

Research paper

Removal of acid orange 7 by surfactant-modified iron nanoparticle supported on palygorskite: Reactivity and mechanism

Guixiang Quan^{a,b}, Lijuan Kong^a, Yeqing Lan^b, Jinlong Yan^{a,*}, Bin Gao^c^a School of Environmental Science and Engineering, Yancheng Institute of Technology, Yancheng 224051, China^b College of Sciences, Nanjing Agricultural University, Nanjing 210095, China^c Department of Agricultural and Biological Engineering, University of Florida, Gainesville, FL 32611, United States

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ABSTRACT

To improve the hydrophobic property of iron nanoparticle (INP) coated on palygorskite (P), cationic surfactant cetyltrimethyl ammonium bromide (CTMAB) was used to modify its surface properties through ion exchange. The structure of the composited P-INP/CTMAB was characterized by various analytical methods. The results showed that the long chain of CTMAB was coated on the surface of P-INP without intercalation effect. The morphology of P-INP was thus not obviously changed after the modification, but the special specific surface area increased from $30.63 \text{ m}^2 \text{ g}^{-1}$ to $127.04 \text{ m}^2 \text{ g}^{-1}$. The decoloration and degradation efficiency of P-INP/CTMAB towards acid orange 7 (AO7) was examined under different pH value, temperature and dosage conditions. The results showed that the reaction was preferred to the acidic environment and its first-order reaction rate constant (k_{obs}) linearly depended on the dosage. With suitable conditions, the removal rate of AO7 and total organic carbon (TOC) by P-INP/CTMAB reached about 98.4% and 59.21% after 2 h, respectively. These values were both higher than that of the unmodified composition P-INP, suggesting the synergistic effects in surface adsorption and the following removal reaction. The degradation intermediate products were investigated using HPLC-MS, and the potential degradation pathway of AO7 by P-INP/CTMAB was determined.

1. Introduction

Azo dyes ($-\text{N}=\text{N}-$ unit as the chromophore) are widely utilized in leather, paper-making, printing and textile dyeing because of their chemical stability and versatility (Brown, 1987). However, they are hazardous to aquatic life, disturbing the natural equilibrium through reducing aquatic diversity and photosynthetic activity by blocking the passage of light through water (Gupta et al., 2011). It is thus necessary to search for novel and efficient approaches to treat industrial wastewater containing azo dyes. Up to now, the removal of azo dyes in wastewater mainly relies on physical, chemical, biological methods or their combinations (Forgacs et al., 2004), such as electrochemical treatment (Martinez-Huitle and Brillas, 2009), ozonation (Szyrkowicz et al., 2001), photocatalysis (Han et al., 2009), Fenton or Fenton like reagents (Malik and Saha, 2003), etc. Nevertheless, many of these methods are often ineffective to remove and degrade azo dyes, resulting in a large quantity of solid waste or other secondary contaminant problems (Azam and Hamid, 2006).

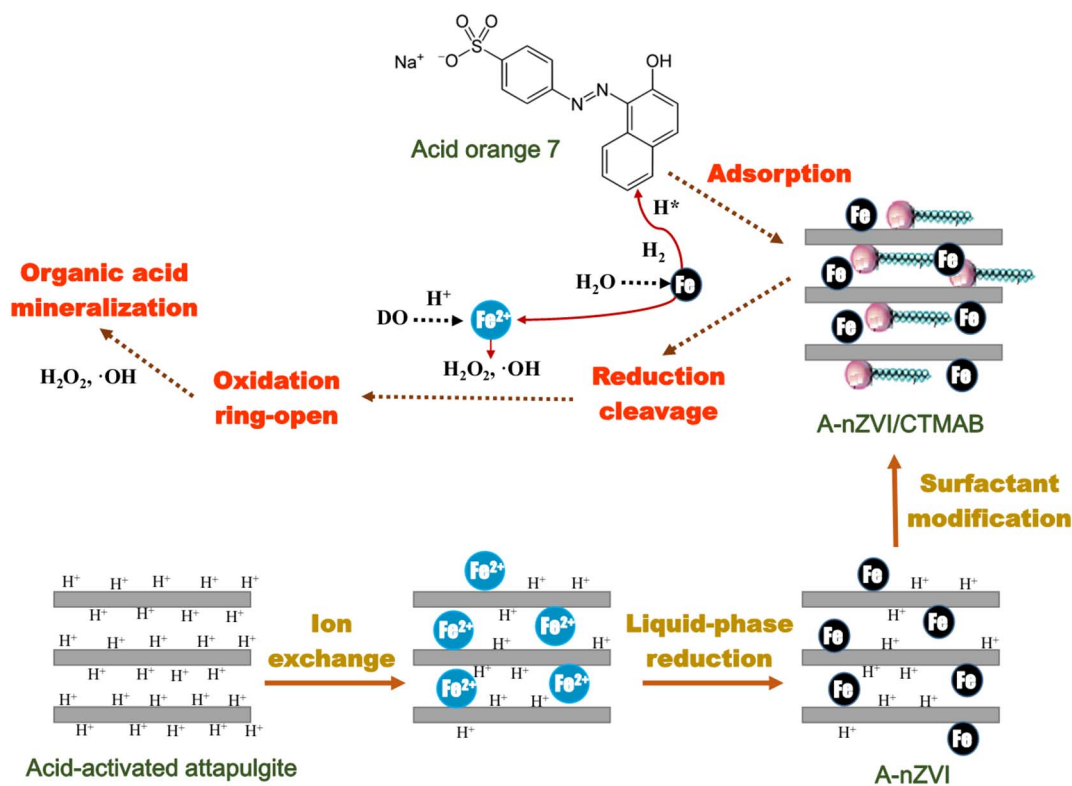
Iron nanoparticle (INP) has been widely used in the degradation of organic contaminants in environment due to its high reductive activity

(Wang and Zhang, 1997). For example, the color and total organic carbon (TOC) removal efficiencies of azo dye by laboratory-synthesized INP can reach 98.9% and 53.8%, respectively, with an initial dye concentration of 100 mg L^{-1} and an INP dosage of 0.3348 g L^{-1} (Shu et al., 2007). However, INP exhibit strong tendency to aggregate via Van der Waals and magnetic attraction forces, forming larger particles with diameters ranging from several microns to several millimeters (Phenrat et al., 2007; Zhan et al., 2008). In addition, the poor transport properties of INP in aquifers limit its practical applications in wastewater treatment or groundwater remediation (Krajangpan et al., 2008). To overcome these drawbacks, several technologies have been developed to enhance the stability of INP, which is thus often modified with various methods including coated with surface stabilizers (He and Zhao, 2005), supported by solids (Qiu et al., 2011), and entrapped in polymers (Liu et al., 2010). As for the surface modification with stabilizers and polymers, the steric or electrostatic repulsion among the INP particles can stabilize them in aqueous solutions. INP particles can also be stabilized through impregnation onto a solid carrier.

A recent study has shown that INP can be grafted onto the surface of acid-activated palygorskite (P-INP) and be used to remove Cr (VI) from

* Corresponding author.

E-mail address: yjlyt@ycit.cn (J. Yan).



Scheme 1. Schematic illustration of preparing P-INP/CTMAB and its degradation for AO7.

solution (Quan et al., 2014a). The results show that 98.73% of 20 mg L⁻¹ Cr (VI) was removed within 60 min by 4 g L⁻¹, which was more effective than iron powder (98%, < 400 mesh) under the same conditions. However, the adsorption capacity of lipophilic pollutants onto P-INP is not so satisfactory due to the hydrophilic characteristic of crystalline hydrate magnesium silicate. Further modification of P-INP by surfactant is thus necessary to increase its hydrophobicity.

In this work, cationic surfactant cetyltrimethyl ammonium bromide (CTMAB) modified P-INP composites (P-INP/CTMAB) were developed for effective removal of azo dye acid orange 7 (AO7) in aqueous solution (Scheme 1). The combined decoloration and mineralization properties and involved removal mechanism of as-synthesized composites were studied in detail. In addition, the intermediate products were analyzed to determine the potential degradation pathway.

2. Materials and methods

2.1. Chemicals

AO7 (≥ 97.0%), potassium borohydride (KBH₄, ≥ 99.9%), silver nitrate (AgNO₃, ≥ 99.5%), CTMAB (≥ 99.0%) and ferrous chloride tetrahydrate (FeCl₂·4H₂O, 99%) of analytical grade were purchased from Sigma-Aldrich. Absolute ethanol (C₂H₅OH), hydrochloric acid (HCl, 37.5%), o-phosphoric acid (≥ 85%) and sodium hydroxide (≥ 99.5%) were obtained from Sino-pharm Chemical Reagent Company (Shanghai, China) and used without further purification. Palygorskite clay was obtained from Jiangsu Junda Material Co., Ltd., and the cation exchange capacity is 15.47 mg/100 g. All the experimental solutions were made with double-distilled water, which was deoxygenated by purging with N₂ for 60 min.

2.2. Synthesis of P-INP/CTMAB

To synthesis the P-INP/CTMAB, the P-INP was firstly prepared according to our previous work (Quan et al., 2014a). In typical procedure,

16 g of FeCl₂·4H₂O was dissolved in the mixture solution of absolute ethanol (72 mL) and deionized water (18 mL), 4.5 g of acid treated palygorskite was then suspended in the solution and oscillated about 24 h in the digital water bath oscillator for fully exchange/sorption of iron (II) ions on the palygorskite clay. The mixture was then centrifuged several times to remove excess iron (II) and then the final product was resuspended into the mixture solution (72 mL ethanol + 18 mL deionized water). After that, 250 mL 1.5 mol/L of freshly prepared KBH₄ solution was added drop-wise into the above solution and kept stirring for 2 h after the addition. The whole process was performed under a N₂ atmosphere with vigorous stirring to avoid the oxidization of P-INP.

The synthesis of P-INP/CTMAB is according to Jia and Wang (2012). The P-INP was firstly dispersion in the aqueous solution containing CTMAB through continuously shaking under room temperature, and ratio of P-INP and CTMAB keeps 1:2. After shaking at 60 °C for 2 h, the suspension was centrifuged at 4500 rpm for 20 min, and the supernatant was removed. Absolute ethanol was used to wash samples instead of water to avoid the immediate oxidation of P-INP/CTMAB, and the obtained composites were stored in a N₂-purged desiccator.

2.3. Characterization of P-INP/CTMAB

The crystalline structure and phase purity of the prepared nanoparticles were examined by a vertical X-ray diffractometer (X'Pert-Pro MPD, Philips the Netherlands) using Cu K_α radiation (λ = 0.15406 nm) generated at 40 kV and 40 mA in the 2θ range from 5° to 90°. Thermo Nicolet Nexus 870 (Nicolet, USA) with the equipment configuration of diamond ATR accessory was used to identify the presence of functional groups in the synthesized composites, and the samples was speed scan 64 times (average was reported) with scan range of 4000–550 cm⁻¹ at a resolution of 0.6329 cm⁻¹ s. Specific surface area of the samples was determined with an ASAP 2020 (Micromeritics, USA) at 77 K. The nitrogen sorption isotherms were measured with the Brunauer-Emmett-Teller (BET) method using the Barrett-Joyner-Halenda (BJH) model (Cai et al., 2014). The

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