



Mechanistic Insight into Disinfection Using Ferrate(VI)

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Abstract

Disinfection, chemical oxidation, and coagulation are key methods in water treatment. A chemical that may be used for all these targets is ferrate(VI). This work tries to bring some light into mechanisms implied throughout killing microbes using ferrate(VI). In acidic pH, the oxidation and reduction capacity of ferrate(VI) is superior to all currently utilized oxidizers and disinfectants in water and wastewater treatment. The technology of using ferrate(VI) for coagulation, chemical oxidation and disinfection of water and wastewater in a reactor simultaneously, can reduce the size of water and wastewater treatment plants and increase the treatment efficiency. Ferrate(VI) allows a novel emergency water treatment design for disaster-affected populations through the repercussions of natural disasters, through the concurrent and efficient elimination of microbial and chemical pollutants. The ferrates' elevated performance and utilization as a green element for water treatment propose that these techniques remain greatly convenient for usage as pre- or post-treatment in traditional wastewater plants. It seems that there is no big difference in terms of ferrate's action on pathogens with other chemical disinfectants. However, ferrate was found to be requiring lower needed doses as compared with other chemicals. More researches on disinfection by-products formation following ferrate injection as a disinfectant are required. Finally, ferrate(VI) deserves more attention to be used more largely through worldwide wastewater treatment plants.

Subject Areas

Chemical Engineering & Technology

Keywords

Ferrate, Wastewater Treatment, Drinking Water, Disinfection, Green

1. Introduction

During the last decades, iron-founded nanoparticles have reached augmented attention for environmental treatment usages [1]-[8]. Their employment in dealing with polluted water has been particularly classified as an extremely interesting option for eliminating biotic and abiotic pollutants [9]. Among these, ferrates are viewed as a significant category of iron-founded materials that possess singular physical-chemical characteristics with crucial capacity for treating water [10] [11].

Ferrate possesses exceptional redox potential and its application has been found to augment under acidic pH [9]. It is well known that it is an outstanding oxidizing chemical that is useful in performing coagulation actions via constituting ferric hydroxide during the reactional mechanism [12] [13] [14] [15]. The objective of this work is to discuss the application of ferrate-like chemicals as killing agents and routes implied in their functions. Moreover, future trends and primary dares to largely employ ferrate are also exhibited.

This work tries to bring some light into routes involved during killing microbes using ferrate(VI).

2. Employing Ferrate as Disinfectant

In water disinfection field, ferrate has been widely examined and has been found to be an exceptional killing agent [9]. Apart from employing it to demobilize bacteria (*Escherichia coli*, *Salmonella*, *Staphylococcus aureus*, *Bacillus sp.*, *Pseudomonas sp.*, *Enterococcus faecalis*) in water [16] [17] [18] [19], ferrate has also been utilized for attractive implementations like demobilizing fish parasites (*Ichthyophthirius multifiliis*) [20] and eliminating harmful algae (such as *Microcystis aeruginosa*) [19] [21] and viruses [22]. Former publications illustrate that the disinfection potential of ferrate augments greatly at pH degrees lower than 8.0 thanks to protonated ferrate species (Figure 1). This implies that it may demobilize most of the pathogenic microorganisms aforesaid employing doses as low as 1 mg/L [23].

For water disinfection estimates, ferrate was found to be greatly influenced by temperature. Indeed, researchers [22] observed that the ferrate demobilization rate constants for coliphage MS2 augmented by up to fourfold via elevating the temperature from 5°C to 30°C (Figure 2). The impact of temperature on the demobilization operation of MS2 was established to fit with the Arrhenius equation, and a dependence resulting from changing oxidation pathways and/or initial attack sites on the MS2 phage protein or genome components has been proposed [9]. Nevertheless, the influence of temperature is complex since various investigations have mentioned marginal or no impact of temperature for the demobilization of MS2 and *Bacillus subtilis* spores employing ferrate [24].

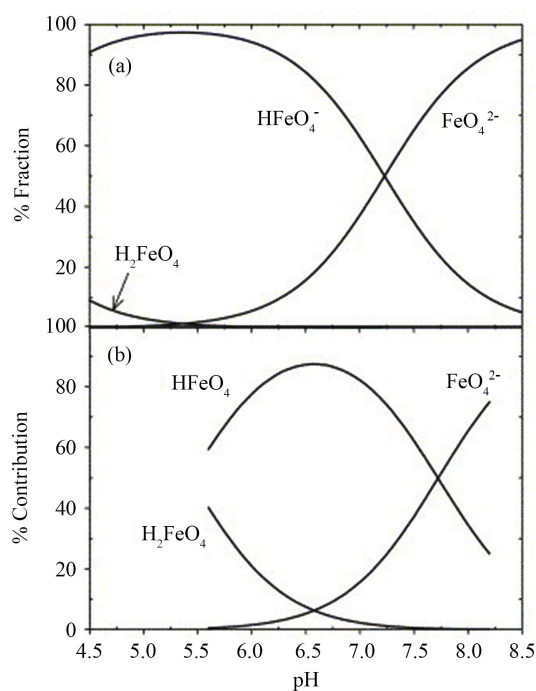


Figure 1. (a) Distribution of Fe(VI) species as a function of pH. (b) Percent contribution from each Fe(VI) species (H_2FeO_4 , HFeO_4^- , and FeO_4^{2-}) to overall *E. coli* inactivation as a function of pH [16].

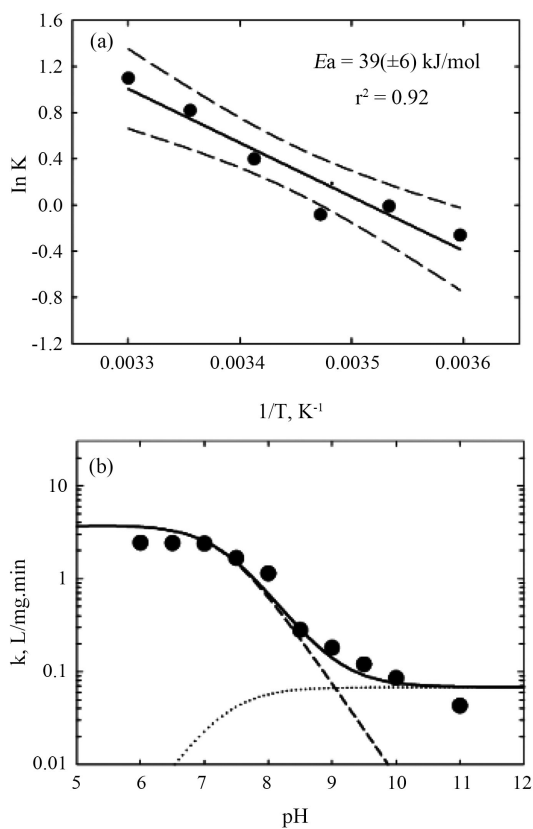
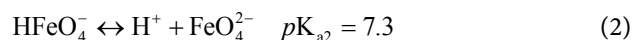
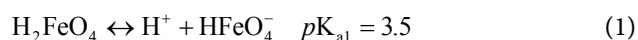


Figure 2. Effect of (a) temperature and (b) pH on the kinetics of MS2 inactivation using ferrate [22].

On the other hand, pH was likewise observed to significantly influence ferrate-founded disinfection techniques. More important demobilization rate constants were determined for the disinfection method of MS2 at a lower pH and the identical was noticed to be correct for *E. coli* demobilization [17]. It has been proposed that the impact of pH on the disinfection efficiency may be linked to the dependence of the reactivity of Fe(VI) on acid-base speciation. For pH comprised between 5 and 8, ferrate is distinguished by two conjugate pairs (Equations (1) and (2)) [9]:



Researchers [25] [26] [27] proposed that protonated ferrate species (Equation (1)) are more powerful oxidants than non-protonated species as protonation decreases the electron-donating potential of the coordinated oxygen ligands encouraging metal center to serve with more elevated oxidative potential [9] [28].

Employing ferrate as a preoxidant for natural organic matter (NOM) [29] [30], reports have mentioned that the generation of THMs was decreased through water disinfection if it is pursued by chlorination [31] [32] [33]. Nevertheless, it has also been mentioned that small injections of ferrate (1 mg/L as Fe) may produce chloral hydrate and halo ketones; while more elevated injections (20 mg/L Fe) importantly diminish the generation of such by-products [34].

In tertiary treated wastewater [35]-[41], big-scale ferrate disinfection has been mentioned to together eliminate total organic carbon (TOC) and pathogens from water (Figure 3) [9] [42]. A configuration was suggested to support coagulation besides the oxidation reaction thanks to its reduction potential, as illustrated in Equations (3) and (4) [42]:

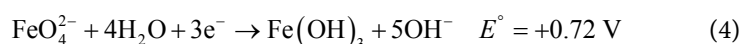


Figure 3(a) illustrates the findings acquired for TOC elimination employing Fe(VI) contrasted with employing hypochlorite as a traditional oxidizer. As displayed, TOC elimination was a fair function of the oxidant concentration and a more important ferrate concentration conducted to a more elevated TOC decrease, perhaps due to the concurrent presence of ferrate-formed coagulation and an oxidative/reductive process [16] [42]. The more elevated TOC elimination was attained employing 14 mg/L of ferrate (like 48% TOC decrease) following one hour of reaction, while less than half of the TOC was eliminated employing the identical reaction circumstances for chlorine. The most important finding was that ferrate was also capable of jointly demobilize pathogens even with reacting with organic matter in the wastewater. Figure 3(b) presents the demobilization of total and fecal coliforms employing ferrate (circles) and hypochlorite (triangles). Figure 3(b) displays that ferrate was apt to demobilize up to 4-log units employing disinfectant injection ($C \times t$) values under 20 (mg/L)·min, while $C \times t$ values bigger than 120 (mg/L)·min of hypochlorite were required to obtain the identical demobilization levels [9].

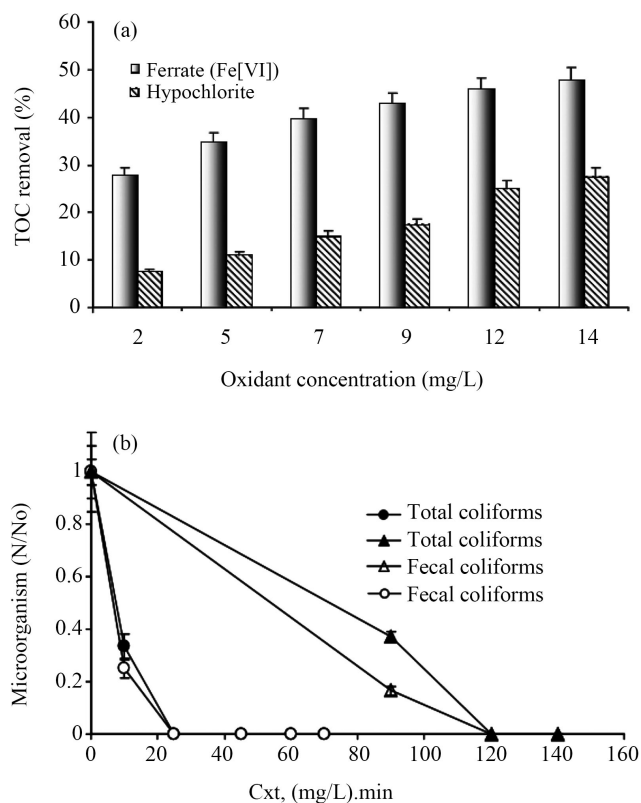


Figure 3. Simultaneous removal of TOC (a) and total and fecal coliforms (b) in wastewater using ferrate (circles) and hypochlorite (triangles) [42].

3. Ferrate(VI) for Emergency Water Treatment (EWT)

Cui *et al.* [43] performed bench-scale trials to estimate ferrate(VI) for treating simulated disaster-polluted water. They proved that ferrate(VI) treatment can together and efficiently demobilize microbial parameters, eliminate metal and metalloid pollutants, decompose dissolved organic matter, and decrease turbidity, whereas insignificantly augmenting total dissolved solids (TDS) to guarantee an allowable $\text{TDS} \leq 1000 \text{ mg/L}$ (the recommended minimum TDS standard for an emergency water supply) [43]. The ferrate(VI) injection possessed a fundamental contribution, as it immediately affected the elimination rates and also influenced the size distributions of iron [44] [45] and metal/metalloid pollutants after treatment. As the ferrate(VI) injection augmented, the portions of soluble and colloidal iron and metal/metalloid pollutants diminished, whilst their particulate fractions augmented, indicating that the pollutants were fixed with ferrate(VI) resultant iron particles. Consequently, an elevated ferrate(VI) injection could quicken the aggregation of fine iron particles, that way helping the elimination of fixed toxic metals and metalloids in resulting solid-liquid separation. Ferrate(VI) reveals a novel chance for emergency water treatment (EWT) to supply secure and sufficient water for disaster-affected populations. The comprehension may be used to ferrate(VI) treatment design for urban water treatment [46] [47] [48] and reuse (Figure 4).

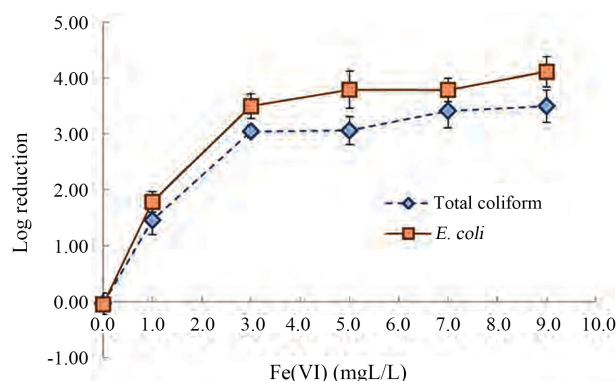


Figure 4. Removals of total coliforms and *E. coli* at various ferrate(VI) injections during the ferrate(VI) treatment of sewage polluted surface water [43].

4. Demobilizing *Bacillus subtilis* Spores via UV/H₂O₂/Ferrate Hybrid Process

Despite the fact that chlorination possesses the capacity to demobilize most of the microbes in water, protozoan parasites such as *C. parvum* oocysts and *Giardia* cysts may fight against it. Consequently, several scientists focused on searching new method for disinfecting water more efficiently [49] [50] [51]. Martin *et al.* [52] assessed the interactive impact of H₂O₂ and ferrate pursued via UV radiation to demobilize *Bacillus subtilis* spores as surrogate microbes. They used response surface methodology (RSM) in optimizing UV/H₂O₂/ferrate and H₂O₂/ferrate techniques. Through employing central composite design (CCD), the impact of three major factors comprising time, hydrogen peroxide, and ferrate concentrations was estimated on process efficiency. Their findings displayed that integrating UV, H₂O₂, and ferrate was the most efficient disinfection technology [53] [54] [55] [56] [57] comparatively with H₂O₂ and ferrate. They established that via UV/H₂O₂/ferrate, around 5.2 log reductions of *B. subtilis* spores were attained at 9299 mg/L of H₂O₂ and 0.4 mg/L of ferrate concentrations following 57 min of residence period, which was the best circumstance (Figure 5). However, H₂O₂/ferrate may demobilize *B. subtilis* spores around 4.7 logs comparatively with the other method. Consequently, these findings proved that UV/H₂O₂/ferrate technique is an encouraging method for both demobilizing spores and disinfecting water.

5. Ferrate as a Coagulant

In its environmental usages [58], coagulation and chemical oxidation have long been known as two main pathways of ferrate(VI) (*i.e.* FeO₄²⁻, an oxyanion holding Fe(VI)). Even if ferrate(VI) oxidation of different pollutants has been widely investigated, fewer works were performed to evaluate the routes and compartments of ferrate(VI)-driven coagulation in water. Lv *et al.* [59] examined coagulation of colloidal kaolin particles using ferrate(VI) in simulated natural water under the circumstances regarding potable water treatment (initial turbidity 25.00 NTU, 0.0 - 9.0 mg/L Fe(VI), pH 7.5, and 0.50 - 10.00 mg/L

DOC). Fe(III) generated from Fe(VI) reduction launched *in situ* coagulation. Minimal efficient iron doses (MEIDs) were detected for Fe(VI) coagulation than for direct Fe(III) coagulation at pH 6.5 and 7.5 (DOC = 2.00 mg/L), at which the colloids were fixed by iron precipitates mainly by sweep coagulation [60] [61]. They employed a citrate-ascorbate iron extraction technique to show that Fe(VI) consequent flocs were constituted of both amorphous and crystalline iron with an amorphous to crystalline Fe ratio of around 2.3:1.0 (pH 7.5, 3.0 mg/L Fe(VI), and 2.00 mg/L DOC). Ferrate(VI) oxidation converted NOM preferentially into more hydrophilic components [62], which possess a lower affinity with colloids and therefore are less adsorbed on colloids to form a less negatively charged surface. Consequently, ferrate(VI) oxidation greatly enhanced the aggregation of colloids via the mitigation of electrostatic repulsion. Nevertheless, NOM at an elevated concentration (8.00 - 10.00 mg/L DOC) may avoid the agglomeration of small iron oxide particles by the generation of a negatively charged NOM coating through adsorption, that way avoiding the development of flocs (Figure 6). If integrated with ferrate(VI) oxidation, ferrate(VI) coagulation gives a more usable treatment solution to remedy numerous water pollution in raw water, like the existence of algal cells and dissolved algal toxins [63] [64] [65] [66] in water throughout a harmful algal bloom [67]. The twin treatment routes authorize an extremely efficient treatment design with an economical physical footprint for supporting a sustainable municipal drinking water supply.

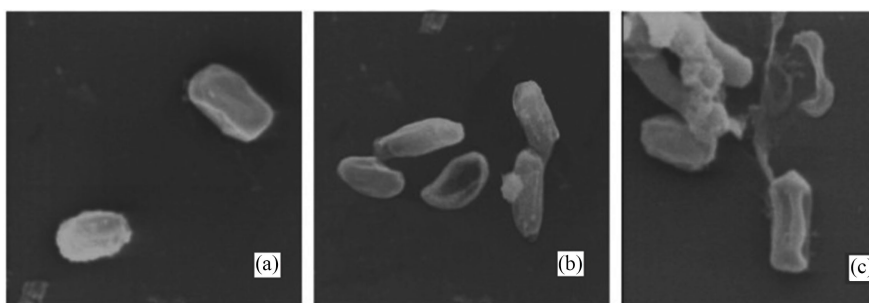


Figure 5. Scanning electron microscope (SEM) images of *B. subtilis* spores under optimal conditions in different processes: (a) Unexposed, (b) H_2O_2 /ferrate and (c) $\text{UV}/\text{H}_2\text{O}_2$ /ferrate [52].

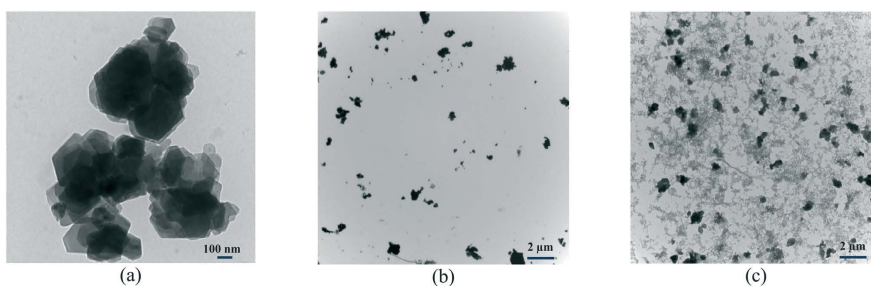


Figure 6. TEM images of particles in simulated natural water: (a) and (b) kaolin particles suspended in water before Fe(VI) coagulation; and (c) kaolin particles incorporated into ferrate(VI)-induced iron precipitates (3.0 mg/L Fe(VI)) (pH 7.5; DOC = 2.00 mg/L; and initial turbidity = 25.00 NTU) [59].

6. Simultaneous Action of Ferrate as a Coagulant and Disinfectant

Barişçi [68] tested the performance of ferrate(IV) prepared electrochemically [69] [70] [71] on both the decomposition of humic acid (HA) [72] [73] and the elimination of *E. coli*. In order to guarantee a total demobilization of *E. coli*, >1.5 mg/L ferrate(VI) concentration has to be injected for only *E. coli* [74] holding samples. For only HA carrying sample, 60% DOC elimination was reached with 25 mg/L ferrate(VI). Ferrate(VI) was observed to be more performant at initial pH of 7. For HA/*E. coli* holding samples, ferrate(VI) concentration must be elevated to 260 mg/L for total demobilization of *E. coli*. Specific UV absorbance (SUVA) values of raw samples illustrated that the samples mainly contain high molecular weight and high hydrophobicity [75] [76] [77] [78]. Following the ferrate(VI) application; whereas almost all hydrophobic fraction was eliminated, a little fraction of the hydrophilic part was also eliminated for only HA holding sample. Nevertheless, for treated HA/*E. coli* carrying sample, the aromatic decrease happened; however, lower and higher molecular weights of NOM remain in the solution in the existence of bacteria. The electro-synthesized ferrate(VI) reduced the trihalomethane formation potential. The remaining total iron concentration was 1.14 mg/L at 1 min of residence period and then remained stable. This signifies that ferrate(VI) was consumed as soon as it was injected and reduced to $\text{Fe}^{3+}/\text{Fe}^{2+}$.

7. Modelling Ferrate(IV) Disinfection

Elnakar and Buchanan [79] examined the action of fast mixing [80] on the rate of potassium ferrate(VI) decomposition and disinfection [81] [82] [83] in bypass wastewaters from extreme wet weather flow events. First-order, second-order, and double exponential models were fit to the potassium ferrate(VI) exhaustion data, and the double exponential model was able to represent the potassium ferrate(VI) decomposition in all circumstances with an elevated coefficient of determination and low mean square error. Further, they examined Chick-Watson and Hom models, and both fit the *E. coli* disinfection [84] [85] findings. The rates of potassium ferrate(VI) exhaustion and disinfection [86] obtained from the models were more elevated employing 500 - 1000 rpm rapid mixing velocities than they were when magnetic stirrer mixing was employed for the same initial injection and wastewater sample. There was no important augmentation in the potassium ferrate(VI) exhaustion or disinfection [87] rates with the elevation of the rapid mixing velocities from 500 to 1000 rpm which discovered that the reactions were kinetically controlled. The coagulation potential of potassium ferrate(VI) improved the sedimentation capacity and participated nearly identical as the chemical disinfection potential to the global *E. coli* elimination. Elnakar and Buchanan [79] concluded that potassium ferrate(VI) may be applied in present plants that mix coagulants to ameliorate primary sedimentation, yet potassium ferrate(VI) may furnish both disinfection [88] and coagulation at lower mixing velocities.

8. Dares and Future Trends

In the face of the considerable performance of ferrate at disinfecting water, scarce publications have given data about its usage or evaluated more kinetic, upscaling, or mechanistic indicators. Additional investigation is necessitated to consistently define many obscure features and deductions for their utilization in treating water [9].

As an illustration, little is recognized concerning the influences of the components of the water employed in ferrate disinfection methods. Even though it has been proposed that ferrate can diminish TOC and bacterial charge simultaneously, there are no publications about the impacts that diverse water components—like ions (such as chloride, carbonate, phosphate) or the existence of nitrogen- or phosphorus-related compounds (such as nitrate and nitrite)—might possess on its performance in disinfection techniques [9].

Utilizing ferrates tested here to demobilize greatly resistant microbes (like helminthes eggs and bacterial or fungal spores) is a different domain in which no accounts have been issued [9]. Employing greatly resistant microbes in a conservative model for water disinfection is very significant to guarantee the treatment of secure potable water even in regions where pathogens like *Giardia* cysts or *Cryptosporidium parvum* oocysts may make a danger to consumers if the adequate precautions for safe potable water are not followed. Moreover, their application for demobilizing else nonpathogenic but very significant microorganisms in water (like harmful algal blooms, phytopathogens, and aquatic invasive species) remains an investigation domain that needs focus.

The capacity of ferrates to function as a coagulant following the oxidation stage [89] gives an extremely motivating post-treatment potential. The ferrates' elevated performance and utilization as a green element for water treatment [90] [91] [92] [93] propose that these techniques remain greatly convenient for usage as pre- or post-treatment in traditional wastewater plants [9].

9. Conclusions

The main points drawn from this work may be given as:

1) Ferrates were tested for their utilization in disinfecting water. Ferrates displayed important implementations for demobilizing frequent water-related microorganisms, as well as viruses and diverse pathogens. Ferrates were also illustrated to be greatly cost-effective for disinfecting water since both may be employed more than once in the treatment procedure and have been mentioned to possess comparatively low synthesis costs. Nevertheless, more study is necessitated for presenting new materials and techniques that can enhance the findings published yet. There is a requirement for more technical ways that can give knowledge beyond the lab scale and let evaluations of the aptness of employing iron-based materials for pilot- or full-scale usages in water disinfection methods [9].

2) Ferrate(VI) allows a novel emergency water treatment design for disas-

ter-affected populations through the repercussions of natural disasters, through the concurrent and efficient elimination of microbial and chemical pollutants. The ferrate(VI) injection is a fundamental parameter as it influences the elimination performances of different pollutants and touches the size distributions of iron and metal/metalloid contaminants following ferrate(VI) degradation [43].

3) Ferrate(VI) coagulates colloids through sweep coagulation under drinking water treatment-related circumstances. Featured from conventional coagulation, coagulation via ferrate(VI) progressively forms iron flocs holding amorphous and crystalline iron oxides in water. Meantime, ferrate(VI) oxidation decomposes NOM to promote particle aggregation. Efficient ferrate(VI) coagulation, integrated with ferrate(VI) oxidation, authorizes a dual-pathway treatment to remedy numerous contaminants for supporting water supply sustainability [59]. Disinfection action occurs simultaneously with oxidizing and coagulating roles. This makes ferrate(IV) as well-known a multi-disciplinary agent [79].

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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