



Simultaneous formation of nanoscale zero-valent iron and degradation of nitrobenzene in wastewater in an impinging stream-rotating packed bed reactor



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HIGHLIGHTS

- An impinging stream-rotating packed bed system is investigated.
- Simultaneous NZVI synthesis and nitrobenzene degradation are achieved.
- Higher liquid flow rate and initial reactant concentrations improve degradation.
- Fe²⁺ is a preferable iron source due to a relatively larger pH application range.
- The dosage of iron can be greatly reduced by high gravity technology.

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ABSTRACT

A novel process based on an impinging stream-rotating packed bed (IS-RPB) was proposed for preparing nanoscale zero-valent iron (NZVI) and degrading nitrobenzene (NB) simultaneously. The obtained NZVI particles were characterized by scanning electron microscopy (SEM), high-resolution transmission electron microscopy experiments (HRTEM), X-ray diffraction (XRD) and X-ray photoelectron spectroscopy (XPS). The experimental results showed that the NB removal efficiency increased with the augment of liquid flow rate and initial reactant concentrations. The NZVI prepared with Fe²⁺ as an iron source had high reactive activity and its removal efficiency of NB could be maintained at a high level in acidic or a weakly basic condition (pH < 8). Compared with the traditionally mixing method, the IS-RPB could reduce the iron dosing amounts by 49.6% and the prepared NZVI could achieve 100% efficiency of the NB removal. Considering the operational cost and treatment efficiency, the optimal ratio between NaBH₄ and iron concentration could be in the range of 2.0–3.0. Through the strong micro-mixing intensification performance of IS-RPB, the reported approach has compensated for the shortcomings of the traditional nanoparticles production such as complicated procedures and the potential oxidation of Fe⁰. This study encourages further optimization and development of the high gravity system towards industrial production of NZVI and large-scale NB-containing wastewater treatment.

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1. Introduction

Nitrobenzene (NB) is an important chemical intermediate and widely applied in many fields such as petrochemicals [1], pharmaceuticals [2], explosive materials and dyes [3]. NB has high toxicity with teratogenic, carcinogenic and mutagenic effects, and has been listed as a priority control pollutant in many countries [4]. Thus, removal of NB from wastes/wastewater is critically important.

However, NB is relatively resistant to biodegradation, because the existence of strong electron-withdrawing nitro-group in NB greatly reduces the electronic cloud density to strongly stabilize the chemical property [5]. Currently, the treatment technologies for NB can be classified into single-element and multiple-element methods. Single-element methods include physical, biological and chemical treatment techniques, such as Fenton oxidation [6], ozone oxidation [7–9], electrochemistry [10] and photocatalytic degradation [3,11]. Some of those methods often have the problems of low treatment efficiency, long treatment time and high treatment cost. To address those problems,

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multiple-element methods are developed by combining multiple single-element processes together to compensate for the drawbacks of each process to improve the NB treatment efficiency [12]. Among those, nanoscale zero-valent iron (NZVI) can be used with multiple-element methods for NB removal. NZVI can facilitate the transformation of NB into degradable aniline (AN), which is then mineralized [13].

Various physical and chemical methods have been developed to prepare NZVI particles, including ball milling [14], physical vapor deposition [15], thermal cracking of iron pentacarbonyl [16], and chemical reduction method [17]. Chemical reduction in the liquid phase becomes a popular method for synthesizing NZVI, because of low cost, high production rate and good dispersion performance of produced particles [18]. In chemical reduction, Fe^{2+} or Fe^{3+} is reduced in the aqueous solution to prepare nanoscale Fe^0 , followed by a series of post-treatment steps including washing, separation and drying. However, these approaches still have several technical problems, such as particle aggregation due to uneven mixing, broad particle size distribution of NZVI, and great effect of process amplification.

As a novel reactor of process intensification, rotating packed bed (RPB) is able to generate a high gravity environment that could strongly intensify the micro-mixing and mass transfer [19–23]. RPB can achieve uniform super saturation and homogeneous nucleation, yielding particles with a narrow size distribution. In addition, the residence time distribution (RTD) of liquid under a strong centrifugation force in a high gravity field can be greatly decreased by the order of magnitude to 10–100 μs [24], much shorter than the typical nucleation time at an order of 1 ms [25]. Thus, the saturation degree can reach the maximum extent of micro-mixing, resulting in nanoparticles within a narrow particle size distribution [26]. Recently, RPB has been widely applied for the preparation of nanoparticle, such as CaCO_3 [27], Fe_3O_4 [28], ZnS [24], and BaTiO_3 [29]. An impinging stream - RPB (IS-RPB) [30,31] was employed to synthesize NZVI within uniform particle size distribution, and the prepared NZVI showed high activity to degrade nitrobenzene in wastewater [32]. In another study, the prepared NZVI by high gravity technology exhibited a better degradation effect for reactive red 2 than the commercially available NZVI [33]. Those prior studies have demonstrated the feasibility of using high gravity technology to synthesize nanoparticles for contaminant degradation.

However, the reported methods still involve complicated procedures including washing, separation, dehydration and storage, especially synthesis and application of NZVI are in different steps/locations. This could lead to the risk of aggregation and oxidation of Fe^0 , which will decrease its reactive ability and thus limit the engineering application of Fe^0 . To solve the problem, in this study, we have developed a new method for continuous preparation of NZVI particles in the same reactor where degradation of NB simultaneously occurs. This method based on IS-RPB could synthesize NZVI from dissolved iron salts and sodium borohydride and then use it for *in situ* NB degradation. The specific objectives of this study were to: (1) demonstrate the feasibility of simultaneous formation of NZVI and NB degradation in the same reactor (IS-RPB); and (2) examine the impact of various key operational factors such as rotating speed, liquid flow rate, initial load of precursor compounds, and solution pH on NB degradation in IS-RPB.

2. Experimental

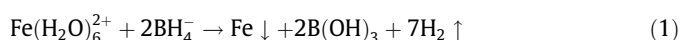
2.1. Chemicals and equipment

Analytic grade chemicals ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), ferric iron (FeCl_3), sodium borohydride (NaBH_4), sulfuric

acid (H_2SO_4), sodium hydroxide (NaOH), and NB were purchased (Tianli chemical reagent co., LTD, Tianjin, China) and used as received without additional treatment. Methanol (CH_3OH) was obtained from Kemiou chemical reagent co., LTD (Tianjin, China). All working solutions were prepared in deionized water. The equipment to prepare NZVI for NB degradation was IS-RPB, developed at Research Center of Shanxi Province for High Gravity Chemical Engineering and Technology (Shanxi, China). As shown in Fig. 1, this IS-RPB consists of packing (inter diameter with 60 mm, and outer diameter with 160 mm, axial height of packing with 60 mm, the volume of 1.32 L), shaft, motor, jet nozzle (jet nozzle with 1.5 mm diameter), shell (200 mm diameter), liquid inlet/outlet, gas inlet/outlet, and other elements. In IS-RPB, two streams flowing along the same axis in the opposite direction are brought into collision to ameliorate the liquid non-uniform distribution of the RPB. The IS surface reaches the inner brim of the RPB along the radial direction to collide each other, and the IS surface with a poorer mixing quality is further mixed with the aid of the RPB. The predominant mixing characteristic of the IS-RPB is that the end-effect in the inner radius of the RPB offsets the fringe effect of the IS, and then the fluid is mixed for the second time. Therefore, the IS-RPB is expanded from the gas-liquid to liquid-liquid fields with higher dispersion and turbulence by the coupling the IS and the RPB, which makes better micro-mixing and mass transfer in the liquid-liquid fields.

2.2. Chemical reaction in the IS-RPB reactor and measurement

NZVI was prepared by a reductive precipitation method in the IS-RPB, according to the following chemical reaction.



The prepared NZVI would react with NB according to the following equation [34]:



After the sufficient reaction in the IS-RPB, the samples were taken from the liquid outlet for measurement of the NB concentration using high performance liquid chromatography (HPLC).

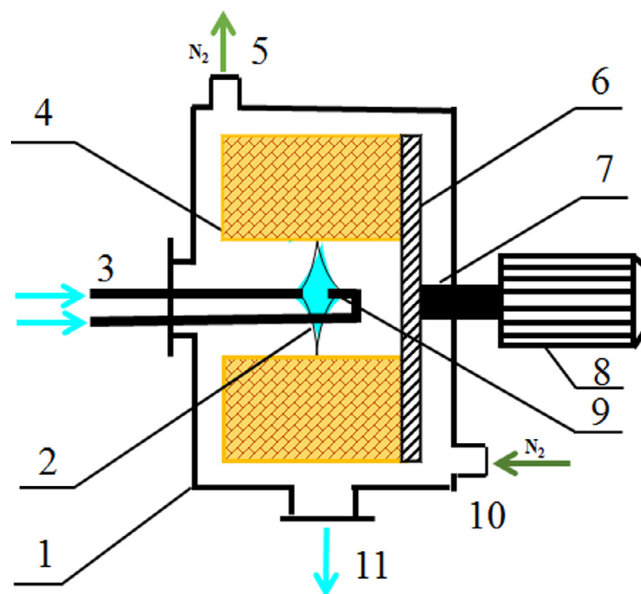


Fig. 1. Schematic diagram of the IS-RPB. 1. shell; 2. impinging ozone; 3. liquid inlet I and II; 4. packing; 5. N_2 gas outlet; 6. plate; 7. shaft; 8. motor; 9. jet nozzle; 10. N_2 gas inlet; 11. liquid outlet.

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