



Research article

Performance of integrated ferrate–polyaluminum chloride coagulation as a treatment technology for removing freshwater humic substances



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ABSTRACT

Ferrate-based technologies can play versatile roles in water treatment because of their potential for *in situ* production and because they do not form any harmful by-products. We compared the oxidative performance of Fe(VI) generated by an electrochemical process, with H₂O₂-UV irradiation (a standard oxidation process) for removing Suwannee River natural organic matter. It took only 5 min for Fe(VI) (Fe: 1.67 × 10⁻⁴ M) to reduce the fluorescence intensity of a humic-like fluorophore by 36% of the original value; in contrast, it took 120 min of irradiation using H₂O₂-UV ([OH•] ~ 1.8 × 10⁻¹³ M) to remove 68% of the original value. In addition to the short reaction time, ferrate can also form aggregates that can remove turbidity and adsorb organics and other contaminants present in water. Simultaneous addition of ferrate and polyaluminum chloride (PACl) to unfiltered natural water displayed the most efficient reduction of UVA₂₅₄, primarily under acidic conditions. Ferrate pre-oxidation followed by PACl coagulation was the most effective process for reducing turbidity and chromaticity, because of the effects arising from the coagulation of ferrate that resulted in Fe(III)_(s) species. Ferrate pre-oxidation generated low-molecular-weight UVA₂₅₄-absorbing organics, whose dissolved organic matter (DOM) peak at 1250 Da was removed by PACl coagulation. Neither the initial pH nor the process order significantly affected the removal of organic carbon by the integrated ferrate-PACl process.

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1. Introduction

A number of oxidation processes (e.g., ozonation, ferrate: Fe(VI) oxidation, H₂O₂-UV, and vacuum-UV: VUV) are promising technologies for eliminating or partially oxidizing natural organic matter (NOM) in raw water. Ferrate exhibits the strongest oxidizing potential under acidic conditions (E⁰ = +2.20 V) among the oxidants used in water and wastewater treatment. In addition to being an effective disinfectant for a variety of microorganisms, ferrate is a selective oxidizing agent due to its high reactivity with electron-rich organic moieties of contaminants (Jiang, 2014; Sharma,

2013). Ferrate was shown to show selectivity for reacting with different disinfection by-product (DBP) precursors, similar to that of the highly selective electrophile, ozone (Jiang et al., 2016). Another advantage of ferrate oxidation is that Fe(OH)_{3(s)} produced from ferrate decomposition can serve as an *in situ* coagulant and adsorbent to promote the physical removal of pharmaceuticals and other contaminants such as DBP precursors (Jiang, 2014; Licht and Yu, 2005). The presence of various ferrate decomposition products in water might enhance the performance of ferrate (<1 mg/L) over ferric sulfate (~10 mg/L) for the removal of suspended solids and phosphorous and the reduction of chemical oxygen demand (COD) (Stanford et al., 2010).

Pre-oxidation means the addition of an oxidant (e.g., chlorine and permanganate) to raw water prior to clarification, and the specific addition of ferrate before clarification is referred to as ferrate pre-oxidation. Several researchers have investigated the effect of ferrate pre-oxidation on specific pollutants in wastewater and

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water treatment; however, the effect of integrating ferrate pre-oxidation with conventional water treatment processes to remove NOM is largely unknown. Past studies have reported that ferrate pre-oxidation enhanced the NOM removal during coagulation (e.g., Goodwill et al., 2016), while another study found minimal or variable effect (Graham et al., 2010). The discrepancies in the literature might be ascribed to variations in the NOM properties. Ferrate pre-oxidation prior to ferric chloride coagulation could improve the removal of UVA₂₅₄-absorbing compounds in continuous flow experiments (Goodwill et al., 2016) and DBP precursors in batch experiments (Jiang et al., 2016). Similar to ozonation and other strong oxidants, ferrate may alter the properties of NOM and influence its removal by subsequent coagulation (Graham et al., 2010; O'Melia et al., 1999).

Advanced oxidation processes (AOPs), including Fenton, photo-Fenton, ozonation, electrochemical oxidation, and photocatalysis, have been widely used for the treatment of a variety of water pollutants (Tunç et al., 2012, 2013). The hydroxyl radical (HO•) can be generated by AOPs from superoxides (e.g., hydrogen peroxide: H₂O₂), while H₂O₂-UV is a well-established AOP for organic abatement (Dotson et al., 2010; Legrini et al., 1993). HO• is considered a non-selective oxidizing agent and it reacts with several contaminants in water, including NOM. The overall process can lead to the mineralization of micropollutants via a series of degradative oxidation reactions. Wang et al. (2006) observed that for the H₂O₂-UV system to degrade NOM in water, the optimum H₂O₂ dose should be 0.0029–0.0147 M (0.01–0.05%) for humic acid oxidation. This resulted in the degradation of humic substances and formation of small-molecular-weight humic intermediate compounds. Nonetheless, because it is a new oxidation process for abatement of organics in water treatment, the performance of ferrate (a selective oxidant) has not yet been compared to that of oxidation by hydroxyl radicals (non-selective oxidant), which is a well-established method.

Therefore, the primary aim of this study was to compare the oxidative performance of Fe(VI), generated by an electrochemical process, with H₂O₂-UV irradiation (a standard oxidation process), for the removal of Suwannee River natural organic matter (SRNOM). UV irradiation was compared to H₂O₂-UV irradiation to see the effect of OH radicals on the degradation of dissolved organic matter (DOM). We compared the H₂O₂-UV treatment to H₂O₂-UV followed by anion exchange resin (AER), to determine whether the degraded (small molecular) DOM could be removed by polystyrene AER. To establish an effective regime for applying ferrate to water treatment systems for organic abatement, we evaluated the effects of process order and initial pH on the performance of a combined process of ferrate oxidation and PACl coagulation. This combination was used to treat natural surface water rich in aromatic NOM (i.e., humic substances), with moderate turbidity. To the best of our knowledge, this is the first study that combines ferrate pre-oxidation with PACl coagulation. Because ferrate is unstable at room temperature, the Fe(VI) solution was generated *in situ* by an electrochemical process that can be put to practical use for water treatment (Alsheyab et al., 2009).

2. Materials and methods

2.1. Water samples

Synthetic water samples were prepared with SRNOM (1R101N, International Humic Substances Society). A stock solution was prepared by dissolving SRNOM powder in Milli-Q water, followed by vigorous stirring with a magnetic stirrer for several hours. The solution obtained was passed through a 0.45 µm hydrophilic PTFE membrane to remove the undissolved fraction. The specific UV

absorbance at 254 nm (SUVA₂₅₄) of the SRNOM solution was 4.5 L/mg-m. Natural surface water sample was collected in November 2015 from the raw water intake (CH) of an advanced water treatment plant on a remote subtropical island of Japan, nearly 1000 km south of Tokyo. CH water had a turbidity of 5.15° (Japanese turbidity unit), chromaticity of 28.7°, pH of 7.4, and alkalinity of 46 mg CaCO₃/L. Dissolved organic carbon (DOC) of the CH water was 3.38 mg/L and UVA₂₅₄ was 0.118 cm⁻¹, which yields SUVA₂₅₄ of 3.5 L/mg-m.

2.2. Chemicals and reagents

H₂O₂ (30% w/w) from Wako Pure Chemical Industries (Japan) was used as an oxidant for the UV experiment. Polyaluminum chloride (PACl, [Al₂(OH)₅Cl]_m, m ≤ 10, Nippon Light Metal Co. Ltd., Japan), in concentration of 10 g/L, was prepared from the concentrate solution received from a full-scale water treatment plant.

2.3. Electrochemical ferrate production

The laboratory-scale experimental setup for the generation of ferrate comprised a 500 mL electrochemical reactor, steel plates as the anode and cathode, and a regulated DC power supply (PWR400L, 0–80 V, 25A, 400W, Kikusui Electronics Corp., Yokohama, Japan). The steel plate (B-77-P22, 50 × 150 × 2 mm, Yamamoto-MS Co. Ltd., Tokyo, Japan) used for the electrode had an Fe content of >99% and a C content of <0.1%. Ferrate was produced in a 12 M NaOH + 4 M KOH electrolyte solution at 60 °C. The electrodes therein were configured in a bipolar connection at the current density of 130–210 A/m² applied for 150 min (Barisci et al., 2014; Mácová et al., 2010). The ferrate solution was refrigerated until use. The Fe(VI) concentration was measured in a 1-cm-path quartz cell using spectrophotometry (UH5300, Hitachi, Japan) at 510 nm with a molar extinction coefficient of 1150 M⁻¹cm⁻¹, which has been used by several researchers (Homonnay and Perfiliev, 2004; Stanford et al., 2010).

2.4. UV irradiation and H₂O₂-UV experiments

LP UV lamps (2 × 15 W at 254 nm, GL Hitachi) were used in this study. A portion (500 mL) of SRNOM solution (initial DOC: 1.5 mg/L, pH 6.0) was placed in a glass Petri dish (114-mm inner diameter), resulting in a water depth of 49 mm. This was irradiated for 120 min at nearly 20 °C. The water sample in the Petri dish was continuously mixed using a magnetic stirrer during the exposure period. The fluence was measured with a radiometer, and afterwards, the UV dose was determined according to Bolton and Linden (2003).

Sarathy and Mohseni (2010) reported that the ranges of UV dose and H₂O₂ concentration feasible for producing drinking water include a fluence of less than 2 J/cm² and an initial H₂O₂ concentration < 15 mg/L (ca. 4 × 10⁻⁴ M). At higher H₂O₂ concentrations, HO• radicals react with H₂O₂ and produce hydroperoxyl (HO₂•) radicals, which are less reactive and do not contribute to the oxidative degradation of NOM. Considering this, UV fluence of 2 J/cm² with 10 mg/L H₂O₂ was applied to evaluate the H₂O₂-UV treatment in this study. The H₂O₂-UV experiment, using the same setup as the UV experiment, was performed by spiking H₂O₂ into SRNOM solution and immediately irradiating the solution for 120 min at nearly 20 °C.

2.5. Ferrate oxidation and combined coagulation-ferrate oxidation treatments

To assess the oxidative performance and coagulation effect of

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