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Photo-assisted degradation of bisphenol A by a novel FeS_2@SiO_2 microspheres activated persulphate process: Synergistic effect, pathway and mechanism

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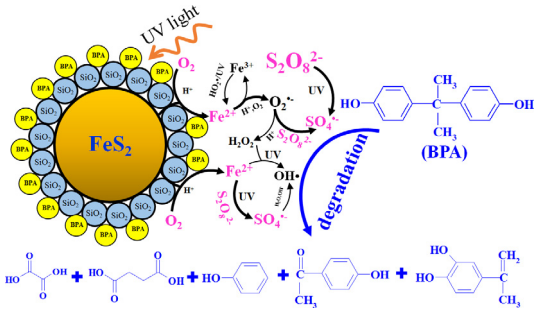
HIGHLIGHTS

- FeS_2@SiO_2 microspheres were firstly employed to activate persulfate (PS).
- UV induced a significant improvement in the degradation of bisphenol A (BPA).
- A synergistic effect between photochemical and catalytic processes was achieved.
- $SO_4^{\cdot -}$ radicals prevailed over HO· in the FeS_2@SiO_2 microspheres/PS/UV system.
- A possible reaction pathway and mechanism of BPA degradation was proposed.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, FeS_2@SiO_2 microspheres were firstly employed as a heterogeneous catalyst to activate persulfate (PS) for the degradation of bisphenol A (BPA) from aqueous solutions. The most relevant findings revealed that UV irradiation induced a significant improvement in the degradation of BPA by the FeS_2@SiO_2 microspheres/PS system. Nearly 100% of BPA degradation by the FeS_2@SiO_2 microspheres/PS/UV system was achieved within 120 min at reaction conditions of 1 mM PS, 0.066 mM BPA, 1.0 g/L FeS_2@SiO_2 microspheres and pH 3.0. A high performance on the degradation of BPA might be attributable to a synergistic effect between the PS/UV and the FeS_2@SiO_2 microspheres/PS catalytic processes. It was found that the BPA degradation could be inhibited by the coexisting anions like Cl^-, HCO_3^- and PO_4^{3-} to different extents at much higher concentrations, whereas NO_3^- had a negligible effect. Organic acids such as ethylene diamine tetra-acetic acid (EDTA) and oxalic acid (OA) would lead to an enhancement with a lower dosage, whereas a significantly negative effect was observed at much higher dosages. Radical scavenging tests revealed that the $SO_4^{\cdot -}$ radicals prevailed over HO·. A total of seven intermediates during the degradation of BPA were identified by GC/MS, and a possible reaction pathway and mechanism of BPA degradation by the FeS_2@SiO_2 microspheres/PS/UV system was proposed. This study

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demonstrated a simple water treatment method involving the use of low cost natural iron minerals for organic pollutants removal.

1. Introduction

Recently, advanced oxidation process based on the sulfate radicals ($SO_4^{\cdot -}$) has received growing attention as a promising alternative for the degradation and mineralization of organic pollutants in water and sediment owing to its advantages of highly stable reactivity, widely operative range and relatively low cost [1–8]. $SO_4^{\cdot -}$ radicals can be generated via activation of persulfate (PS) or peroxymonosulfate (PMS) using a series of physic-chemical processes such as UV, heat, electron, microwave and transition metals ions (Me^{n+}) [2,4,9–13]. Among these initiators, it has been reported that Fe^{2+} is the most preferable initiator to activate PS for the generation of $SO_4^{\cdot -}$ in environmental application [1,14,15]. However, the reaction between Fe^{2+} with PS is too fast to control and excess Fe^{2+} will react with as-formed $SO_4^{\cdot -}$ in a short time, leading to a sharply decreased in removal efficiency of organic pollutants [16,17]. To overcome this drawback, $SO_4^{\cdot -}$ based heterogeneous Fenton-like system has now considered as an alternative to make reaction between Fe^{2+} with PS smoothly since the heterogeneous catalyst is used as a slow-releasing source of Fe^{2+} [3,18,19]. Iron-containing solid materials such as iron minerals and iron oxide have been employed as a heterogeneous catalyst for activation of PS in this system [3,16,18,19,20]. Among these materials, FeS_2 has been proved to be the most effectively heterogeneous catalyst for the degradation of organic pollutants by comparison with other iron oxides [3,18,21,22]. However, Fe^{2+} could be conveniently dissolved from FeS_2 under an acidic condition, not to mention a strong oxidizing condition including oxidizing agents such as HO·, $SO_4^{\cdot -}$ and H_2O_2 [3,18,21–23]. It might still happened that excess Fe^{2+} dissolved from FeS_2 will react with as-formed $SO_4^{\cdot -}$ within a short time, leading to a poor utilization of $SO_4^{\cdot -}$. Most recently, our previous work reported that FeS_2@SiO_2 microspheres were used as catalysts to activate H_2O_2 for the degradation of ciprofloxacin in water [22]. Previous results indicated that SiO_2 microspheres on the surface of FeS_2 could make the reaction between Fe^{2+} and H_2O_2 smoothly continuous by controlling the release of Fe^{2+} from FeS_2. Additionally, ciprofloxacin could be more conveniently adsorbed on the SiO_2 microspheres surface, leading to an enhanced in removal efficiency. Since $SO_4^{\cdot -}$ radicals are more stable reactivity and widely operative range than HO·, thereby it was attempted to use FeS_2@SiO_2 microspheres as catalysts to activate PS for the degradation of organic pollutants under UV irradiation. But it is still unclear whether a synergistic effect between the PS/UV and the FeS_2@SiO_2 microspheres/PS catalytic processes could be achieved.

Nowadays, bisphenol A (BPA), identified as an endocrine disrupting chemical, has been deteriorated the generative function of living things [24–26]. Hence, a rapid and simple treatment of BPA is now required as soon as possible. To the best of our knowledge, activation of PS by both the FeS_2@SiO_2 microspheres and UV for the degradation of BPA in water has not yet been reported. The main objectives of this study were: (1) evaluate a synergistic effect between the PS/UV and the FeS_2@SiO_2 microspheres/PS catalytic processes; (2) determine the degradation of BPA at various conditions; (3) elucidate PS decomposition under different systems; (4) identify reactive oxygen species and intermediate products of BPA degradation; and (5) explore a possible reaction pathway and mechanism of BPA degradation by the FeS_2@SiO_2 microspheres/PS/UV system.

2. Materials and methods

2.1. Materials

The detail synthesis procedure and characterization of FeS_2@SiO_2 microspheres samples were described by our previous study [22]. SEM and TEM analysis in pervious study indicated that the FeS_2 cores of particles were covered with SiO_2 microspheres, and they possess a core–shell structure inevitably (Fig. 1) [22]. Additionally, the SiO_2 microspheres presented uniform and consistent spherical shapes, and the BET specific surface area of the FeS_2@SiO_2 microspheres was 119.15 m^2/g. Bisphenol A (BPA, purity 99%), 5,5-dimethyl-1-pyrroline N-oxide (DMPO, > 99%) and 2,2,6,6-tetramethyl-4-piperidone (TEMP, > 99%) were purchased from Sigma-Aldrich (USA) and stored at 4 °C. Potassium persulfate (PS), ethylenediaminetetraacetic acid (EDTA), oxalic acid (OA) and other chemicals were obtained from Guangzhou Chemical Reagent Co., Ltd., China. All chemicals used in the experiments were of analytical reagent grade without further purification. The double distilled water was used in the whole experiment. Stock standard solution of BPA (1000 mg/L) was prepared in ethanol and then diluted with the double distilled water.

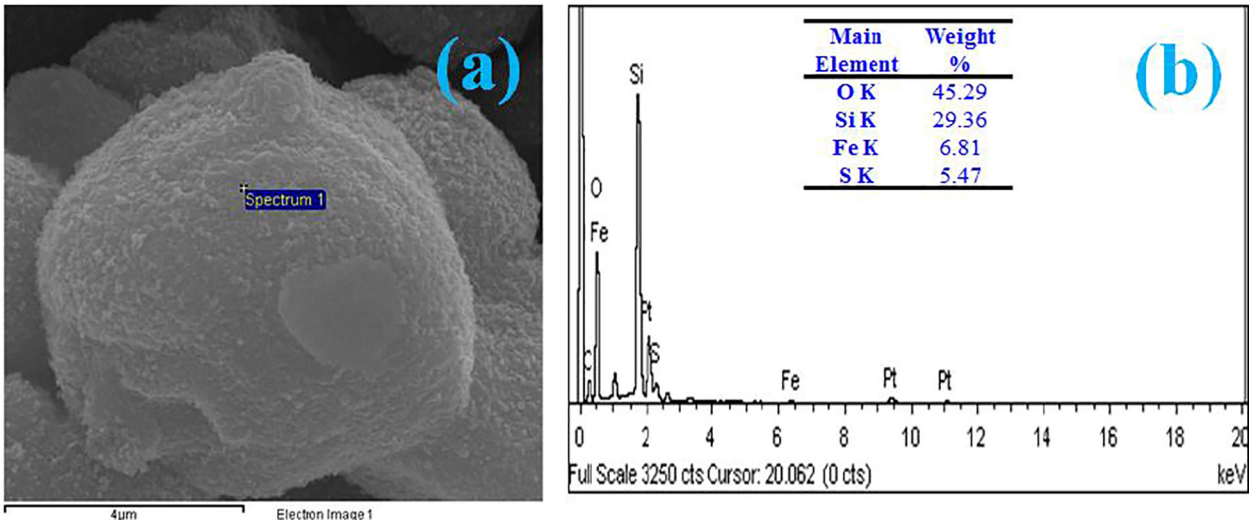

Fig. 1. SEM image and EDS data of FeS_2@SiO_2 microspheres.

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