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Research progress in the use of ferrate(VI) for the environmental remediation

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Abstract

The aim of this paper is to review the research progress of using ferrate(VI) in following fields of environmental remediation: (1) water disinfection; (2) degradation of synthetic organic pollutants; (3) treatment of emerging organic pollutants; (4) oxidation of inorganic pollutants; (5) removing humic substance; (6) wastewater treatment and disinfection; and (7) sewage sludge treatment. Whilst the superior performance of potassium ferrate(VI) as an oxidant/disinfectant for the environmental remediation has been demonstrated in various recent researches, challenges have existed to the implementation of ferrate(VI) technology in full-scale water, wastewater and sewage sludge treatment owing to either the instability property of a ferrate(VI) solution or a high preparation cost of a solid ferrate(VI). In addition to this, there are some fundamental issues which have not yet been studied thoroughly which are crucial for the implementation of ferrate(VI)—these lead to the future research work recommended by this paper.

Keywords: Potassium ferrate(VI); Disinfection; Oxidation; Degradation; Water and sewage treatment; Sludge treatment

1. Introduction

Oxidation and disinfection are two important unit processes for the environmental remediation. A wide range of oxidants/disinfectants can be used in the process. However, the common used chemical oxidants have limitations, one example of which is the formation of potential harmful disinfectant/disinfection by-products (DBPs) in chlorination and ozonation processes, and examples of DBPs are trihalomethanes [1] and bromate [2]. In order to meet the more stringent environmental regulations or standards of drinking water, an ideal water treatment chemical reagent should be developed and assessed, which could not form any harmful by-products in the treatment processes but give more efficient inactivation, degradation and oxidation of harmful micro-organisms and organic and inorganic micro-pollutants.

Potassium ferrate(VI) (K_2FeO_4) possesses all above functions. Under acidic conditions, the oxidation-reduction potential of the ferrate(VI) ions (2.2 V) is greater than that of ozone (2.0 V). As well as the high oxidation capacity, the ferrate(VI) also possesses the coagulation function by the formation of ferric hydroxide in the oxidation of other contaminants. Due to such a unique property, ferrate(VI) has re-attracted a great attention in last 10 years and various researches have been carried out in the use of it for the environmental remediation.

The aim of this paper is to review the progress of the application of ferrate(VI) in following fields: (1) water disinfection; (2) degradation of synthetic organic pollutants; (3) treatment of emerging organic pollutants such as endocrine disrupting chemicals (EDCs); (4) oxidation of inorganic pollutants; (5) removing humic substance from water resources; (6) wastewater treatment and disinfection; and (7) sewage sludge treatment. The limiting factors of using ferrate(VI) are discussed and future work in the field is recommended.

2. Disinfection with ferrate(VI) for drinking water treatment

2.1. The need of alternative disinfectants

Chlorination is the most common disinfection technology for potable-water treatment. Since the discovery of DBP in the use of chlorination and their potentially negative health effects [1], great efforts have been made to minimise the concentration of DBP by removing natural/synthetic organic compounds prior to disinfection, or removing the DBP after disinfection. However, this will greatly increase the overall cost of water treatment.

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Table 1
Redox potential for the oxidants/disinfectants used in water and wastewater treatment (after Ref. [7])

Disinfectant/oxidant	Reaction	E° (V)
Chlorine	$Cl_2(g) + 2e \leftrightarrows 2Cl^-$	1.358
	$ClO^- + H_2O + 2e \leftrightarrows Cl^- + 2OH^-$	0.841
Hypochlorite	$HClO + H^+ + 2e \leftrightarrows Cl^- + H_2O$	1.482
Chlorine dioxide	$ClO_2(aq) + e \leftrightarrows ClO_2^-$	0.954
Perchlorate	$ClO_4^- + 8H^+ + 8e \leftrightarrows Cl^- + 4H_2O$	1.389
Ozone	$O_3 + 2H^+ + 2e \leftrightarrows O_2 + H_2O$	2.076
Hydrogen peroxide	$H_2O_2 + 2H^+ + 2e \leftrightarrows 2H_2O$	1.776
Dissolved oxygen	$O_2 + 4H^+ + 4e \leftrightarrows 2H_2O$	1.229
Permanganate	$MnO_4^- + 4H^+ + 3e \leftrightarrows MnO_2 + 2H_2O$	1.679
	$MnO_4^- + 8H^+ + 5e \leftrightarrows Mn^{2+} + 4H_2O$	1.507
Ferrate(VI)	$FeO_4^{2-} + 8H^+ + 3e \leftrightarrows Fe^{3+} + 4H_2O^-$	2.20

Alternative disinfectants (e.g., bromine, iodine, chlorine dioxide, and ozone) have been thus considered to replace the chlorine. However, they form a range of other by-products, which are also considered to be toxic to some extent to the human population and to aquatic life. Owing to this, alternative disinfectants have been sought and studies been carried out.

Ferrate(VI) is a powerful oxidising agent. Under both acidic and neutral conditions, the redox potential of ferrate(VI) ions is greater than that of many other disinfectants (see Table 1), and it is potentially the strongest of all the oxidants/disinfectants realistically applicable to water and wastewater treatment.

2.2. *Disinfection performance of ferrate(VI)*

Pioneer studies of disinfection with ferrate(VI) in lab-scale [3–5] demonstrated a superior performance of ferrate(VI) in the inactivation of non-recombinant and recombinant *Pseudomonas*, *Escherichia coli* (*E. coli*), and a virus, f2 Coliphage. The results also showed that ferrate(VI) has sufficient disinfection capability to kill *E. coli.* At pH 8.2 and a dose of 6 mg/l as Fe, the *E. coli* percentage kill was 99.9% when the contact time was 7 min. The results also demonstrated that the disinfecting ability of ferrate(VI) increased markedly if water pH was below 8.0. The authors [5] found that ferrate(VI) can rapidly inactivate f2 Coliphage at low concentrations; 99% of f2 Coliphage was inactivated at 1 mg/l of ferrate(VI) in 5.7 min at pH 6.9 and only 0.77 min at pH 5.9. A higher dose (10 mg/l of ferrate(VI)) was required in order to achieve 99.9% inactivation at pH 7.8 with a contact time of 30 min.

The findings from these authors have been confirmed by the subsequent researches conducted either as pilot-scale trials in water industries [6–8] or at lab-scale experiments [9–12]. Fig. 1 shows the comparative disinfection performance at pH 8 of ferrate(VI) and sodium hypochlorite with ferric sulphate (FS) for a given contact time (30 min) and for various doses. It can be seen that under studying conditions, 4 mg/l FS (as Fe) with 10 mg/l Cl_2, or, 8 mg/l FS (as Fe) with 8 mg/l Cl_2, were required to achieve 100% inactivation of *E. coli*, whilst a small ferrate(VI) dose of 6 mg/l as Fe was needed to achieve the same target. The results demonstrated that in order to achieve 100% inactivation, the relative lower doses of ferrate(VI) was required in comparison with FS plus Cl_2.

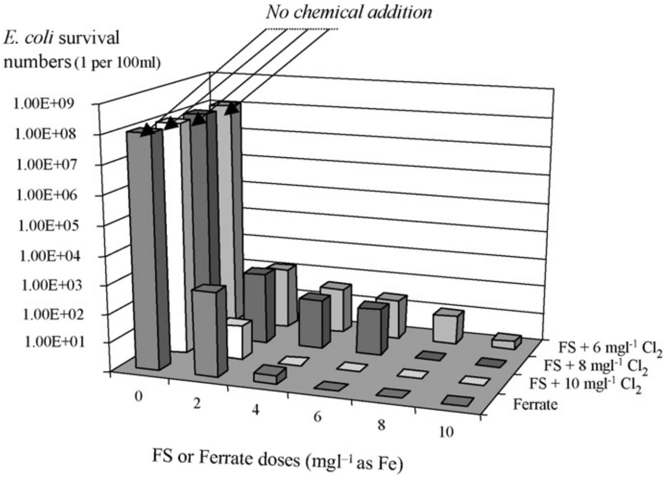

Fig. 1. Comparative disinfection performance of ferrate(VI) and ferric sulphate with chlorine at pH 8 (after Ref. [8]).

3. Degradation of synthetic organic pollutants

A range of organic contaminants has been shown to be readily oxidised by ferrate(VI). The organic compounds investigated were alcohol [13], aliphatic sulphur [14], amino acids [15], carboxylic compounds [16], organic nitrogen compounds [17], phenol and its related compounds [18,19], recalcitrant organics [20] and thiourea [21]. The percentage oxidation of these pollutants strongly depends on the dose of ferrate(VI); and overdoses of ferrate(VI) were proved to be most effective in reducing organic concentration [22]. In a case study [23], the maximum oxidation percentages with ferrate(VI) at pH < 8 were 18–47, 23–47, 85–100 and 32–55% for oxidising benzene, chlorobenzene, allylbenzene and phenol, respectively. The maximum oxidation efficiency was achieved when the molar ratios of the ferrate to organic impurities were in the range of 3:1–5:1.

For a pH range of 8–9 and the molar ratio of 5 as ferrate(VI) to pollutant, rate constants and half-lives of reactions between ferrate(VI) and selected pollutants could be as short as several seconds to minutes or as long as several hours, depending on the types of pollutants, and this can be seen in Table 2.

4. Treatment of emerging organic pollutants

Endocrine disrupting chemicals and drug related compounds are of current research interest owing to concern about their environmental impact. Pioneer studies using ferrate(VI) for the degradation of EDCs have been reported recently [27–30]. The oxidation of estrone (E1), 17β-estradiol (E2), and 17α-ethynylestradiol (EE2) by potassium ferrate(VI) was studied as a function of pH and dosages. The results suggest that pH 9 is the most favourable condition to obtain the highest removal efficiency and complete removal can be obtained at a molar ratio of ferrate(VI) to estrogens >3:1 in water samples (Fig. 2). A study [28] showed that in comparison with electrochemical

Table 2
Ferrate(VI) oxidation of pollutants at 25 °C

Pollutants	pH	k ($M^{-1}\,s^{-1}$)	$t_{1/2}$	Reference
Thioacetamide	9.0	5.6×10^3	0.36 s	[24]
Thiourea	9.0	3.4×10^3	0.59 s	[21]
p-Toluidine	9.0	1.3×10^3	1.5 s	[25]
Glyoxylic acid	8.0	7.0×10^2	2.9 s	[26]
Thiodiethanol	8.0	7.0×10^2	20.0 s	[26]
Phenol	9.0	8.0×10^1	25.0 s	[26]
p-Aminobenzoic acid	9.0	4.3×10^1	46.9 s	[25]
Methylamine	8.0	4.0×10^1	50.0 s	[26]
Nitriloacetic acid	8.0	2.0×10^0	16.7 min	[26]
Diethylamine	8.0	7.0×10^{-1}	47.6 min	[26]
Neopentyl alcohol	8.0	1.0×10^{-1}	5.55 h	[26]
Isopropyl alcohol	8.0	6.0×10^{-2}	9.26 h	[26]

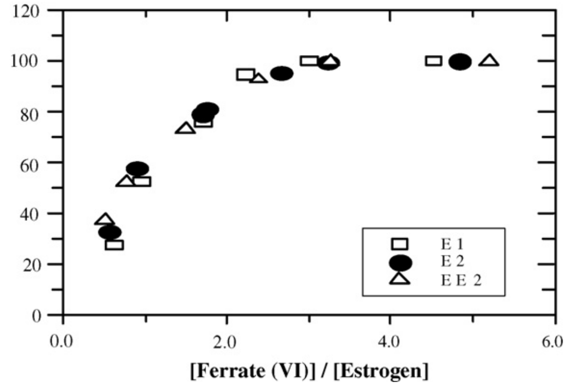

Fig. 2. Degradation of estrogens at pH 9, ferrate dose vs. removal percentage (after Ref. [27]).

oxidation, ferrate(VI) can reduce much more bisphenol A, E2 and 4-*tert*-octylphenol (Fig. 3).

The effectiveness of Fe(VI) for the oxidative removal of phenolic EDCs was also confirmed in both natural water and wastewater [29]. The apparent second-order rate constants for the reaction of Fe(VI) with selected EDCs (E2, EE2, and bisphenol A) ranged from 6.4×10^2 to $7.7 \times 10^2\ M^{-1}\ s^{-1}$ at pH 7. A study result [30] demonstrated that ferrate(VI) has the potential to be an oxidative chemical for removing sulphamethoxazole (SMX) in water.

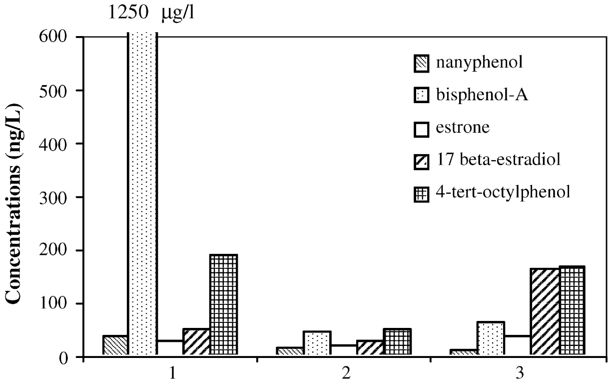

Fig. 3. Comparative EDCs residual concentrations. (1) Wastewater sample taken from the post-sedimentation; (2) treated sample with ferrate oxidation; (3) treated sample with electrochemical oxidation (after Ref. [28]).

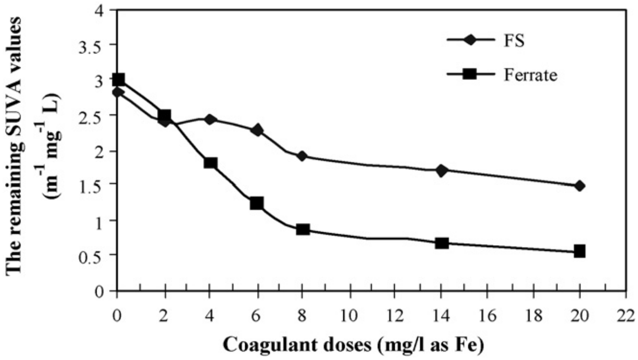

Fig. 4. Residual SUVA values in the treated effluent with ferrate(VI) and FS at pH 8, FA model water (after Ref. [40]).

5. Oxidation of inorganic pollutants

The oxidation of inorganic pollutants such as cyanide [31], ammonia [32], hydroxylamines [33] and hydrogen sulphide [34] was well presented. Potassium ferrate can also remove a range of metals (e.g., Mn^{2+}, Cu^{2+}, Pb^{2+}, Cd^{2+}, Cr^{3+}, and Hg^{2+}) to a low level at a dose range of 10–100 mg/l as K_2FeO_4 by oxidation and co-precipitation [35]. Arsenic(III) oxidation efficiency with ferrate(VI) was examined recently [36]. Under given test conditions, the mole ratio of Fe(VI) to As(III) and the reaction time were found to be important to achieve a high removal As(III) efficiency. As(III) was oxidised to As(V) (arsenate) by ferrate(VI), with a stoichiometry of 3:2 [As(III):Fe(VI)] [37]. Arsenic removal tests with river water showed that with minimum 2.0 mg/l Fe(VI), the arsenic concentration can be lowered from an initial 517 to below 50 μg/l.

6. Removing humic substances

Several studies [6,38,39] have demonstrated that potassium ferrate can perform better than ferric sulphate (FS) at lower doses for treating humic and fulvic acids (HA and FA) in terms of removing UV_{254} absorbance and dissolved organic carbon (DOC) and lowering the trihalomethane formation potential (THMFP). Fig. 4 [40] shows that the specific UV-abs (SUVA) values in the treated water with ferrate(VI) were much lower than that with FS for the same dose compared, indicating that ferrate(VI) can degrade FA first and the degraded organic matter could be easily removed by coagulation. More effective FA reduction was achieved in a pilot-scale trial in comparison with that by jar test studies [39]. Before these studies, a preliminary study was carried out [41] where sodium ferrate(VI) was studied for its performance to remove colour, iron and manganese.

7. Municipal wastewater treatment and disinfection

7.1. Overall treatment efficiency

A recent study [12] showed that for wastewater treatment, ferrate(VI) can achieve a high efficiency in the removal or reduction of aromatic organic compounds (as colour (Vis_{400}-abs)), COD and bacteria in comparison with coagulants of aluminium

Table 3
Comparative performance of wastewater treatment (after Ref. [12])

	Aluminium sulphate (AS)	Ferric sulphate (FS)	Potassium ferrate(VI)
pH	6.75–7.48	6.75–7.48	7
Optimum dose as Al or Fe (mM)	0.37	0.36	0.36
Turbidity removal (%)	80	86	94
Colour (Vis_{400}-abs) removal (%)	50	50	92
Total COD removal (%)	6	16	32
Bacteria reduction or inactivation (in $\log_{10}$ terms)[a]	1	1.05	>4

[a] AS and FS achieved $1-\log_{10}$ bacteria reduction at doses >0.50 mM as either Al or Fe, whilst ferrate(VI) achieved $>4-\log_{10}$ bacteria inactivation at doses <0.27 mM as Fe.

Table 4
The inactivation rate constant $-k$ in the disinfection of *E. coli* with sodium hypochlorite and potassium ferrate (after Ref. [42])

pH	$-k$ (min^{-1})					
	Dose = 4 mg/l as either Cl_2 or Fe		Dose = 6 mg/l as either Cl_2 or Fe		Dose = 8 mg/l as either Cl_2 or Fe	
	NaOCl	Ferrate	NaOCl	Ferrate	NaOCl	Ferrate
5.5	0.41	3.53	1.50	4.08	1.98	5.69
7.5	0.24	2.94	0.38	3.73	0.46	5.65

sulphate and ferric sulphate for the same or even smaller dose compared (Table 3). In addition, ferrate(VI) produced less sludge volume, which should then make sludge treatment easier.

7.2. *Comparative disinfection performance*

The superior disinfection performance of ferrate(VI) was also demonstrated by proposing disinfection kinetics using the Chick–Watson's rate law [42]. The rate constants were proposed in terms of the experimental results obtained. As shown in Table 4, that for both disinfection pHs (i.e., 5.5 and 7.5) and for different dosages, disinfection rate with ferrate(VI) was always greater than that with hypochlorite (as chlorine). In addition, the disinfection rate constant with hypochlorite was affected significantly by pH values of the solution; higher pH (7.5) gave lower k values due to decreasing the concentrations of HOCl but increasing OCl^- species when water pH increased. It has been well acknowledged that the disinfection efficiency of HOCl is 100 times greater than that of OCl^- and therefore, decreasing the concentration of HOCl results in less efficiency of disinfection. In contrast, the disinfection rate constant of the ferrate(VI) was slightly affected by pH increase (e.g., 7.5) at lower doses (4 and 6 mg/l as Fe) but was not affected by pH at higher dose (8 mg/l as Fe). In terms of the rate constant derived, potassium ferrate(VI) can achieve effective disinfection much faster than hypochlorite for a relative low dose and a short contact time, and the disinfection performance with ferrate(VI) could be less affected by pH values.

8. Sewage sludge treatment

In the development of municipal wastewater treatment strategies, the issues associated with the sewage sludge production are always taken into account. Toxic pollutants together with a large number of pathogens are concentrated in the sludge, and this increases the risks to the health and environment. Moreover, a number of organic sulphides and amines are produced in wastewater treatment which results in unpleasant odours. Complaints of illness related to the land application of biosolids have been increasing, and the original application of the sludge as a fertiliser in agricultural systems has thus become increasingly under pressure. The legislation and regulations regarding the application of sludge in agriculture have changed considerably (e.g., EU Directive on sewage sludge). The most important new aspects are the requirement of sludge hygienization and odour reduction using advanced treatments.

Due to all these, there is a need of innovative sludge technologies, which could not only effectively treat a wide range of contaminants and health hazardous pathogenic organisms, but could also remove unconventional contaminants (e.g., personal care products and endocrine disruptors) from sewage sludge.

Ferrate(VI) has been observed to be superior in disinfecting coliforms in sewage sludge [43]. With a small dosage of ferrate(VI) (20 ml or 0.4 g ferrate per 2 kg of sludge), complete inactivation of coliforms was achieved (Fig. 5). Oxidation of sludge by ferrate(VI) to remove odour-causing compounds such as hydrogen sulphide, mercaptans and amines has been studied

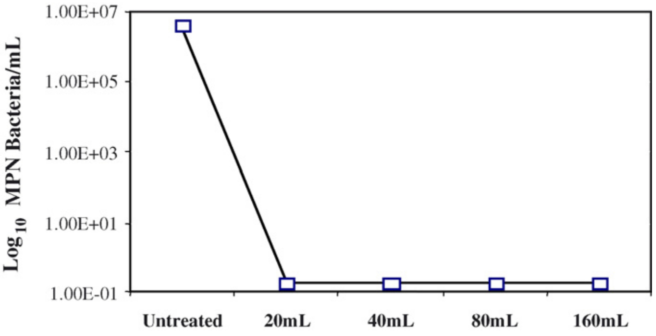

Fig. 5. MPN counting of coliforms in sludge with ferrate (after Ref. [43]).

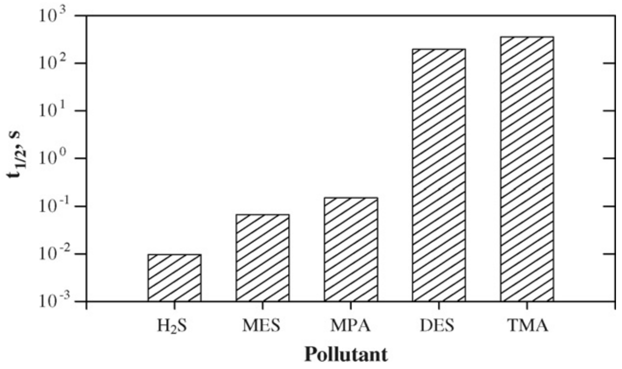

Fig. 6. Half-lives of the reactions between Fe(VI) (500 mM) and pollutants (100 mM) at pH 9 and 25 °C. MES: 2-Mercattoethanesulphonic acid; MPA: 2-mercaptopropionic acid; DES: diethylsulphide; TMA: trimethylamine (after Ref. [43]).

Table 5
The dosage required to remove 90% H_2S (after Ref. [45])

Chemical	Dose as chemical to S^{2-} (g:g)
Ferrous iron	2.2
Ferric iron	1.7
Ferrate(VI)	0.4

[44]. The reaction rate law and observed rate constants at pH 9 were used to determine half-lives of the oxidation processes (Fig. 6). The half-lives of the reactions vary from milliseconds to seconds and ferrate(VI) tends to react faster with sulphur-containing pollutants than with amines (Fig. 6).

In a study [45], the dose requirement for the removal of 90% sulphide from wastewater sludge with ferrous and ferric irons and ferrate(VI) was compared and this can be seen in Table 5. It is evident that using ferrate(VI) to replace ferrous and ferric iron, the required dose was reduced by 80 and 76%, respectively, which significantly reduce the sludge production and therefore, the sludge treatment cost.

Dewatered sludge was treated with four different doses of ferrate(VI) and the odour intensity and hedonic tones were quantified (Fig. 7). From the experiment, sludge treated with 40 ml ferrate(VI) produced least odour, which was one third of untreated sludge. The trend of hedonic tone also confirmed that sludge treated with ferrate(VI) of 40 ml was the best in terms of odour reduction. In comparison, lime treated sludge still produced similar level of odour with untreated sludge [43].

9. Toxicity assessment of the ferrate(VI) treated water

It is important to determine whether the ferrate(VI) treated water contains any toxic substances as this should relieve public health concerns when a new chemical is employed for water treatment. The Ames test is used to screen for the existence of mutagenic compounds. If a positive result is obtained, mutagenic compounds are considered to be present. It is claimed that about 90% of known carcinogens can be shown to be mutagens by use of Ames test [46]. The Ames test was applied to ferrate(VI) treated water and a preliminary study demonstrated negative results [47], suggesting that ferrate(VI) does not produce mutagenic by-products for the study conditions used. However, systematic studies need to be conducted to confirm that there are no mutagens to be produced in treating different types of water with ferrate(VI).

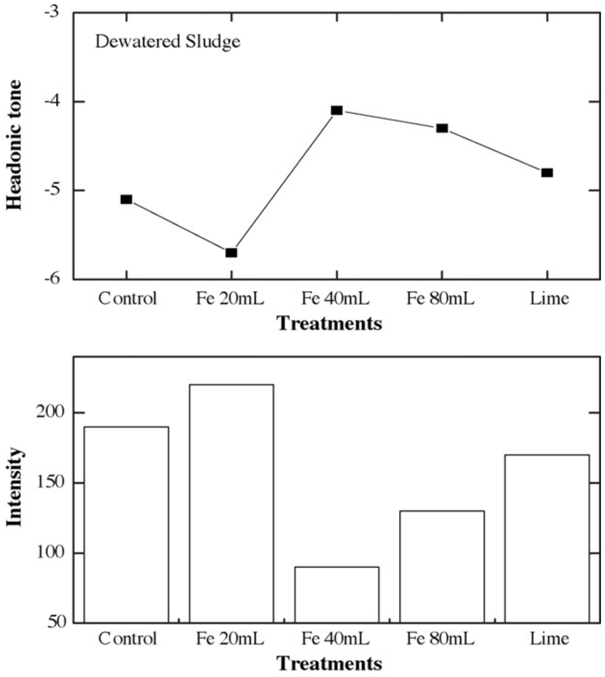

Fig. 7. Odour intensity and hedonic tone of sludge with ferrate(VI) and lime treatment (after Ref. [43]).

10. Discussions and concluding remarks

The superior performance of potassium ferrate(VI) as an oxidant/disinfectant in the environmental remediation has been demonstrated in various recent researches. Especially, the superior performance of ferrate(VI) in the treatment of emerging organic pollutants, humic substances and arsenic(III) provide alternatives to ozonation or other advanced oxidation processes. Challenges have existed, however, to the implementation of ferrate(VI) technology in the full-scale treatment of water, wastewater and sewage sludge owing to either instability property of a ferrate(VI) solution or high preparation cost of a solid ferrate(VI) product. Therefore, further work should be carried out aiming to prepare the ferrate(VI) with high stability but in a low manufacturing cost.

Although a number of researches have been conducted in recent years to study the overall efficiency of ferrate(VI) as an oxidant or a disinfectant in water and wastewater treatment, 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. The author suggests following future work to be carried out:

(A) To classify and assess the toxicity of the potential degraded by-products when ferrate(VI) is used to oxidise various micro-pollutants;

(B) to investigate the inactivating capability of ferrate(VI) in treating different types of harmful micro-organisms such as Cryptosporidium;
(C) to study the effects of dosing points, dosing methods, dosing farcicalities and mixing schemes on the ferrate(VI) performance in water and wastewater treatment;
(D) to investigate the impact of water quality characteristics on the ferrate(VI) efficiency as a disinfectant and as an oxidant;
(E) to assess the effect of ferrate(VI) dose and pH on the reduction of heavy metals and various micro-pollutants and on the inactivation of bacteria and virus in sewage sludge treatment with ferrate(VI), and finally;
(F) to carry out a full-scale trial to validate the superior treatment performance obtained in the laboratory studies and to evaluate economic suitability of using ferrate(VI) comprehensively.

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