



Mitigation and degradation of natural organic matters (NOMs) during ferrate(VI) application for drinking water treatment



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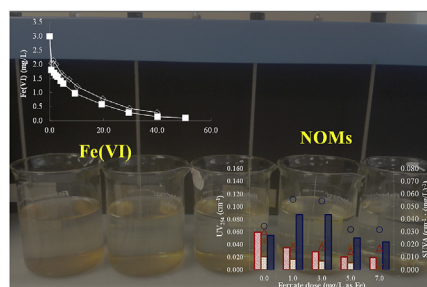
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HIGHLIGHTS

- Ferrate(VI) decay exhibited a pseudo 2nd order reaction pattern in a typical surface water sample.
- A sufficiently high ferrate(VI) dose was required to effectively destruct NOM molecules.
- An acidic condition accelerated ferrate(VI) decay and favored the NOM degradation.
- Ferrate(VI) oxidation is selective, effectively reducing UV₂₅₄ and SUVA, but poorly mineralizing NOM.
- Ferrate(VI) preferentially removes hydrophobic/transphilic NOM fractions and high MW molecules.

GRAPHICAL ABSTRACT



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ABSTRACT

Ferrate(VI), as an alternative for pre-oxidation in drinking water treatment, has recently captured renewed interest. However, the knowledge in ferrate(VI) chemistry remains largely undeveloped. The information regarding ferrate(VI) reactions with natural organic matters (NOMs), an important water matrix component affecting water treatment, is highly limited. In this study, bench scale tests were performed to study ferrate(VI) decay and reactions with NOMs in a typical surface water matrix. Results showed that ferrate(VI) decay exhibited a pseudo 2nd-order reaction pattern ($k_{\text{obs}} = 15.2 - 1.6 \text{ mM}^{-1} \text{ min}^{-1}$ and $36.3 - 4.0 \text{ mM}^{-1} \text{ min}^{-1}$ with $1.0 - 7.0 \text{ mg/L Fe(VI)}$ at initial pH 7.8 and 5.8, respectively), suggesting that self-decomposition is principally responsible for ferrate(VI) consumption. Ferrate(VI) tended to attack electron-rich moieties in NOM molecules, but had limited capability to mineralize NOMs. Consequently, ferrate(VI) effectively reduced UV₂₅₄ and specific UV absorbance (SUVA₂₅₄), but poorly removed dissolved organic carbon (DOC). Generally, lower pH and higher ferrate(VI) dose favored the NOM destruction. Fe(VI) (3.0 mg/L) could remove 16% of initial DOC (4.43 mg/L), 56% of initial UV₂₅₄ (0.063 cm^{-1}), and 48% of initial SUVA₂₅₄ ($0.033 \text{ cm}^{-1} (\text{mg/L})^{-1}$) at pH 5.80. Further organics analyses indicate that ferrate(VI) readily degraded hydrophobic and transphilic NOM fractions, but scarcely decomposed hydrophilic fraction. Fluorescence excitation-emission matrix (EEM) and fluorescence regional integration (FRI) analyses revealed that ferrate(VI) preferentially reacted with fulvic-like (region III) and humic-like (region V) substances and certain aromatic proteins (region II),

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difficultly decomposed soluble microbial byproducts (region IV), and rarely oxidized aromatic proteins in region I.

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1. Introduction

Interests in ferrate(VI), as a potential alternative for pre-oxidation and disinfection, for drinking water treatment has been gradually increased (Yang and Ying, 2013; Lee et al., 2014; Jiang, 2014; Jiang et al., 2015; Goodwill et al., 2015; Sharma et al., 2015). Ferrate(VI) is the oxyanion FeO_4^{2-} containing iron in +6 oxidation state. It can address different traditional and emerging water contaminants through multiple mechanisms, chemical oxidation in particular. Moreover, ferrate(IV) reportedly produces little toxic disinfection byproducts, and the end product (iron precipitates) is non-toxic itself, thereby making it an “environmentally friendly” water treatment chemical (Lee et al., 2004; Tiwari et al., 2007; Eng et al., 2006). Although the ability of ferrate(VI) for water treatment was early demonstrated in the 1970s (Waite and Gilbert, 1978; Waite, 1979; Gilbert et al., 1976), it has captured renewed interest as a “new”, green, and multi-purpose drinking water treatment agent only over the past decade (Yngard et al., 2007; Sharma et al., 2008; Anquandah et al., 2011; Ramseier et al., 2011; Sharma et al., 2013).

Different from other water treatment oxidants that have acquired considerable maturity, ferrate(VI) has lately gained sufficient attention, partially as a result of recent advances in ferrate(VI) synthesis. Although ferrate(VI) was intensively studied for removal of various traditional and emerging water contaminants, the knowledge in ferrate(VI) chemistry remains largely underdeveloped. Particularly, the information regarding ferrate(VI) removal of natural organic matters (NOMs) is very limited. NOM is a complex matrix of organic chemicals derived from natural sources (Crittenden et al., 2011). Understanding of reactivity of ferrate(VI) toward NOMs is crucial for drinking water treatment, at least, due to the following five reasons: 1) NOMs may be a principal sink of ferrate(VI) exerting a major fraction of ferrate(VI) demand; 2) the reaction rate of ferrate(VI) and NOMs can affect the formation rate of Fe(III) serving as the ensuing *in-situ* coagulant; 3) NOMs impact color to water, causing esthetic effects; 4) NOMs are potential precursors of certain disinfection byproducts (DBPs) that have health effects; and 5) NOMs can increase the solubility of metals in water through complexation.

Few efforts were made to study ferrate(VI) oxidation of NOM in water, most of which only focused on one or two NOM fractions such as humic acid (HA) and fulvic acid (FA) (Qu et al., 2003; Jiang and Wang, 2003; Lim and Kim, 2009; Graham et al., 2010; Gan et al., 2015). Qu et al. (2003) applied ferrate(VI) to oxidize FA extracted from the sediment of a Chinese reservoir. They reduced 90% of UV_{254} (2 mg/L FA) at a ferrate dose of 6.8 mg/L as Fe at pH 7.1–7.8. Of interest, they found that the optimal pH was between 8 and 9, because more ferrate(VI) was consumed by self-decomposition at a lower pH, while its oxidation potential was decreased at a higher pH. However, Lim and Kim (2009) reported that ferrate(VI) more readily removed NOMs at pH 3 than at pH 7.8 or 11. With 2–46 mg/L (as Fe) ferrate, they decreased UV_{254} by 21–74% for 10 mg/L HA and by 48–78% for 10 mg/L FA, separately, at pH 7.8. In contrast, TOC reduction was much lower (<20% removal for HA and <30% removal for FA under the identical conditions). Graham et al. (2010) also found that the DOC reduction was increased with a decreasing pH from 7 to 5 when ferrate(VI) was employed to oxidize HA in

three model waters and NOM in an England reservoir water, separately. Furthermore, they found that humic macro-molecular structures are cleaved into more hydrophilic fractions during ferrate(VI) oxidation of HA at pH 7. Jiang and Wang (2003) reported that ferrate(VI) performed better in terms of the DOC and UV_{254} reduction than ferrous sulfate during oxidation of HA (2–8 mg/L Fe(VI) at pH 6), and FA (2–16 mg/L Fe(VI) at pH 6 and 8).

Gan et al. (2015) further explored the reaction of NOM with ferrate(VI) to reduce the ensuing formation of disinfection byproducts. They oxidized Suwannee river natural organic matter (SRNOM) (3 mg/L DOC) at pH 7.0, and reduced DOC by 12% and 28% with 1 and 20 mg/L Fe(VI), respectively. They further found that a low ferrate(VI) dose (1 mg/L Fe(VI)) could not significantly reduce DOC in the 1–10 kDa and >10 kDa fractions, and a high dose (20 mg/L Fe(VI)) preferentially oxidized <1 kDa molecules and slightly decomposed >10 kDa molecules into 1–10 kDa compounds. With the fluorescence excitation-emission matrix (EEM) technique, they noticed that only 20 mg/L Fe(VI) could substantially diminish FA and HA-like substance-induced peaks; in contrast, the removal by 1 mg/L Fe(VI) was almost marginal. Although the work of Gan et al. (2015) provides more detailed findings on ferrate(VI) oxidation of NOM than previous studies, the information obtained is somewhat limited. Firstly, the species and oxidative capability of ferrate(VI) are actively pH dependent. However, only a pH level (pH 7.0), though it falls within a typical water treatment pH range, was tested, so that the effect of various pH levels is unclear. Secondly, their tests were all performed in the presence of phosphate buffer (10 mM). Such a high concentration of phosphate does not exist in real water sources in practice. Recently, Jiang et al. (2015) pointed out that phosphate buffer was able to considerably sequester Fe(III) during ferrate(VI) oxidation, inhibited the formation of iron decomposition products capable of catalyzing the ferrate(VI) decomposition, and slowed down the ferrate(VI) decay. Consequently, the behaviors of ferrate(VI) reduction in the presence and absence of phosphate buffer are not same or similar. Thirdly, the tested ferrate doses in their work were too low (1 mg/L) or too high (20 mg/L) compared with the ferrate dose during ferrate application. To sum up, most of the previous works merely targeted at ferrate(VI) oxidation of certain fractions of NOM, or were not performed under the experimental conditions closely approximating a water treatment scenario.

The objective of this study was to investigate the ferrate(VI) decay and the degradation behaviors of NOM and its different fractions during ferrate(VI) treatment. Effects of pH and ferrate (VI) dose on ferrate(VI) decay and NOM decomposition were evaluated. Two pH levels (5.80 and 7.80), both of which fall within a real water treatment pH range, and under which different active ferrate(VI) species were predominant (HFeO_4^- dominant at pH 5.80 and FeO_4^{2-} prevailing at pH 7.80), thereby facilitating our understanding of the effect of different ferrate(VI) species. Ferrate(VI) doses were tested within a water treatment relevant range (1.0–7.0 mg/L Fe(VI)). DOC, UV_{254} , and SUVA were employed as key parameters to assess the alternation of different NOM fractions during ferrate(VI) oxidation. Fluorescence analyses techniques were also applied to qualitatively and quantitatively investigate the reduction of different organic groups. Finally, implication of the ferrate(VI) reactions with NOM in drinking water treatment are further discussed.

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