



## Decolorization of azo dye Orange II by ferrate(VI)–hypochlorite liquid mixture, potassium ferrate(VI) and potassium permanganate

Guoting Li<sup>a,\*</sup>, Ninggai Wang<sup>a</sup>, Bingtao Liu<sup>a</sup>, Xiwang Zhang<sup>b</sup>

<sup>a</sup> Department of Environmental and Municipal Engineering, North China University of Water Conservancy and Electric Power, Zhengzhou 450011, China

<sup>b</sup> School of Civil and Environmental Engineering, Nanyang Technological University, 639798, Singapore

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### ABSTRACT

Ferrate(VI)–hypochlorite liquid mixture was prepared using hypochlorite, an industrial by-product, via wet oxidation method. Its oxidizing ability was investigated by decolorizing azo dye Orange II in batch experiments, and compared with potassium ferrate(VI) and potassium permanganate. Effects of the oxidant concentration, dye concentration, initial pH of dye solutions and UV 254 nm irradiation were examined. The color removal by potassium permanganate, potassium ferrate(VI) and the ferrate(VI)–hypochlorite liquid mixture at 30 min reached 17.7%, 62.0% and 95.2%, respectively. The ferrate(VI)–hypochlorite liquid mixture maintained a high decolorization efficiency over a wide pH range from 3.0 to 11.0, indicating that the initial solution pH had little impact on its oxidizing power. However, the decolorization efficiency by potassium permanganate was proved to be highly pH dependent and the lowest efficiency was observed at neutral pH. UV 254 nm irradiation did not enhance the decolorization efficiencies significantly for both the ferrate(VI)–hypochlorite liquid mixture and potassium permanganate over a wide pH range.

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### 1. Introduction

The conventional biological treatment methods are widely applied and considered to be suitable for sewage treatment because about two thirds of sewage contaminants are organic and biodegradable constituents [1]. However, biological treatment becomes ineffective especially for biological recalcitrant and toxic pollutants such as textile dyes, phenols and aniline [2,3]. In contrast, non-biological methods such as physical and chemical technologies have received ever-increasing concern around the world for the removal of these toxic pollutants for water and wastewater treatment. These physical technologies consist mainly of conventional phase separation techniques such as adsorption processes, ultrafiltration, reverse osmosis and air-stripping techniques. Nevertheless, these non-destructive techniques just transfer organics from water to another phase which needs further disposal of the solid wastes [4]. These more-destructive chemical methods are designed to destroy the contaminants by chemical oxidation or reduction with the aim to completely mineralize or transform the contaminants into harmful products. Therefore, chemical oxidation is always considered as an essential and fundamental step for water and wastewater treatment though the other methods are proved to be effective to some extent.

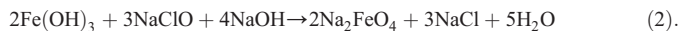
A series of chemical oxidants such as ozone, chlorine dioxide, chloramines, ferrate(VI) and permanganate have been widely studied due to their strong oxidation ability. However, some of these oxidants

can produce a large variety of by-products and the resulted by-products are proved to be toxic to the aquatic environment and human populations [5]. For example, ozone can form the potent carcinogenic bromate ion by reacting with bromide in water and monochloroamine produces N-nitrosodimethylamine (NDMA) which is suspected to be human carcinogen [6,7]. Potassium permanganate and ferrate(VI) has received much attention because they can avoid the formation of chlorinated DBPs and bromate from chlorination and ozonation. The use of potassium permanganate has been systematically studied and it is practically applied in some water treatment plants so far. However, ferrate(VI) salt is still a potential chemical reagent used in the field of water treatment. In fact, the oxidation potential of ferrate(VI) in acidic solution is the strongest of all the oxidants employed in water and wastewater treatment till now, and it is regarded as a dual-function chemical reagent with excellent oxidation, coagulation and disinfection ability [8–13]. The super-iron batteries using ferrate(VI) was reported to have a higher capacity and energy advantage compared to the conventional alkaline batteries indicating its peculiar oxidizing ability as well [14]. Ferrate(VI) can reduce the levels of THMs during water treatment and enhance the coagulation of surface water by preoxidation [15], but itself does not produce any toxic by-products when used. It has been proved to be effective for the application such as organic and inorganic treatment based on its excellent oxidation and coagulation capacity [9,16–21]. Therefore, ferrate(VI) is a promising oxidant to be put into practical application for environmental remediation.

Ferrate(VI) can be easily prepared by wet oxidation method. It only involves a simple reaction of a ferric and hypochlorite under high

\* Corresponding author. Tel.: +86 371 69127426; fax: +86 371 65790239.  
E-mail address: [liguoting@ncwu.edu.cn](mailto:liguoting@ncwu.edu.cn) (G. Li).

alkaline conditions. The basic chemical reactions are presented as the following equations [22]:



Chlorine is always employed for preparing hypochlorite in order to produce ferrate(VI) by wet oxidation in high alkaline media especially for lab study. Actually, hypochlorite is a by-product of industrial production and the cheap product can be directly used for preparing ferrate(VI) instead of chlorine. In this research, the ferrate(VI) solution was prepared by utilizing an industrial by-product hypochlorite, and then the ferrate(VI)–hypochlorite liquid mixture was directly used for dye decolorization without any purification. The oxidizing ability of potassium permanganate, potassium ferrate(VI) and the ferrate(VI)–hypochlorite liquid mixture was comparatively investigated. In addition, dye effluents are mostly resistant to biological treatment and constitute a considerable source of non aesthetic pollution and eutrophication. Decolorization by oxidation can improve the biodegradability of dye, which makes conventional biological processes available for further treatment [23]. About 60–70% of the dyes commercially available are bio-recalcitrant azo dyes. Orange II is a kind of soluble azo dye commonly used for decolorization and it was selected as a model compound in this study as well.

## 2. Materials and methods

### 2.1. Materials

Sodium ferrate(VI) was prepared by wet oxidation between hypochlorite (NaClO solution, 10%) and  $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  in high basic media at 50 °C [24]. Mechanical stirring was maintained for over 60 min. The resulted sodium ferrate(VI) solution (0.07 mol/L) is designated as the ferrate(VI)–hypochlorite liquid mixture unless otherwise stated in this paper. The dosage of ferrate(VI)–hypochlorite liquid mixture refers to that of pure sodium ferrate(VI) in the mixture. It was centrifuged to remove  $\text{Fe}(\text{OH})_3$  precipitate each time before being used and then diluted for chemical oxidation. Potassium ferrate(VI) was precipitated and isolated by adding KOH into the ferrate(VI)–hypochlorite liquid mixture, which was regarded as free of hypochlorite. It was stored in a refrigerator and used within a week.

Potassium permanganate and Orange II were purchased from Beijing Chemical Reagents Company. Orange II was selected as a model pollutant and used without further purification. The chemical structure of Orange II is illustrated in Fig. 1. Other materials used in the test were of analytical purity. Deionized and doubly distilled water was used throughout the study.

### 2.2. Decolorization of Orange II by the ferrate(VI)–hypochlorite liquid mixture and permanganate in the presence and absence of ultraviolet irradiation

The reaction was carried out in a series of beakers. The dye solution (50 mL) was stirred vigorously by a magnetic stirrer. The low pressure

UV lamp (15 W, Wenzhou Zhengtai Aosilan electric appliance Co., Ltd.), which had the main wavelength of 254 nm, was placed vertically 60 mm above the dye solution. The reaction was ceased with  $\text{Na}_2\text{SO}_3$  if needed and then used for spectrophotometer analysis. A series of stock Orange II solutions with different initial pH values were prepared and they were enough for the subsequent decolorization reactions involved. All the experiments were carried out at least in duplicate.

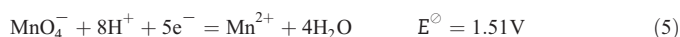
### 2.3. Analyses

The concentration of the dye was determined by measuring the absorbance at a fixed wavelength of 484 nm, which corresponds to the maximum absorption wavelength of Orange II in visible region. The concentration of ferrate(VI) was determined by measuring its maximum absorbance at 505 nm according to the literature [25]. The solution pH values were adjusted with NaOH or  $\text{H}_2\text{SO}_4$ . Analyses of Orange II and its degradation products were carried out on a high-performance liquid chromatograph (HPLC, Waters 996 photodiode array detector and 2695 separations module) analysis. The other experimental parameters can be referred to in our previous study [26].

## 3. Results and discussion

### 3.1. Orange II decolorization by the ferrate(VI)–hypochlorite liquid mixture and potassium permanganate

The color removal for Orange II using the ferrate(VI)–hypochlorite liquid mixture, potassium ferrate(VI) and potassium permanganate with the same dosage of 10 mg/L was compared and presented in Fig. 2a. It is observed that the color removal of 62.0% was achieved by potassium ferrate(VI), while it only reached 17.7% for potassium permanganate by 30 min, indicating the strong oxidizing ability of pure ferrate(VI). The oxidation potentials of ferrate(VI) and potassium permanganate are illustrated in the following reactions:



Potassium permanganate is another strong oxidant with high oxidation potentials but its oxidizing ability is not so strong as that of ferrate(VI), as presented by Eqs. (5) and (6). As we know, three electrons are transferred between  $\text{MnO}_4^{-}$  and  $\text{MnO}_2$  at neutral pH which is the same as that between  $\text{FeO}_4^{2-}$  and  $\text{Fe}(\text{OH})_3$ . The molecular weight of both oxidants is similar and the total number of electrons transferred is close as well. The above indicates that the difference of color removal basically resulted from the disparity of their oxidation potentials. Meanwhile, it is worthwhile to point out that the color removal amounted to as much as 95.2% using the ferrate(VI)–hypochlorite liquid mixture. Due to the residual hypochlorite in the ferrate(VI)–hypochlorite liquid mixture, the color removal was dramatically improved compared to pure potassium ferrate(VI). As a result, there may not be a great need to employ pure ferrate(VI) such as potassium ferrate(VI) in all the applications [27,28]. In addition, the direct use of newly prepared ferrate(VI) solution avoids the costly purification process and long time store despite its instability. On the contrary, potassium permanganate is stable enough for a long time store, which favors its wide application in water plant. Therefore, if high-purity ferrate(VI) is not a great need and there is no concern of chlorinated products, the ferrate(VI)–hypochlorite liquid mixture will

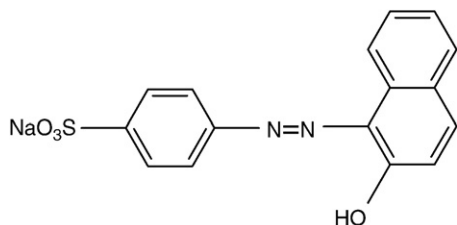


Fig. 1. Molecular structure of azo dye Orange II (C.I. 15510).

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