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The role of ferrate(VI) in the remediation of emerging micropollutants: a review

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ABSTRACT

The adverse effects of micropollutant residues in water on the environment and public could take place under a very low range of concentrations; from several μg/L to ng/L. Since there is no efficient unit process to remove these pollutants, efficient technologies are sought to treat them. Ferrate(VI) exhibits high oxidation/reduction potentials and has many advantages because of its dual functions of oxidation and coagulation. Removal of micropollutants by ferrate(VI) was pH dependant and this was in coordinate with the chemical/physical properties of the pollutants and ferrate(VI) speciation. Promising performance of ferrate(VI) in the treatment of real wastewater was observed. It is important to determine whether the ferrate(VI)-treated water contains any toxic or mutagenic substances as this should relieve public health concerns when a new chemical is employed for water treatment. The toxicity studies on the ferrate(VI)-treated effluent were carried out initially via Ames tests and recently via zebrafish embryos tests conducted at author's group. These results suggest that ferrate(VI) reagents do not produce mutagenic by-products for study conditions. However, a recent study showed the formation of adsorbable organic haloids as by-products in the ferrate(VI)-treated wastewater effluents. Obviously, more researches are needed to investigate the potential formation of harmful by-products during ferrate(VI) treatment. Other future work is suggested in order to implement ferrate(VI) into full-scale water treatment and other environmental remediation requirements.

Keywords: Emerging micropollutant; Environmental remediation; Ferrate(VI); Oxidation; Water treatment

1. Emerging micropollutants in water and wastewater

Pharmaceutical and personal care products (PPCPs) and endocrine disrupting chemicals (EDCs) are classified as emerging micropollutants, which have been a significant issue of environmental and public health concern because they may be significantly adverse environmental and human health effects although the occurrence of these pollutants in the environment is usually very low.

Pharmaceuticals such as antibiotics, anti-inflammatory drugs, β-blockers and X-ray contrast media are widely used, these pharmaceuticals and their

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metabolites cannot be fully utilized by human beings or animals and are inevitably emitted into the waters by excretion [1–4] and/or through the discharge of industry effluents and hospitals wastewaters [5,6]. Results of toxicology studies have revealed that some pharmaceuticals are suspected to have direct toxicity to certain aquatic organisms [7–9] and they could accumulate slowly, and finally lead to irreversible change on wildlife and human beings [10]. The adverse environmental and human health effects could take place under a very low range of concentrations; from several μg/L to ng/L.

EDCs are defined as the natural and/or synthetic compounds which would affect endocrine systems of fishes and other aqueous animals. Since the middle of last decade, a variety of adverse effects of EDCs on the endocrine systems of animals have been observed [11,12]. These effects may be cumulative, possibly will only appear in subsequent generations, and then the resulting effects may be irreversible, threatening the human's sustainable development. Most EDCs are synthetic organic chemicals being introduced to the environment by anthropogenic inputs (e.g. bisphenol A) but they can also be naturally generated estrogenic hormones e.g. estrone (E1) and 17β-estradiol (E2), and therefore, are ubiquitous in aquatic environments receiving wastewater effluents.

PPCPs represent a large group of compounds which include non-prescription and prescription pharmaceuticals for human and veterinary use, and the active and inert ingredients for personal care purposes. Examples of PPCPs include analgesics, lipid regulators, synthetic hormones, steroids, fragrances, sun screens, shampoos and cosmetics. Most PCPs, in their original or biologically altered form, are discharged into wastewater and make their way to wastewater treatment plants (WWTPs). Possible fates of PPCPs and their metabolites within a WWTPs are: (1) mineralization to CO_2 and water; (2) retention to the solids portion (sludge/biosolids) if the compound entering the plant or the product of biologically mediated transformation is lipophilic; (3) release to the receiving water either as the original compound or as a degradation product.

The presence of emerging micropollutants and their potential toxicity is a challenge to the global water industries as there is no unit process specifically designed to remove these pollutants; activated sludge and secondary sedimentation in most wastewater treatment works seems to be inefficient to eliminate them [5,13–16]. Thus, a number of recent studies have been carried out to explore suitable technologies to treat pharmaceutical residuals from water and wastewater [17] including ozonation, nanofiltration and reverse osmosis membrane filtration and activated carbon adsorption.

2. Ferrate(VI) and its application in water and wastewater treatment and in the degradation of emerging micropollutants

A promising technique which can address the concerns on emerging micropollutants is ferrate(VI) which exhibits dual functions of the oxidation and coagulation [18–22] and it has green chemical properties [23]. Ferrate (VI) has been successfully applied into water remediation processes [24–28] and to the oxidation of carbohydrates [29] and nitrogen-containing pollutants [30]. The removal of pharmaceuticals and other micro-pollutants by ferrate(VI) has been extensively studied [31–36].

2.1. General water and wastewater treatment by ferrate(VI)

Ferrate(VI) can act as coagulant, disinfectant and oxidant. An early study conducted by the author [20] demonstrated that in comparison with ferric sulphate (FS), ferrate(VI) can achieve the high removal of organic matter as DOC.

In a study [27] using ferrate(VI) for raw sewage treatment, much smaller dose of ferrate(VI) was required, in comparison with FS (Fe^{3+}), to efficiently remove suspended solids (SS), chemical oxygen demand (COD), biochemical oxygen demand (BOD) and phosphorous (P). Dashed lines in Fig. 1 are lower and upper limits of the required percentage removal for controlling quality parameters in post-sedimentation. The removal efficiency of SS, COD, BOD and P exceeded the high requirement by ferrate(VI) at lower doses.

Ferrate(VI) is efficient in inactivating *Escherichia coli*, total coliforms and f2 coliphage viruses [37]. Fig. 2 shows significant fast rates of *E. coli* inactivation by ferrate(VI) with smaller C_t values required to achieve 4-log inactivation at a low dose, 1.5 mg/L and in neutral pH range of 6.8–7.2 [38].

Ferrate(VI) was studied in comparison with ozone to kill protozoan pathogens such as *Cryptosporidium parvum* which are known as frequent cause of recent outbreaks of waterborne disease because of their strong resistance against chlorine disinfection. It was found [39] that ozone was superior over ferrate(VI); the large difference of the performance between ozone and ferrate(VI) in spore inactivation was attributed mainly to ferric iron produced from ferrate(VI) decomposition at the coat layer of spore which might coagulate spores and make it difficult for free ferrate(VI) to attack live spores.

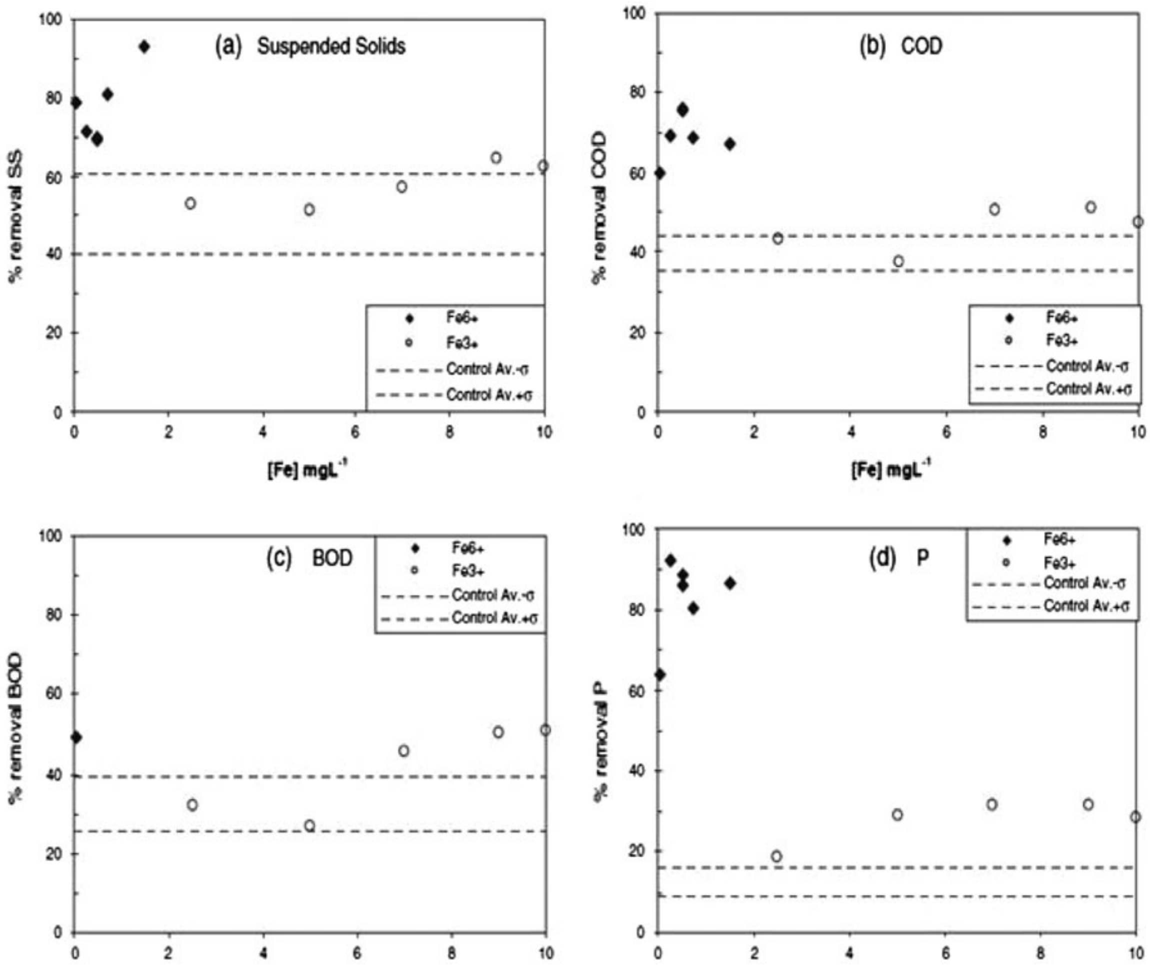

Fig. 1. Comparison of Fe^{6+} with Fe^{3+} for (a) SS, (b) COD, (c) BOD and (d) P% removal [27].

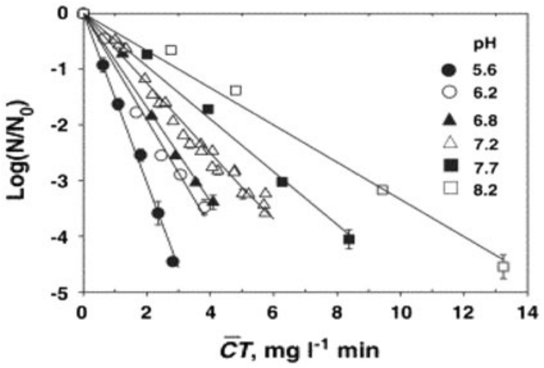

Fig. 2. Inactivation level of *E. coli* (log $[N/N_0]$) and exposure amount of *E. coli* to ferrate(VI) at various pH values [38].

Bacteriophage MS2 is a commonly studied surrogate for human enteric viruses detected in drinking water, demonstrated a major mechanism of ferrate(VI) disinfection [40]. The study suggests that the damage of both capsid protein and genome of MS2 caused by the attack of ferrate(VI) may contribute to the inactivation of Bacteriophage MS2; the efficiency of this was increased by increasing the ferrate(VI) access to the interior of the virion.

Ferrate(VI) has been used as an oxidant in conjunction with coagulation for algal removal [41]. The combined use of ferrate(VI) and alum, algal removal increased significantly in comparison with that using alum alone (Fig. 3). The study showed that ferrate(VI) oxidation resulted in the release of intracellular components of algae into the surrounding medium, which might behave as anionic and non-ionic polyelectrolytes [42] and act as coagulant aids, and therefore, improved algal removal by alum coagulation.

Arsenic (As) is a global concern as a pollutant of drinking water and groundwater (presented as As^{3+}) due to its significant toxicity. Considering that the removal of arsenic requires two steps that As^{3+} is to

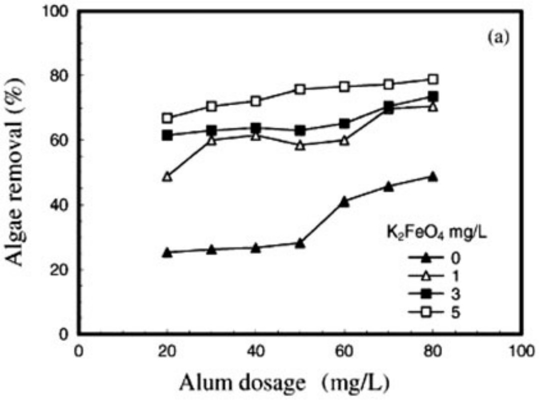

Fig. 3. Effect of ferrate(VI) oxidation on the removal of algae by alum [41].

be oxidized to As^{5+} first, and subsequent to be precipitated by the coagulating flocs, ferrate(VI) might be an ideal reagent for the removal of arsenic as it possesses dual functions of oxidation and coagulation. A number of researches [43–45] reported the stoichiometric relationship of As^{3+} oxidation by ferrate(VI) with a stoichiometry of 3:2 [As^{3+}:Fe(VI)] in order to achieve a satisfactory removal performance to comply the regulatory requirement.

Although potentials of ferrate(VI) to act as coagulant, disinfectant and oxidant for water and wastewater treatment have been widely investigated, researches will still be required to assess treatment efficiency and cost effectiveness considering simultaneously removing turbidity, dissolved organic carbon, particles and lowering residual iron, microbial activity in drinking water treatment and the removal of SS, COD, BOD, phosphorous and micropollutants in wastewater treatment.

2.2. *Application of ferrate(VI) for emerging micropollutant removal*

The ferrate(VI) efficiency to remove 68 selected EDCs and PPCPs spiked in a wastewater matrix was studied in two WWTPs [33]. Thirty-one target EDCs and PPCPs were detected in the effluents of the two WWTPs with concentrations ranging from 0.2 to 1,156 ng/L. Ferrate(VI) treatment resulted in selective oxidation of electron-rich organic moieties of these target compounds, such as phenol, olefin, amine and aniline moieties. But ferrate(VI) failed to react with triclocarban, three androgens, seven acidic pharmaceuticals, two neutral pharmaceuticals and erythromycin-H_2O.

In a recent study [46], selected pharmaceuticals were spiked into the effluent samples with concentration of 10 µg/L for each compound. Results showed that removal efficiencies of ciprofloxacin and naproxen were up to 70 and 50%, respectively, for ferrate(VI) doses up to 5 mg/L. Except ciprofloxacin and naproxen, raising ferrate(VI) dose did not improve the removal of other pharmaceutical significantly (Fig. 4). The relative high reactivity of ciprofloxacin and naproxen with ferrate (VI) may be attributed to electron donation by the methoxy group to the naphthalene moiety [33,45]. The acidic pharmaceuticals compounds (such as ibuprofen) showed less reactivity with ferrate(VI) because a carboxylic group is an electron-withdrawing functional group, which can depress the reaction of aromatic ring with ferrate(VI). Therefore, the reactivity of ferrate(VI) with carboxylic acids is usually slow.

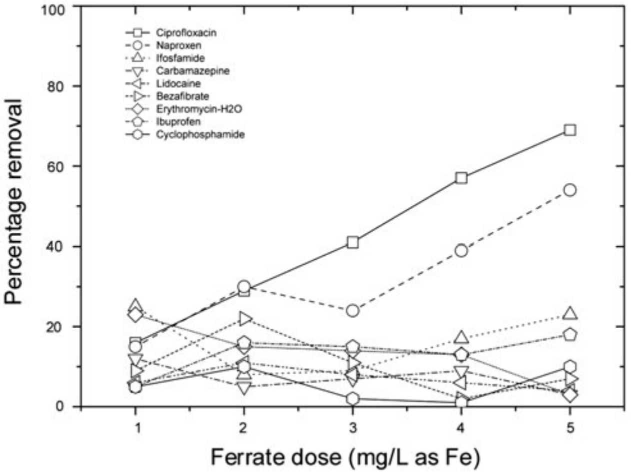

Fig. 4. Pharmaceuticals removal by ferrate(VI) at pH 6 [46].

The rate constant for the reaction of ferrate(VI) with selected EDCs and PPCPs can be seen in Table 1. The data indicate relative reaction activities between ferrate(VI) and various compounds. Ferrate(VI) can degrade most listed EDCs at rapid speed except for Buten-3-ol which has low reactivity with ferrate(VI). For the PPCPs, atenolol, carbamazepine and ibuprofen have low reactivity especially ibuprofen has the slowest reaction rate with ferrate(VI). On the other hand, the relative high reactivity of most EDCs and PPCPs with ferrate(VI) may be attributed to electron donation by the specific group to the naphthalene moiety [36,45].

3. Assessment of the toxicity of ferrate(VI)-treated water and wastewater

It is important to determine whether the ferrate (VI)-treated water contains any toxic or mutagenic substances as this should relieve public health

Table 1
Second-order rate constants k, $M^{-1}s^{-1}$ for the reaction of ferrate(VI) with selected EDCs and PPCPs

Group	Contaminant	pH	Rate constant k, $M^{-1}s^{-1}$	Half-life $t_{1/2}$	Refs.
Endocrine disruptors	17α-Ethinylestradiol	7	7.3×10^2		[47]
	17α-Ethinylestradiol	8	4.2×10^2		
	Bisphenol A	7	6.4×10^2		
	â-estradiol	7	7.7×10^2		
	Phenol	7	7.7×10^1		
	Bisphenol-A	7	7.7×10^2		[45]
	17β-estradiol	7	7.6×10^2		
	17β-estradiol	8	4.6×10^2		
	4-methyphenol	7	6.9×10^2		
	4-methyphenol	8	3.3×10^2		
	Buten-3-ol	7	12		
	Buten-3-ol	8	3		
PPCPs	Atenolol	8	7		[48]
	Bisulphite	8	1.90×10^4	0.7 s	[49]
	Bisulphite	7	8.24×10^4	0.2 s	
	Carbamazepine	8	16		[45]
	Ciprofloxacin	7	4.7×10^2	29.4 s	[47]
	Enrofloxacin	7	4.6×10^1	300 s	
	Ethionine	8	8.3×10^2	17 s	[49]
	Glycolic acid	8	4.0×10^{-1}	1.39 h	[50]
	Glycolic acid	12.4	7.2×10^2		[23]
	Glycylglycine	7	8.2×10^2	17 s	[51]
	Ibuprofen	8	0.4		[48]
	Iodide	7	6.67×10^3	2.1 s	[49]
	Iodide	8	1.54×10^3	9.0 s	
	Sulphamethizole	7	4.1×10^2	33.9 s	[52]
	Sulphamethoxazole	8	95		[45]
	Sulphamethoxazole	7	1.3×10^3	10.4 s	[52]
	Sulphisoxazole	7	1.5×10^3	9.2 s	

concerns when a new chemical is employed for water treatment. The Ames test has been applied to ferrate (VI)-treated water and a preliminary study demonstrated negative results [53], suggesting that ferrate (VI) reagents do not produce mutagenic by-products for the study conditions. Moreover, in a recent study by the author's group in Glasgow, UK, the toxicity of the ferrate(VI)-treated wastewater effluent was assessed and compared with that of raw wastewater effluent by the zebrafish embryos model. The zebrafish, a small tropical fish native to the rivers of India and South Asia [54], has become one of the most popular model organisms in developmental genetics and (eco) toxicology [55–57]. The zebrafish embryos represent an attractive model for environmental risk assessment of chemicals since they offer the possibility to perform small-scale, high-throughput analyses. The results of both mortality of zebrafish embryo tests and

Table 2
Harmful by-products formation in the ferrate(VI)-treated wastewater

Compound	By-product	Refs.
Carbohydrates	Aldehydes	[60]
Aniline	Azobenzene and nitrobenzene	[61]
Phenol	Pbenzoquinone and biphenols	[61]
Methanol	Formaldehyde	[62–64]
Carbamazepine	Aldehyde, ketone and carboxyl groups	[65]
Sulphamethoxazole	Methyl group compounds	[52]
Trimethoprim	3,4,5,-trimethoxybenzaldehyde and 2,4-dinitropyrimidine	[66]

microscopic images demonstrated that raw wastewater effluents possessed toxicity to zebrafish but ferrate (VI)-treated effluents had no adverse effects [58].

As shown in Table 2, a number of other studies have reported the potential formation of harmful by-products. Most recently, A study showed the formation of adsorbable organic haloids (AOX) as by-products in the ferrate(VI)-treated wastewater effluents although the AOX concentration rise was lower than that in the chlorination process [59].

Obviously, more researches need to be carried out to investigate the potential formation of harmful by-products during ferrate(VI) treatment. For example, it is to be studied under which operating conditions and for which original pollutants that harmful by-products would be formed.

4. Concluding remarks—feasibility of ferrate(VI) for the full-scale water and wastewater treatment

The high oxidation properties of ferrate(VI) were discovered a long time ago but systematic studies of oxidizing a number of organic pollutants using ferrate (VI) can be dated back to the early 1970s. Oxidation of micropollutants existing in water and wastewater by ferrate(VI) has been studied extensively since the beginning of this century. The advances in analytical chemistry theory and instrument allow tiny and low-level concentrations of micropollutants could be detected which helps the new legislated regulations could be set up and quality of water could be monitored. Water industries have to meet the requirement of stringent water and wastewater quality regulations and therefore, alternative technologies are sought by water industries.

A number of laboratory-based studies have investigated schemes and mechanisms, possible by-products formation and kinetics or rate constants of the degradation of micropollutants; these have advanced knowledge of the use of ferrate(VI) for the environmental remediation. In comparison with these fundamental studies, relatively smaller number researchers have focused on the practical application of ferrate(VI) for water and wastewater treatment or the environmental remediation such as odour removal and sewage sludge treatment. Nevertheless, a few of cases have been reported that ferrate(VI) has been used in full-scale applications so far. There are some fundamental issues which have not yet been studied thoroughly and are critical to implement ferrate(VI) into full-scale water treatment and other environmental remediation.

This author suggests following future work to be carried out:

(1) to classify and assess the toxicity of the potential degraded by-products when ferrate(VI) is used to oxidize various micropollutants;
(2) to study the effects of dosing points, dosing methods, dosing facilities and mixing schemes on the ferrate(VI) performance in water and wastewater treatment;
(3) to investigate the impact of water quality characteristics on the efficiency of disinfection and oxidation by the ferrate(VI);
(4) to assess the effect of ferrate(VI) dose and pH on the reduction of various micropollutants and on the inactivation of bacteria and virus in sewage sludge treatment and finally; and
(5) to carry out a full-scale trial to validate the treatment performance obtained in the laboratory studies and to evaluate economic suitability of using ferrate(VI) comprehensively.

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