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# Enhancing zero valent iron based natural organic matter removal by mixing with dispersed carbon cathodes



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#### HIGHLIGHTS

#### GRAPHICAL ABSTRACT

- Interspersed carbon cathodes increase the iron corrosion rate of zero valent iron.
- Coagulation and adsorption mechanisms dominate natural organic matter removal.
- Oxidation occurs to a less degree and forms biodegradable organic carbon.
- Empty bed contact times as low as 0.5 min could be effective.
- Graphite and granular activated carbon were both suitable cathode materials.

#### A R T I C L E I N F O

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#### ABSTRACT

Former studies have shown that adding granular activated carbon (GAC) cathodes could enhance the overall performance of the zero valent iron (ZVI) process for organics removal. The present study evaluates for the first time the performance of such an enhanced ZVI process to remove natural organic matter (NOM), an important water quality parameter in drinking water.

Lab-scale batch tests were conducted with surface reservoir feed water from a drinking water plant. In the GAC enhanced ZVI process dissolved organic carbon (DOC) and UV<sub>254</sub> were reduced by  $61 \pm 3\%$  and  $70 \pm 2\%$ , respectively, during 24 h treatment corresponding to 1.8 min empty bed contact time. The process was superior to ZVI alone, particularly during the earlier stages of the process due to the synergistically increased iron dissolution rate. Besides GAC, graphite and anthracite also prove to be suitable and potentially more cost-effective options as cathode materials for the enhanced ZVI process, whereby electrically conductive graphite clearly outperformed anthracite.

The dominant mechanisms in terms of NOM removal from surface water were found to be coagulation following iron dissolution and adsorption in the case of employing GAC. Oxidation was also occurring to a lesser degree, converting some non-biodegradable into biodegradable DOC.

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#### 1. Introduction

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The objective of drinking water treatment is to produce water that is chemically and microbiologically safe and aesthetically pleasing (Gray, 1994; Spellman and Drinan, 2012). Natural organic matter (NOM) has

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been reported to increase considerably in many areas (Matilainen and Sillanpää, 2010), which challenges drinking water treatment processes to meet increasingly strict water quality standards and raises the demand for advanced treatment. In particular, the presence of NOM affects disinfection efficiency and related to that acts as precursor for disinfection by-products (Jacangelo et al., 1995).

Therefore, several new technologies, such as UV-light based advanced oxidation (Bond et al., 2009; Wang et al., 2000), ozone based advanced oxidation (Chin and Bérubé, 2005; Von Gunten, 2003), activated carbon adsorption (Velten et al., 2011) and membrane filtration (Cho et al., 1999; Zularisam et al., 2006), have been developed and applied to improve the performance of conventional coagulation-filtrationbased water treatment processes. However, despite their effectiveness, due to the increased complexity and significant capital investments and operational costs, these technologies have achieved limited market penetration (Jacangelo et al., 1995; Matilainen and Sillanpää, 2010; Metsämuuronen et al., 2014). Therefore, innovative and effective, yet simple technologies are still in demand from drinking water utilities.

Zero valent iron (ZVI) has been used in drinking water treatment to remove arsenic (Kanel et al., 2005; Sun et al., 2006), reduce chlorinated organic groundwater pollution (Ma et al., 2012; Ruhl et al., 2012) and inactivate pathogens (Shi et al., 2012; You et al., 2005) based on its relatively cheap price and easy availability. With regard to NOM, some studies have shown that ZVI could remove NOM through adsorption and coprecipitation with iron corrosion products (Chiu, 2013; Wei et al., 2011). Fenton-like reactions were also reported on the ZVI surface, which may oxidise NOM (Kang and Choi, 2009; Keenan and Sedlak, 2008).

A recent review summarised the general limitations and countermeasures of the ZVI technology (Guan et al., 2015). Among them perhaps the most important drawback is a low intrinsic reaction rate due to mass transfer limitations imposed by the available surface, which is aggravated by gradually increasing passivation as iron corrosion products accumulate on the ZVI grains over time. Two of the countermeasures described in the aforementioned review aiming to overcome these limitations are adding complexing agents that reduce passivation of the ZVI surface by iron oxides (Keenan and Sedlak, 2008) and adding a second material acting as cathode promoting spontaneous galvanic corrosion accelerating the process (e.g. granular activated carbon (GAC)) (Wang et al., 2009; Ying et al., 2012).

When GAC is incorporated as a cathode material into this enhanced ZVI process, the electrode reactions can be represented as follows (Yang et al., 2009):

Iron anode (oxidation):

$$Fe(s) \to Fe^{2+}(aq) + 2e \ E^0 \Big( Fe^{2+} / Fe \Big) = -0.44V \tag{1}$$

$$Fe^{2+} \rightarrow Fe^{3+}(aq) + e \quad E^0 \Big( Fe^{3+} / Fe^{2+} \Big) = +0.77 V \,. \tag{2}$$

Carbon cathode (reduction):

$$2H^+ + 2e \rightarrow 2[H] \rightarrow H_2(g) \quad E^0(H^+/H_2) = 0.00V$$
 (3)

Carbon cathode (reduction) in the presence of oxygen:

$$O_2(g) + 4H^+(aq) + 4e \rightarrow 2H_2O \quad E^0(O_2/H_2O) = +1.23V$$
 (4)

$$O_2(g) + 2H^+(aq) + 2e {\rightarrow} H_2O_2(aq) \quad E^0(O_2/H_2O_2) = + \ 0.68V \eqno(5)$$

$$O_2(g) + 4H^+(aq) + 4e \rightarrow 40H^-(aq) \quad E^0(O_2/0H^-) = + \ 0.40V. \eqno(6)$$

To date, this ZVI process enhanced by GAC was mainly used to treat highly polluted wastewater, whereby the dissolved pollutants should ultimately be the primary electron acceptor (Lai et al., 2013; Ruan et al., 2010; Ying et al., 2012). From a technology point-of-view, the application in concentrated wastewater is very different compared to drinking water, which has a rather dilute nature. Then, partial reduction of the limited available dissolved oxygen may provide sufficient electron acceptors to promote the necessary reactions to achieve the treatment goal of NOM removal, including iron dissolution and generation of reactive oxygen species (Matilainen et al., 2010).

In drinking water treatment, to date only a couple of studies have been reported on the GAC-enhanced ZVI process. These evidence reductive removal of trichloroethylene (Song and Gao, 2014) and removal of arsenic contamination (Chen et al., 2012), but no study has been reported about how this enhancement impacts natural organic matter (NOM) removal during drinking water production from surface waters.

The aims of the present work are (i) to verify that under oxic conditions a significant acceleration of iron corrosion and consequential NOM removal is achieved by the enhanced ZVI process incorporating GAC; (ii) compare GAC with other, cheaper carbon cathode materials for NOM removal; (iii) verify if NOM removal occurs primarily due to adsorption and co-precipitation of iron corrosion products with NOM, or if oxidation plays also an important role.

#### 2. Material and methods

#### 2.1. Raw water

Raw water used in this study was collected from a surface water reservoir, which is the source water for a drinking water treatment plant in Southeast Queensland, Australia. 200 L raw water were collected onsite and concentrated with a lab-scale reverse osmosis system using 2.5" spiral wound reverse osmosis membrane (RO ESPA2540, Hydranautics, USA) to around 45 L (concentration factor of 4.44) for increased ease of storage in a cold room at 4 °C for future use. Prior to experimentation the concentrate was reconstituted with deionised water obtained from a Milli-Q Advantage system (see section on experimental procedures for detail).

#### 2.2. Materials

ZVI was purchased from Alfa Aesar (Australia) with a size of 1–2 mm and purity of 99.98% (metals basis). GAC (ACTICARB GA1000N) and anthracite (ACTICARB ANTHRACITE) particles were obtained from Activated Carbon Technologies Pty Ltd., Australia, and had a size of 1.2–2.4 mm. Granular graphite (1–2 mm) (El Carb 100) was provided by Graphite Sales Inc., USA. The rough bulk densities for ZVI, GAC, anthracite and graphite were 7.14 g/cm<sup>3</sup>, 1.43 g/cm<sup>3</sup>, 1.27 g/cm<sup>3</sup>/g and 1.28 g/cm<sup>3</sup> respectively. Before use, new GAC, anthracite, and graphite were rinsed with Milli-Q water (Millipore Pty Ltd., USA) several times to remove impurities, and then dried at 100 °C overnight. After cooling down to room temperature in a drying closet, these particles were stored in sealed bottles.

#### 2.3. Experimental procedures

All experiments were conducted in a flow-through column reactor with a diameter of 8 mm and total length of 120 mm, with a bed height of 10 mm corresponding to 0.5 cm<sup>3</sup>. The configuration of the reactor is shown in Fig. S1. According to former studies (Lai et al., 2012; Ying et al., 2012), the enhanced ZVI process achieved the best performance when the volumetric ratio of ZVI and GAC was 1:1. Hence, the same volume of ZVI and GAC were thoroughly mixed first, and then transferred to the column reactor where the bed height of 10 mm, corresponded to about 1.45 g ZVI and 0.23 g GAC. In each experiment, 90 mL of raw water concentrate were mixed with 310 mL Milli-Q water and the initial pH was adjusted to 7.0 or 3.0 with 0.5 N H<sub>2</sub>SO<sub>4</sub> or 0.5 N NaOH solutions. Then, the feed water was circulated through the bed with a flow rate of 40 mL/min for 24 h, which corresponded to a total empty bed contact time (EBCT) of 1.8 min over the whole experiment. For the enhanced ZVI process without dissolved oxygen (DO), the DO in the feed water Download English Version:

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