



# Highly effective iron–carbon–bentonite–alginate beads (Fe<sup>0</sup>/C-BABs) as catalyst to treat benzalkonium chloride in fixed-bed column systems



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## ABSTRACT

Novel iron–carbon–bentonite–alginate beads (Fe<sup>0</sup>/C-BABs) were successfully prepared with the ion-gelation method and demonstrated high activities in persulfate activation for benzalkonium chloride (BAC) oxidation. The beads were measured in terms of size, mass, and Fe content and then characterized via scanning electron microscopy, energy dispersive spectrometry, Fourier transform infrared spectroscopy, and thermogravimetric analysis. The performance of Fe<sup>0</sup>/C-BABs as filler was evaluated in a continuous fixed-bed with paralleled PS and BAC feeding. The effects of initial PS (100 mM) concentration, pH (3–11), fixed bead weight (10–30 g), and flow rate (225–540 mL h<sup>-1</sup>) were investigated. Results indicated that the Fe<sup>0</sup>/C-BABs effectively activated PS to treat BAC in the solution. The optimal conditions for the degradation of BAC were pH 7.0, [BAC] = 50 mg L<sup>-1</sup>, [PS] = 100 mM, bead weight = 30 g, PS flow rate = 25 mL h<sup>-1</sup>, and BAC flow rate = 225 mL h<sup>-1</sup>; the removal rate of BAC reached 93.3%. The economic cost analysis was carried out under the optimal experimental conditions. To deeply understand the degradation mechanism of pollutants, the intermediates of BAC degradation were analyzed via gas chromatography/mass spectrometry analysis, and a degradation path pathway was proposed.

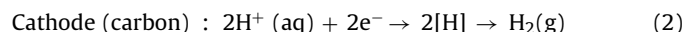
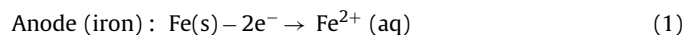
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## 1. Introduction

Benzalkonium chloride (BAC), as a cationic surfactant, is widely used in industries and households. However, it is a type of quaternary ammonium compound and known as the most dangerous compound for the ecosystem (Ivankovic and Hrenovic, 2010). The discharge of BAC wastewater that shows relatively high toxicity and biodegradation inhibition will pollute soil and water. The toxicity of BAC toward Gram-negative bacteria *Vibrio fischeri* and *Pseudomonas putida* (Awadhesh, 2008) and algae (such as *Chlorella vulgaris* (Chen et al., 2014), *Isochrysis galbana*, and *Chaetoceros gracilis*) (Perez et al., 2009) was confirmed by many studies.

For such recalcitrant compounds, the advanced oxidation process (AOPs) have been successfully developed and used because of their relatively high solubility, stability, non-selectivity, and activity (Antoniou et al., 2010). Persulfate (PS, S<sub>2</sub>O<sub>8</sub><sup>2-</sup>), as one type of

AOP, can be activated via heating (Hori et al., 2010), light exposure (Lau et al., 2007), or transition metal ions (Kislenko et al., 1997) to trigger the generation of free radicals. The generated radicals of SO<sub>4</sub><sup>2-</sup>• (E<sub>0</sub> = 2.6 eV) are more powerful than those of PS (E<sub>0</sub> = 2.1 eV), and relatively efficient in organic contaminant degradation (Huang et al., 2005). Among all the activation methods, Fe-C micro-electrolysis, an economical and environmentally friendly catalysis for PS activation, has attracted considerable attention in recent years (Ma et al., 2017). When iron and carbon are mixed in a solution, numerous microscopic galvanic cells form between the particles of iron (anode) and carbon (cathode) (Ying et al., 2012). This process promotes the activity of iron ions, as shown in Eqs. (1) and (2) (Yang et al., 2009).



However, Fe-C micro-electrolysis results in defects in homogeneous catalysts, such as sludge formation, Fe loss, and separation difficulties (Costa et al., 2010; Iglesias et al., 2015). As shown in Table 1, the pre-magnetized Fe<sup>0</sup> (Li et al., 2017), nanoscale zero-valent iron supported on biochar (Hussain et al., 2017) or carbon

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(Ramire et al., 2007) in the activated persulfate reaction system show serious iron loss, which can reach up to 40%, 30% and 10%, respectively. These disadvantages limit the wide utilization of these methods. To overcome those defects, sodium alginate was used to parcels zero-valent iron to reduce iron release, as reported by some researchers. As shown in Table 1, iron release was only 0.85% and 2.4% at 2 h and 4 h in the persulfate system. For example, sodium alginate has been mainly used as the supporter and framework for Fe-C or Fe immobilization (Lee and Mooney, 2012). Fe<sup>0</sup>/C-BABs reduce Fe loss during reactions and improve the recyclability of catalysts. Moreover, C-Fe loading can replace polyvinyl alcohol (Jang and Lee, 2016), bentonite (Benhouria et al., 2015), and montmorillonite (Barreca et al., 2014) to improve the strength and stability of sodium alginate. Although Fe/alginate beads have been used in the degradation of organic compounds in several studies on electro-Fenton or light Fenton processes (Iglesias et al., 2015; Titouhi and Belgaied, 2016; Ben Hammouda et al., 2016; Rosales et al., 2012), sodium alginate beads are rarely used in PS systems, with Fe<sup>0</sup>/C-bentonite-alginate beads (Fe<sup>0</sup>/C-BABs) being the preferable choice.

In the present study, Fe<sup>0</sup>/C-BABs were synthesized to degrade BAC from aqueous solutions. The bentonite-alginate beads (BABs) with iron powder and activated carbon powder on the cross-linked structure were prepared and packed in a fixed-bed column system. The influence of various experimental parameters, such as initial BAC concentration, flow rate, fixed bead weight, and initial PS concentration, on the removal efficiency of BAC in the fixed-bed column system was investigated. Fe release, radical generation, and pathway degradation were then clarified to further understand the underlying mechanism.

## 2. Materials and methods

### 2.1. Chemicals

Analytical grade sodium alginate, 1,10-phenantroline, carbon powder, and hydroxyl ammonium chloride were purchased from Sinopharm Chemical Reagent Co., Ltd. (China). BAC was obtained from Aladdin Industrial Corporation. Sodium PS, calcium chloride, bentonite, and iron powder were purchased from Shantou Xilong Chemical Co., Ltd. (China).

### 2.2. Synthesis of Fe<sup>0</sup>/C-BABs

Fe<sup>0</sup>/C-BABs were prepared in calcium chloride solution, which act as cross-linking agent. For the synthesis of Fe<sup>0</sup>/C-BABs, 2 g of bentonite (2% w/v), 0.2 g of activated carbon powder (0.2% w/v), 2 g of sodium alginate (2% w/v), 2 g of iron powder were added into deionized water. This mixture was stirred for 6 h to make it homogeneous. Then, the mixture dropped into the aqueous solution of calcium chloride. During this process, sodium ions in sodium alginate are replaced by calcium ions to form calcium alginate gels. To complete the ion exchange process (from Na<sup>+</sup> to Ca<sup>2+</sup>), the beads were soaked for 1 h in an aqueous solution of calcium chloride. Finally beads were washed with deionized for at least 3 times prior to practicing and then stored at room temperature. The average sizes of wet beads and dry beads were 4.66 ± 0.02 and 2.01 ± 0.04 mm, respectively (Table 2).

### 2.3. Column experiments

A diagram of a fixed-bed column is shown in Fig. 1. The fixed-bed column reactor was made of PVC material. The height and inside diameter of the column reactor were 40 and 2 cm, respectively. Before reaction, the fixed-bed column was filled with Fe<sup>0</sup>/C-BABs at different column weights (10, 20, or 30 g) for the advanced

**Table 1**  
Performance comparison with literatures.

catalyst	Reaction System	Contaminants	Experimental conditions	T (min)	Removal efficiency (%)	Iron release efficiency (%)	References
nanoscale zero-valent iron supported on biochar(BC)	persulfate	Nonylphenol	[NP] = 20 mg/L; [PS] = 5 mM; [nZVI]/[BC <sub>3</sub> ] = 0.4 g/L, pH = 7.0.	120	96.2%	30%	Hussain et al., 2017
Biochar (BC) supported nanoscale zerovalent iron (nZVI) composite	persulfate	Trichloroethylene	[nZVI] = 4.5 mmol L <sup>-1</sup> , [PS] = 4.5 mmol L <sup>-1</sup> , and [TCE] = 0.15 mmol L <sup>-1</sup> .	5	99.4%	31.3%	Yan et al., 2015
Carbon support Fe	H <sub>2</sub> O <sub>2</sub>	Azo-dye Orange II	[H <sub>2</sub> O <sub>2</sub> ] = 6 mM, [Catalyst] = 91.5 mg/L, T = 30 °C, pH = 3.	240	94.6	10%	Ramire et al., 2007
Iron/carbon-bentonite-alginate beads	persulfate	benzalkonium chloride	[BAC] = 50 mg L <sup>-1</sup> , [PS] = 100 mM, bead weight = 30 g, PS flow rate = 25 mL h <sup>-1</sup> , and BAC flow rate = 225 mL h <sup>-1</sup> ; pH = 7.	240	90.9%	2.4%	Present work
the metal-organic framework MIL-53(Fe) pre-magnetized Fe <sup>0</sup>	persulfate	Orange G	[OG] = 0.2 mM, [PS] = 32 mM, catalyst dosage = 1 g L <sup>-1</sup> , T = 25 °C, ambient pH.	120	98%	3.2%	Pu et al., 2017
Bentonite-supported nanoscale zero-valent iron	persulfate	2,4-dichlorophenol	[2,4-DCP] = 4 mg/L, [Fe] = 1.0 mM, [PS] = 1.0 mM, pH = 7.	60	100%	40%	Li et al., 2017
	persulfate	Cr(VI) and phenol	[Cr(VI)] = 0.38 mM, [nZVI] = 0.50 g/L, [B-nZVI] = 0.50 g/L, pH = 5.	120	99.30% and 71.50%	5.2%	Diao et al., 2016

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