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## Weak magnetic field accelerates chromate removal by zero-valent iron

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### ABSTRACT

Weak magnetic field (WMF) was employed to improve the removal of Cr(VI) by zero-valent iron (ZVI) for the first time. The removal rate of Cr(VI) was elevated by a factor of 1.12–5.89 due to the application of a WMF, and the WMF-induced improvement was more remarkable at higher Cr(VI) concentration and higher pH. Fe<sup>2+</sup> was not detected until Cr(VI) was exhausted, and there was a positive correlation between the WMF-induced promotion factor of Cr(VI) removal rate and that of Fe<sup>2+</sup> release rate in the absence of Cr(VI) at pH 4.0–5.5. These phenomena imply that ZVI corrosion with Fe<sup>2+</sup> release was the limiting step in the process of Cr(VI) removal. The superimposed WMF had negligible influence on the apparent activation energy of Cr(VI) removal by ZVI, indicating that WMF accelerated Cr(VI) removal by ZVI but did not change the mechanism. The passive layer formed with WMF was much more porous than without WMF, thereby facilitating mass transport. Therefore, WMF could accelerate ZVI corrosion and alleviate the detrimental effects of the passive layer, resulting in more rapid removal of Cr(VI) by ZVI. Exploiting the magnetic memory of ZVI, a two-stage process consisting of a small reactor with WMF for ZVI magnetization and a large reactor for removing contaminants by magnetized ZVI can be employed as a new method of ZVI-mediated remediation.

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### Introduction

Chromium can be present in different oxidation states in aquatic environments, of which the most stable states are the hexavalent (Cr(VI)) and trivalent Cr(III) states. Chromium and its compounds are extensively used in many industrial processes and products, such as metallurgy (as a corrosion inhibitor), paint pigments, antifouling agents, fungicides,

leather tanning, and electroplating (Baron et al., 1996; Hug et al., 1997; Naftz et al., 2002; Pratt et al., 1997; Puls et al., 1994). Leakage, poor storage practices and improper disposal of chromium waste have released Cr(VI), a potential carcinogen, into groundwater and surface water (Zazo et al., 2008). The high mobility of Cr(VI) in soils and aquifers (Guan et al., 2011) makes its environmental effect more significant. On the other hand, Cr(III) is much less toxic, and a trace level of Cr(III) is

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needed as an essential trace nutrient for animals and humans (Cespon-Romero et al., 1996; Guertin et al., 2004). The World Health Organization (WHO) and the Ministry of Health of China have established a provisional guideline of 50  $\mu\text{g/L}$  for Cr(VI) in drinking water (Guan et al., 2011). Thus, Cr(VI) should be effectively removed from industrial wastewater before discharge, or from Cr(VI)-contaminated natural water when it is used as a source for potable water (Gheju, 2011).

Cr(VI) removal by reduction to Cr(III) and subsequent precipitation, coprecipitation, or coagulation is well documented. Many chemical, electrochemical and biological methods have been proposed to reduce Cr(VI) to Cr(III) (Barrera-Díaz et al., 2012). In particular, zero-valent iron (ZVI) has been widely used as a reducing agent, especially for *in-situ* treatment of Cr(VI)-contaminated groundwater, since iron is readily available, relatively inexpensive, and nontoxic (Blowes et al., 1997; Liang et al., 2013). However, due to the formation of a passive layer of iron oxides on the surface of ZVI during the high-temperature manufacturing process (Ritter et al., 2002), ZVI has low reactivity toward Cr(VI). Moreover, the reduction of Cr(VI) by ZVI is a self-inhibiting process in near natural groundwater because the generation of (oxy)hydroxide films on the ZVI surface may block further access of Cr(VI) to the iron surface (Hu et al., 2010). Some methods including ZVI pretreatment (acid washing,  $\text{H}_2$ -reduction), dosing with complexing agents, and sonication have been applied to improve the removal of Cr(VI) by ZVI (Hung et al., 2000; Lai and Lo, 2008; Zhou et al., 2008). Although the aforementioned methods were able to enhance Cr(VI) removal by ZVI, the inconvenience and cost of these measures are major drawbacks in applying them. Moreover, the effectiveness of polishing pretreatments is problematic because iron (hydr)oxides will be rapidly produced on the ZVI surface before its contact with contaminants (Lai and Lo, 2008). One way to overcome this deficiency is to coat a small amount of another metal, nobler than iron (*e.g.*, Cu, Ag), onto the surface of ZVI. It has been reported that coating the surface of ZVI with copper not only significantly enhanced the Cr(VI) reduction rate but also increased the Cr(VI) removal capacity per unit weight of ZVI (Hu et al., 2010). However, there are still considerable concerns over the ecotoxicity and material cost of the noble metals (Crane and Scott, 2012), and the bimetal may be more inactive than pure ZVI (Kim and Carraway, 2000). Therefore, it is critical to explore an environmentally friendly method to significantly improve the reactivity of ZVI for Cr(VI) removal.

Our previous studies observed that Se(IV) removal by pristine or aged ZVI could be significantly improved by applying a weak magnetic field (WMF) ( $B_{\text{max}} < 20 \text{ mT}$ ) and the accelerated Se(IV) sequestration was accompanied with the accelerated corrosion of ZVI with a faster release of  $\text{Fe}^{2+}$  (Liang et al., 2014a, 2014b). Moreover, As(III)/As(V) removal by ZVI could also be considerably enhanced over a wide pH range upon the application of a WMF (Sun et al., 2014), ascribed to the improved ZVI corrosion in the presence of WMF. Cr(VI) is different from Se(IV), As(III) and As(V) because ferric (oxy)hydroxide generated in the process of Cr(VI) removal by ZVI can easily passivate ZVI. Although it is expected that the Cr(VI) removal by ZVI can also be accelerated in the presence of WMF, this speculation needs verification. It was proposed in our previous study that the enhanced contaminant removal by ZVI in the presence of WMF could be

mainly ascribed to the enhanced mass transport and localized corrosion induced by WMF. If this speculation is correct, the activation energy of contaminant removal by ZVI would not be affected by WMF, which has not yet been confirmed. Besides affecting the mass transport via the Lorentz force and field gradient force, the applied WMF may affect the morphology of corrosion products (Hinds et al., 2001) and thus affect the process of contaminant removal by ZVI.

Therefore, the major objectives of the present study were: (1) to investigate the effect of WMF on Cr(VI) removal by ZVI as a function of ZVI dosage, initial Cr(VI) concentration, pH, and magnetizing time; and (2) to understand the role of WMF in Cr(VI) removal process by determining the influence of WMF on the activation energy and corrosion products.

## 1. Materials and methods

### 1.1. Chemicals and materials

All chemicals used in this research were of analytical grade and used as received. All stock solutions were prepared with deionized (DI) water obtained from a Milli-Q water system. The Cr(VI) stock solutions were prepared by dissolving  $\text{K}_2\text{Cr}_2\text{O}_7$  in DI water. Iron powder with a median size of 32.1  $\mu\text{m}$  was obtained from Beijing Dk Nano technology Co., Ltd., China and used as received. All other chemicals were purchased from Shanghai Qiangshun Chemical Reagent Company. The magnetic field was provided by two pieces of neodymium-iron-boron permanent magnet on an iron sheet placed under the reactor. The magnetic flux density was measured by a Teslameter (Shanghai Hengtong Magnetic & Electric Technology Co., Ltd., Shanghai, China) and the maximum magnetic flux density was ca. 20 mT at the bottom of the reactor. This magnetic field was weak, and is referred as weak magnetic field or WMF hereafter. The experimental setup and details were schematically illustrated in our previous study (Liang et al., 2014b).

### 1.2. Batch experiments

Batch experiments were carried out to investigate the effect of WMF on Cr(VI) removal by ZVI as a function of ZVI dosage, pH, initial Cr(VI) concentration and temperature. The test solutions of Cr(VI) with varying concentrations were prepared freshly for each batch test in 0.01 mol NaCl and 0.10 mol sodium acetate. Sodium acetate was employed as a buffer to maintain constant pH conditions ( $\pm 0.1$ ) in the range of 4.0–5.5. Batch tests were started by adding ZVI to 500 mL of test solution, and the solution was mixed at 400 r/min with a mechanical stirrer (the aggregation of ZVI induced by the WMF could be avoided with intense stirring). The experiments were carried out in open reactors in the presence of WMF for 3 hr and at room temperature ( $(25 \pm 0.1)^\circ\text{C}$ ) unless otherwise specified. To estimate the activation energy of Cr(VI) removal by ZVI, batch experiments were conducted in the temperature range of 15 to 35°C at pH 5.0 by fixing the initial Cr(VI) concentration at 3.12 mg/L and ZVI dosage at 0.10 g/L. The permanent magnets placed under the reactor were immediately withdrawn after a specified magnetizing time (0, 3, 5, 10, or 120 min) to examine the effects of magnetizing time on Cr(VI) removal. At a fixed

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