. Herein, based on the excellent properties of ZnO and graphene, we designed and constructed a hybrid of magnetic 3D Ni@GE composites and ZnO nanoparticles. To the best of our knowledge, there is no report on the controlled synthesis of Ni@GE-ZnO photocatalysts (designated as NGZ). Firstly, the graphene membranes in Ni@GE composite prepared by our previous reported method [15] acted as carriers to support ZnO nanoparticles. Hereafter, the growth of the outer ZnO nanoparticles were accomplished by a simple one-step sol-gel method, which does not require expensive equipment or facilities for patterning or for chemical processing, illustrating the advantages of being simple, fast, low cost, controllable and highly scalable [16]. Besides, the introduction of 3D structured graphene materials not only prevented the ZnO nanoparticles agglomeration, but also afforded ZnO nanoparticles better charge separation efficiency, and then enhanced the photocatalytic reduction activity and stability, which were better than that of ZnO and ZnO-graphene reported by other groups [7].

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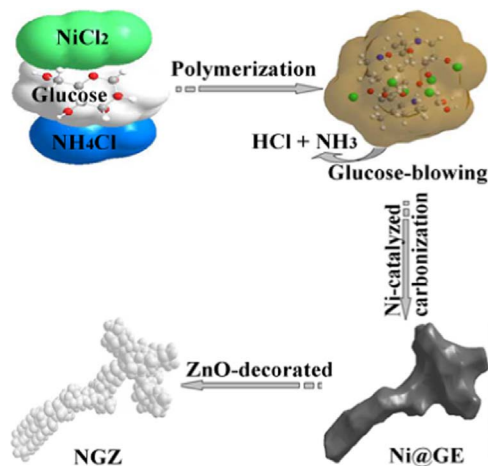


Fig. 1. Schematic illustration of NGZ fabrication.

2. Experimental

2.1. Preparation of NGZ

The synthetic route of NGZ was illustrated in Fig. 1. A series of NGZ composites with different ZnO content were synthesized via a simple and scalable sol-gel method, and denoted as NGZ-X (X=70, 80 and 90 wt%, respectively). Typical procedure of NGZ-80 was as follows: 0.01 g of Ni@GE composites [15] were dispersed into 60 mL of ethanol solution, containing 0.15 g of $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, followed by pH adjustment to 10 by 6 M NaOH. The mixture was stirred at 40 °C for 6 h. Finally, the as-obtained precipitate was washed with deionized water and ethanol, then dried at 60 °C. Additionally, the synthesis methods of ZnO and Ni@GE composites have been provided in Supplementary material S1 and S2.

2.2. Characterization

The structure of the samples was analyzed by powder X-ray diffraction (XRD, Bruker D8, Cu $K\alpha$, $\lambda=0.15418$ nm), Fourier-transform infrared (FT-IR, Nicolet 370FT-IR), field emission scanning electron microscopy (FESEM, SUPRA55) and transmission electron microscopy (TEM, JEOL JEM-2100). The content of Ni and Zn in NGZ composites were indirectly determined by inductively coupled plasma-atomic emission spectrometer (ICP, novAA300). The UV-vis diffuse reflectance spectra of the samples were recorded by a Shimadzu UV-2700 UV-vis spectrophotometer. BaSO_4 was used as a reflectance standard. Photo Luminescence (PL) measurements were performed

using a Jobin Yvon SPEX Fluorolog-3-P spectroscopy. The removal of Cr(VI) ions were performed by measuring the absorbance values at various intervals of time using a UV-visible spectrometer (UV-Vis, DDR).

2.3. Photocatalytic activity measurement

The photocatalytic performance of the samples was evaluated by reducing Cr(VI) at room temperature (25 °C). 0.01 g of photocatalysts was suspended in 50 mL of $\text{K}_2\text{Cr}_2\text{O}_7$ solutions (10 mg L^{-1}) in a quartz tube, and stirred for 60 min under conditions of dark to reach adsorption-desorption equilibrium. Then the mixed suspensions were exposed to UV irradiation produced by a 300 W high pressure Hg lamp. At given time intervals of irradiation, 3 mL of aliquots was withdrawn, and then magnetically separated to remove essentially all the catalyst. The characteristic adsorption peak at 350 nm for Cr(VI) was used to evaluate the variation of Cr(VI) concentration by a Shimadzu UV-2700 UV-vis spectrophotometer.

3. Results and discussion

The XRD patterns of Ni@GE and NGZ composites are shown in Fig. 2A. The XRD pattern of Ni@GE depicted diffraction peaks at $2\theta=44.51$, 51.85 , and 76.37° , which corresponded to the (111), (200) and (220) planes of cubic Ni (JCPDS: 04-0850), indicating that Ni^{2+} ions were converted into metallic Ni after calcination process [4,15]. Compared with the XRD patterns of Ni@GE, additional diffraction peaks appeared in the XRD patterns of the obtained NGZ at 31.77 , 34.42 , 36.25 , 47.54 , 56.60 , 62.86 , 66.38 , 67.96 , 69.10 and 72.56° , which could be indexed to the (100), (002), (101), (102), (110), (103), (200), (112), (201) and (004) planes of cubic ZnO (JCPDS: 36-1451), confirming the formation of ZnO and revealing the successful synthesis of NGZ.

FTIR measurements provided further evidence for the formation of NGZ catalyst. As shown in Fig. 2(B), the broad and intense band observed at 3340 cm^{-1} was ascribed to the stretching vibration of O-H [17]; and the band at about 2924 and 2850 cm^{-1} corresponded to the asymmetric and symmetric C-H stretching vibrations [18], but 1470 and 645 cm^{-1} for the scissoring and out-of-plane bending vibration of the C-H [19,20]. Moreover, the presence of the band of relatively medium intensity at 1621 and 1068 cm^{-1} indicated the presence of C=O stretching vibration [21] and C-O-C deformation vibration [22], respectively. However, after the sol-gel treatment, the peaks at 1621 cm^{-1} in the FTIR spectrum of Ni@GE shifted to 1647 cm^{-1} in the spectrum of NGZ, while a new prominent absorption band appeared at about 479 cm^{-1} [23,24], which corresponded to the stretching mode of Zn-O, implying the combination of ZnO nanocryst-

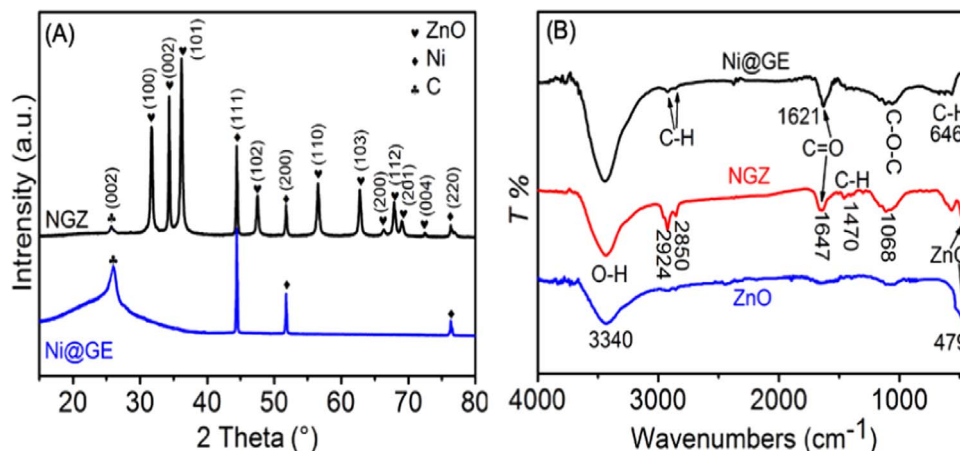


Fig. 2. (A) XRD patterns of Ni@GE and NGZ; (B) FTIR spectra of Ni@GE, NGZ and ZnO.

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