



Bio-regeneration of spent Fe_3O_4 laden quaternary-ammonium shaddock peel after perchlorate capture: Considering the oxygen, coexisting anions, bio-fouling and indirect bio-regeneration



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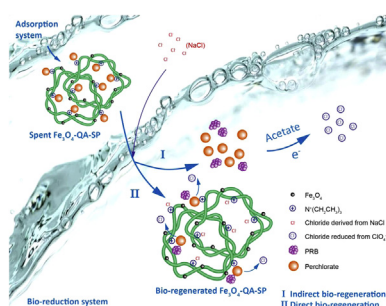
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HIGHLIGHTS

- A new bio-sorbent based on the shaddock peel was prepared for perchlorate capture.
- Bio-regeneration of perchlorate laden bio-sorbent considering oxygen was conducted.
- Bio-fouling on the bio-sorbent was evaluated.
- Indirect bio-reduction of spent bio-sorbent by adding amounts of Cl^- was conducted.

GRAPHICAL ABSTRACT



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ABSTRACT

A new bio-sorbent based on the shaddock peel was prepared for perchlorate capture. The shaddock peel was introduced with quaternary-ammonium groups and Fe_3O_4 nanoparticles, forming the Fe_3O_4 laden quaternary-ammonium shaddock peel (Fe_3O_4 -QA-SP). Bio-regeneration of perchlorate laden Fe_3O_4 -QA-SP considering the oxygen, coexisting anions, bio-fouling and indirect bio-regeneration was conducted. Results indicated that the bio-reduction rate increased as the increase in the amounts of perchlorate laden on Fe_3O_4 -QA-SP and the optimal reduction rates (0.0226–0.0865 $\text{mg}_p/\text{mg}_{ss}/\text{d}$) were obtained at pH 7.0. Bio-reduction of laden perchlorate in the presence of Cl^- (indirect bio-regeneration) help to keep a relatively high capture capacity (86.5%) of Fe_3O_4 -QA-SP for perchlorate even after 7 cycles of indirect bio-regeneration. In contrast, only 58.3% of capture capacity was left after 3rd of direct bio-regeneration. Bio-fouling was also observed on surface of Fe_3O_4 -QA-SP after sterilization process.

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1. Introduction

Perchlorate (ClO_4^-) has been considered as an emerging contaminant in groundwater and drinking water worldwide because different levels of ClO_4^- have been detected in the most industrialized countries, e.g. European Union, China, United States,

Japan, India [1–6]. The explosions of attention and interest in perchlorate recently include the advances in both the understanding of perchlorate's health impacts as well as analytical techniques. The salts of perchlorate were extensively employed in industries, e.g. fireworks, weapons and rocket propellants, lubricating oils and paints. The development in analytical chemistry have allowed detection at low part-per-billion (microgram per liter [$\mu\text{g}/\text{L}$]) concentrations, and the toxicological research indicated that such levels of ClO_4^- should be of concern because it would affect the thy-

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roid function for iodine uptake, which led to the decrease in the formation of thyroid hormones [6–9]. For this situation, 15 µg/L has been recommended as the preliminary remediation goal (PRG) for perchlorate by US EPA. However, there still has been a great deal of controversy, and uncertainty, regarding perchlorate's impacts on other organisms [10–12].

Various traditional physicochemical or biological technologies have been employed to eliminate ClO_4^- from water, e.g., chemical reduction, physical adsorption/ion exchange, and biological remediation [10,12,13]. Biological remediation is a cost-effective method for water with high levels of ClO_4^- . This process required the ClO_4^- as electron acceptor, and reduced the ClO_4^- into Cl^- ($\text{ClO}_4^- \rightarrow \text{ClO}_3^- \rightarrow \text{ClO}_2^- \rightarrow \text{Cl}^-$) using perchlorate reducing bacteria (PRB) in the presence of organic or inorganic electron donors (e.g., hydrogen, Fe(II), S^{2-} , lactate, acetate or methane) [2,12]. However, biological remediation was quite unsuitable for aqueous with low concentration of ClO_4^- . In addition, the key challenge of biological remediation is the lack of public acceptance for the introduction of microorganisms into drinking water systems.

In the physical adsorption methods, anion exchange resins cross-lined with quaternary amine groups have showed preferable capacity for ClO_4^- [14,15]. In contrast, adsorption capacity of activated carbon as well as its ion selectivity to perchlorate was poor. Therefore, a series of surface modification accesses have been employed to enhance the ClO_4^- adsorption capacity of activated carbon (e.g. oxidation, cationic surfactants, and ammonia-tailor) [16–19]. However, the disadvantage for adsorption application was the disposal of spent adsorbents and brine after desorption. Some previous reports indicated that regeneration of spent adsorbents and perchlorate decontamination can be made possible by integrating adsorption process with downstream biological reduction step [20–25]. Such integrated technologies have shown multiple merits as compared with the conventional technologies. For example, they have been proved to be robust and economically viable, and also they could reduce the laden perchlorate synchronously achieve the bio-regeneration of spent adsorbents [20]. Direct bio-regeneration of perchlorate resin as well as bio-reduction of the brine using PRB have been intensively investigated [15,26]. However, most relevant work were based on the application of commercial anion-exchange resins, few researches were related to the adsorption and subsequent bio-reduction of laden perchlorate on surface of modified biomaterials. In addition, it was a bit difficult to separate the bio-regenerated adsorbents from the biological system, and also the improvement of bio-regeneration capacity of spent adsorbents was required.

Considering all above problems, a new bio-sorbent prepared from the shaddock peel was applied for perchlorate capture and subsequent bio-regeneration process. The shaddock peel was introduced with quaternary-ammonium groups and Fe_3O_4 nanoparticles, forming the magnetic Fe_3O_4 laden quaternary-ammonium shaddock peel (Fe_3O_4 -QA-SP). Effect of coexisting anions and organic matters on perchlorate capture as well as the subsequent bio-regeneration of spent Fe_3O_4 -QA-SP was evaluated. Effect of oxygen on bio-regeneration of spent Fe_3O_4 -QA-SP by mixed PRB was also determined. Indirect bio-regeneration of spent Fe_3O_4 -QA-SP by adding amounts of Cl^- was also conducted. Biofouling on the surface of Fe_3O_4 -QA-SP as well as characteristics of virgin samples, spent samples and bio-regenerated samples were presented.

2. Materials and methods

2.1. All materials and Fe_3O_4 -QA-SP preparation

The shaddock peel was the residual by-product of shaddock obtained from Jinan, Shandong Province. The virgin shaddock peel

was dried at 105 °C until constant, and then it was smashed into particles with diameter of 0.2–1 mm. All used chemicals were obtained from Tianjing Kermel Chemical reagent Co., (China) with analytical reagent grade.

The seeds derived for mixed PRB cultivation were collected from the digester sludge in a paper mill in Zibo, Shandong Province (Bohui Paper Co., Ltd.). The detailed cultivation process was given in Appendix S1 and nutrient stock solution for culture enrichment was presented in Table S1.

The detailed preparation method for Fe_3O_4 -QA-SP was presented in Supporting Information Appendix S2.

2.2. Bio-regeneration of spent Fe_3O_4 -QA-SP at different conditions

2.2.1. Bio-regeneration of spent Fe_3O_4 -QA-SP considering coexisting laden anions and organic matters

Bio-regeneration of spent Fe_3O_4 -QA-SP considering coexisting anions (SO_4^{2-} , NO_3^- and F^-) and organic matters (humic acid, HA) included two steps: (i) perchlorate capture as a function of coexisting anions (SO_4^{2-} , NO_3^- and F^-) and organic matters (humic acid, HA), and (ii) the subsequent bio-regeneration of spent Fe_3O_4 -QA-SP in existence of these laden substances. Perchlorate capture was conducted by mixing 0.2 g of Fe_3O_4 -QA-SP with 50 ml of perchlorate solution (300 mg/L). Different amounts of NaNO_3 , Na_2SO_4 , NaF and humic acid were added into the suspensions to control the ionic strength and TOC (Total Organic Carbon) level. The adsorption was conducted at 25 °C with pH of 6.0 and contact time of 240 min so as to achieve the equilibrium of adsorption [27]. The adsorbed perchlorate was then determined using ion chromatograph (ICS-900, Dionex). Thereafter, the spent Fe_3O_4 -QA-SP was separated from the adsorption system. It was then mixed with 10 ml of mixed PRB and 30 ml of tap water with 10 g/L of $\text{CH}_3\text{-COONa}$ in a 125 ml of ground-glass stoppered flasks (pH 7.0). The flask was purged with N_2 for 5 min and then sealed. After shaking in an orbital incubator for 144 h (180 rpm, 30 °C), the Fe_3O_4 -QA-SP was separated from the bio-reduction system using a magnet. It was then sterilized with sodium hypochlorite (0.1 g/L) and reused for a second adsorption for perchlorate in the same adsorption conditions.

2.2.2. Bio-regeneration of spent Fe_3O_4 -QA-SP considering oxygen

In the oxygen test, 0.2 g of spent Fe_3O_4 -QA-SP samples (50 ml of perchlorate solution with concentration of 300 mg/L) were bio-regenerated in different aerobic/anaerobic conditions (anaerobic condition (144 h), anaerobic (48 h) + aerobic (12 h) + anaerobic (84 h), and anaerobic (48 h) + aerobic (96 h)). The Fe_3O_4 -QA-SP was separated, sterilized and then reused for a second adsorption for perchlorate in the same adsorption conditions to evaluate the recovery of adsorbed perchlorate.

2.2.3. Direct bio-reduction kinetic of laden perchlorate at different pH conditions

To evaluate the direct bio-reduction kinetic of laden perchlorate at different pH conditions, the spent Fe_3O_4 -QA-SP laden with different amounts of perchlorate (14.8–86.2 mg/g) was bio-regenerated at three pH conditions (6.2, 7.0 and 7.9). The bio-reduction time was conducted for 96 h. The recovery of adsorbed perchlorate as well as bio-reduction rates were then determined after the bio-regeneration.

2.2.4. Indirect bio-regeneration of spent Fe_3O_4 -QA-SP with different cycles

Indirect bio-regeneration with different cycles was also conducted by adding 0.5% of NaCl into the bio-reduction system. Adsorption capacity of perchlorate by the indirect bio-

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