

# Different iron salt impregnated granular activated carbon (Fe-GAC) for perchlorate removal: Characterization, performance and mechanism



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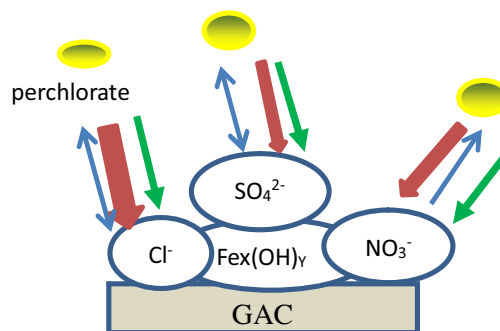
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## HIGHLIGHTS

- The type of iron salts impregnated on GAC play strongly affects perchlorate adsorption.
- FeCl<sub>3</sub>-GAC is most effective for perchlorate uptake in the wide pH range of 2.0–9.0.
- Electrostatic interactions are the key mechanism for perchlorate adsorption by Fe-GAC.
- Fe-GAC prepared with iron nitrate showed the lowest perchlorate uptake.

## GRAPHICAL ABSTRACT



power of attraction   
 exchange   
 reject   
 complex

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## ABSTRACT

A new reactive material (Fe-GAC) was synthesized and tested by impregnating various iron salts on granular activated carbon for perchlorate removal from water. SEM, FTIR and XRD characterization of Fe-GAC revealed that Fe-GAC had different morphology or structure and NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup> or Cl<sup>-</sup> attached. For Fe-GAC prepared with the same Fe concentrations but different iron salts, the perchlorate adsorption capacity (q) followed the sequence of: FeCl<sub>2</sub>-GAC (0.121 mmol/g) ≈ FeCl<sub>3</sub>-GAC (0.117 mmol/g) > Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>-GAC (0.09 mmol/g) > FeSO<sub>4</sub>-GAC (0.08 mmol/g) > Fe(NO<sub>3</sub>)<sub>3</sub>-GAC (0.06 mmol/g) > GAC (0.054 mmol/g). Furthermore, FeCl<sub>3</sub>-GAC was found more suitable for perchlorate adsorption in the wide pH range of 2.0–9.0. The type of iron salt impregnated on GAC played an important role in perchlorate adsorption, and electrostatic attraction was the main mechanism. NO<sub>3</sub><sup>-</sup> on Fe(NO<sub>3</sub>)<sub>3</sub>-GAC acted as strong competing ion with perchlorate, resulting in the lowest perchlorate capacity. The results may help to guide the selection of the optimal iron salt impregnated on GAC for perchlorate adsorption in continuous reactor (CTR) from contaminated water.

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## 1. Introduction

Perchlorate (ClO<sub>4</sub><sup>-</sup>), commonly used as a rocket fuel additive, has been inevitably released into the environment during

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the fabrication, operation and demilitarization of weaponry [1,2]. Meanwhile, perchlorate can remain stable in the environment for several decades due to its characteristics of high solubility, low-reactivity, and poor adsorbability by soil [3,4]. As an emerging environmental contaminant, perchlorate can inhibit the uptake of iodide by the thyroid gland and subsequently causes the malfunction of certain metabolic processes and results in a series of diseases, e.g. neurological damage and anemia [5]. In order to prevent the health risk from perchlorate pollution, United States Environmental Protection Agency (USEPA) adopted a drinking water equivalent level (DWEL) of  $0.2 \mu\text{mol/L}$  [6] in 2005.

A variety of water treatment technologies have been developed for removal of perchlorate from water, such as biological/chemical reduction [7–12], adsorption [13–25], ion exchange (IX) [26] and membrane processes [27]. Among these methods, biological/chemical reduction, ion exchange (IX) and membrane processes are significantly limited by their respective performance and/or economic drawbacks such as slow treatment rates, high energy consumption and production of waste residuals that required further disposal. Consequently, adsorption has attained increasing attention due to its affordable cost and high treatment efficiency.

Of various adsorbents tested so far, granular ferric hydroxide (GFH) and  $\text{Mg}/(\text{Al}-\text{Fe})$  hydrotalcite-like compounds have shown high affinity for perchlorate however, these materials are not suitable for uses in continuous reactors (CTR) [20,23]. In recent researches, Yang et al. [25] have developed the hydroxy-iron pillared bentonite (Fe-Bent) and found that Fe-Bent was the optimal perchlorate adsorbent among the materials of Al-Bent, Fe-Bent and Fe-Al-Bent; Our group have developed hydroxy-iron impregnated granular activated carbon (Fe-GAC) by hydrolysis of the ferrous sulfate ( $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ ) for perchlorate adsorption. The above composite materials can be used in continuous reactor (CTR). At the same time, Fe-GAC can be obtained in the nature by hydrolysis of the various kinds of iron salts such as ferrous sulfate ( $\text{FeSO}_4$ ), ferrous chloride ( $\text{FeCl}_2$ ), ferric chloride ( $\text{FeCl}_3$ ) and ferric nitrate ( $\text{Fe}(\text{NO}_3)_3$ ). Recently, several researchers reported that Fe-GAC was very effective for arsenic removal. For example, Munize et al. [28] impregnated iron on activated carbon using  $\text{FeCl}_2$  salt and in acidic conditions. Gu et al. [29] loaded iron onto activated carbon by applying an oxidation step (by  $\text{NaClO}$  or  $\text{H}_2\text{O}_2$ ) to convert ferrous sulfate to ferric ion. Fierro et al. [30] developed iron-doped activated carbons by forced hydrolysis of ferric chloride. Jang et al. [31] achieved deposition of iron hydroxide onto the surface of activated carbon via addition of ferric nitrate ( $\text{Fe}(\text{NO}_3)_3$ ). However, the preparation of iron-doped activated carbons for arsenic removal was very complex, and it remains unknown how the type of iron salts will affect the adsorption effectiveness. In the study, the preparation of Fe-GAC by hydrolysis of different iron salt under heat was very simple and economic. Therefore, it is a significant thing to the environmental protection to find an optimal iron salt for perchlorate removal.

In this study, a systematically comparative study was carried out to compare the perchlorate adsorption using Fe-GAC prepared using five types of iron salts. The specific objectives were to: 1) Prepare Fe-GAC using different iron salts and through the hydrolysis method, 2) characterize the materials by field emission scanning electron microscope (FESEM), Fourier Transform Infrared Spectroscopy (FTIR), X-ray diffraction (XRD) and zeta potential measurements; 3) Compare the adsorption kinetics, isotherms and pH effect on perchlorate removal; and 4) Elucidate the removal mechanisms and reasons for the effects of different iron salts on perchlorate removal.

## 2. Experimental section

### 2.1. Chemical reagents

Ferrous sulfate ( $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ ), ferrous chloride ( $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ ), ferric chloride ( $\text{FeCl}_3 \cdot 7\text{H}_2\text{O}$ ), ferric nitrate ( $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ ), ferric sulfate ( $\text{Fe}_2(\text{SO}_4)_3 \cdot 9\text{H}_2\text{O}$ ), sodium perchlorate ( $\text{NaClO}_4$ , Sigma), sodium hydroxide ( $\text{NaOH}$ ), ( $\text{HCl}$ ) and coal based GAC with grain sizes of  $105\text{--}148 \mu\text{m}$  were purchased from Sinopharm Chemical Reagent (Shanghai, China). All the chemical reagents were of analytical grade or higher and solutions were prepared by Milli-Q water ( $\text{Q-H}_2\text{O}$ , Millipore Corp).

### 2.2. Synthesis of Fe-GAC

The preparation method of different iron salt impregnated granular activated carbon followed the similar hydrolysis approach to the preparation of ferrous sulfate impregnated GAC reported by our group [19], except that five different iron salts ( $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ ,  $\text{FeCl}_3 \cdot 7\text{H}_2\text{O}$ ,  $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ ,  $\text{Fe}_2(\text{SO}_4)_3 \cdot 9\text{H}_2\text{O}$  or  $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ ) were used for the impregnation. As iron is the key component in perchlorate adsorption, the iron content was kept identical at  $2 \text{ mol/L}$  Fe during the material synthesis in all cases. For each case, 5 g of GAC were mixed with 50 mL of an iron salt solution in a 250 mL glass flask under strong magnetic stirring. After 24 h hydrolysis at  $100\text{--}120^\circ\text{C}$ , the solids were washed with DI water three times to remove impurities and dried at about  $120^\circ\text{C}$  for 4–6 h until its mass became constant. The dried Fe-GAC was stored in sealed bags prior to use.

### 2.3. Characterization

The surface morphology of the adsorbents was determined by FESEM (JSM-6701F, Japan). FTIR analysis was carried out to identify the function groups. To this end, the adsorbent materials were palletized with KBr powder. FTIR spectra were recorded in the range of  $400\text{--}4000 \text{ cm}^{-1}$  with a Nicolet 5700 spectrometer. XRD patterns of the adsorbents were determined with a Bruker AXS D8 Advance diffractometer system using  $\text{Cu K}\alpha$  ray ( $\lambda = 1.5418 \text{ \AA}$ ). The powder samples were scanned from  $10^\circ$  to  $85^\circ$ , using a step size of  $0.02^\circ$  and a run time of 1 s/step. The surface charges of Fe-GAC at pH 2.0–11.0 were determined using a zeta potential analyzer (ELS-8000, Phonal, Yokosuka Alec).

To determine the iron content of the Fe-GAC prepared in the same Fe concentrations but using different types of iron salts, 0.1 g of an Fe-GAC sample was added into 100 mL of a 1:1 HCl solution in a flask installed on a rotary shaker. Upon vigorous shaking (200 rpm) for 4 h, the iron concentration in water was determined by a Spectro Genesis Inductively Coupled Plasma-Optical Emission Spectroscopy (ICP-OES).

### 2.4. Perchlorate adsorption experiments

Batch adsorption kinetic tests were carried out in 250 mL conical flasks containing 200 mL of a  $0.1 \text{ mmol/L ClO}_4^-$  solution without pH adjusted. The pH before and after the reaction were monitored. The reactors were installed on a rotary shaker and the adsorption was initiated by addition of 0.2 g Fe-GAC or GAC into the solutions. The mixing speed was set at 200 rpm to simulate a complete mixed reactor. At the pre-designated sampling times, 3 mL samples were collected from the reactors and then filtered through a  $0.22 \mu\text{m}$  membrane filter. The filtrates were analyzed for perchlorate remaining in the aqueous phase.

Adsorption isotherms were constructed for each adsorbent following the same batch experimental procedures with an equilibrium time of 48 h. In a typical run, 0.1 g of Fe-GAC or GAC was added

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