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On-line batch production of ferrate with an chemical method and its potential application for greywater recycling with Al(III) salt

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ABSTRACT

Ferrate(VI) salt is an oxidant and coagulant for water and wastewater treatment. It is considered as a possible alternative method in greywater treatment. However, challenges have existed in putting ferrate(VI) technology into full-scale practice in water and wastewater treatment due to the instability of ferrate solution and high production cost of solid ferrate products. This study demonstrated a new approach of greywater treatment with on-line batch production of Fe(VI) to which Fe(III) salt was oxidized at a weak acidity solution. A series of experiments were conducted to investigate the effect of Fe(VI) on light greywater (total organic carbon (TOC) = 19.5 mg/L) and dark greywater (TOC = 55 mg/L) treatment under different conditions with varying pH and Fe(VI) doses. In addition, the combination use of Fe(VI) and Al(III) salts was proved to be more efficient than using the Fe(VI) salts alone at greywater recycling. The optimum dosage of Fe(VI)/Al(III) salts was 25/25 mg/L for light greywater, 90/60 mg/L for dark greywater, respectively. The TOC values of both light greywater and dark greywater were reduced to less than 3 mg/L with the dosages. The cost for treating greywater was 0.06–0.2 \$/ton at ferrate(VI) dosage of 25–90 mg/L and 0.008–0.024 \$/ton at AlCl₃ dosage of 25–60 mg/L. The full operating cost needs further assessment before the Fe(VI)/Al(III) technology could be implemented in greywater treatment.

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Introduction

Greywater is commonly defined as wastewater generated from kitchen, bath and laundry, excluding wastewater from toilets (Alfiya et al., 2013; Friedler et al., 2005; Organization, 2006). Wastewater from bathroom, showers, tubs and cloth washing machine sources is termed as light greywater (Friedler et al., 2005), accounted for around 50% of the total greywater. Greywater including more contaminated waste from laundry

facilities, dishwashers and, in some instances, kitchen sinks is called as dark greywater (Birks and Hills, 2007). Due to rapid industrialization and development, greywater management is becoming more and more important. There is an increased opportunity for greywater reuse especially in developing countries. Greywater should, therefore, be regarded as a valuable resource and not as a wastewater.

Around 22 treatment systems comprising different treatment processes are discussed in detail for removal efficiency

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of pollutants, effluent concentrations and their compliance with wastewater reuse guidelines and standards which was issued by USEPA (2004). A series of technologies has been used for greywater recycling from simple 2-stage processes (coarse filtration and disinfection) to physical, physicochemical and biological processes (Diaper et al., 2001). The majority of the suggested treatments are biological processes, which can be affected, especially at small scale, by the variability of strength and flow of the greywater and potential shock loading (Hasan et al., 2015; Nolde, 2000; Santala et al., 1998; Surendran and Wheatley, 1998; Teh et al., 2015). These problems can be avoided by using simple physical processes such as cartridge filters or depth filtration beds, which are effective in removing the physical pollution within the greywater. However, they could not significantly remove the organic fraction (Santala et al., 1998).

Using ferrate(VI) as an oxidant and coagulant is considered as a possible alternative method in greywater treatment. It can overcome the above problems. Ferrate(VI) is an emerging water-treatment disinfectant and coagulant, which acts as a strong oxidant to degrade a wide range of compounds present in wastewater and industrial effluents (Jiang and Lloyd, 2002; Jiang, 2007; Lee et al., 2003; Li et al., 2009; Sharma, 2002, 2011, 2013; Tiwari et al., 2005). It can achieve disinfection at relatively low dosages over wide pH ranges (Cho et al., 2006; Jiang et al., 2007; Sharma, 2007) and can also be effective in treating emerging toxins in the aquatic environment (Yuan et al., 2002). Ferrate(VI) is also an efficient coagulant in removing metals, nutrients, radionuclides, and humic acids (Jiang, 2014; Jiang and Lloyd, 2002; Jiang et al., 2001; Jiang and Wang, 2003; Lee et al., 2003, 2009; Sharma, 2010; Song and Ma, 2013). Thus, the multifunctional properties of ferrate(VI) can be utilized in a single dose for recycling and reusing of water and wastewater.

The exploration of the use of ferrate(VI) for water and wastewater treatment has been well addressed recently (Jiang, 2014). However, challenges have existed in putting ferrate(VI) technology into full-scale practice in water and wastewater treatment due to the instability of ferrate solution and high production cost of solid ferrate products (Jiang, 2014; Jiang and Lloyd, 2002). Fe(VI) solutions are generally unstable, and their reduction to Fe(III) species occurs rapidly at room temperature. The instability may be retarded but not stopped at low temperatures or with careful control of solution concentrations (Johnson and Sharma, 1999). Hence, without steps of refrigeration or high purification, the solutions cannot be stored for use in practice. Solid ferrate(VI) salts are stable, but they are costly since they require multiple chemical reagents and long synthesis time. Thus, it is difficult to be used in industry. Therefore, it would be an ideal solution to generate ferrate *in situ* and apply the generated ferrate(VI) directly for wastewater treatment. Generating ferrate(VI) *in situ* not only omit time and money on ferrate(VI) purification, but also avoid electrical energy consumption and save steps on the transportation and storage of ferrate(VI) (Ding et al., 2013; Ghernaout and Naceur, 2011). Unfortunately, there are few studies on generating ferrate(VI) *in situ* and using ferrate(VI) directly for greywater treatment processes. On the other hand, some researchers have shown that the combination of ferrate with Al-based coagulant which has a larger particle size and

more absorption sites could enhance the coagulation efficiency (Jain et al., 2009).

The objectives of this study were: (1) to investigate the efficiency of on-line produced ferrate(VI) as a disinfectant and as an oxidant; (2) to study whether combined use of Fe(VI) and AlCl_3 can give better removal performance than that of Fe(VI) alone; (3) to evaluate economic suitability of using ferrate(VI) comprehensively. Our research focused on the practical application of ferrate(VI) that generated *in situ* for greywater treatment.

1. Materials and experimental methods

1.1. Synthesis of ferrate(VI)

On-line batch production of ferrate(VI) with chemical method was conducted by the wet oxidation method (Jiang and Lloyd, 2002). Solutions were prepared by the addition of volumes of the solid KClO_4 (1.2 mol/L) to 1 mol/L NaOH/2.5 mol/L FeCl_3 . In the process of the reaction continued to add 0.03 mol/L HCl. An excess of oxidant (about 0.2 mol/L solid KClO_4) and the appropriate pH ($\text{pH} = 9 \pm 0.5$) was used to make the Fe(VI) remain stable. Fe(VI) concentration was determined using an established spectroscopy method, and the absorbance of Fe(VI) solution was measured at the characteristic wavelength (510 nm) (Sharma, 2007). Then the absorbance data was converted to Fe(VI) concentration using a computational program based on the calibration with Fe(VI) standard. Fe(VI) was stored in cool place and used within 7 day after produced. Ferrate concentration is expressed as Fe(VI) (mg/L) throughout this article.

1.2. Water samples

Greywater was collected from a purpose built facility which diverted water from the bath, shower and hand basin of 6 flats within a student hall of residence located at Northeast Petroleum University. The tested greywater samples originated from two different sources. Two sources of greywater varied considerable in terms of their organic concentration (Table 1).

1.3. Jar tests

Light greywater (LGW) was chosen as the study object firstly. Then, the obtained optimum technological condition from LGW treatment was applied to dark greywater (DGW) treatment to determine whether the obtained optimal technical parameters is adaptable to the variation of greywater quality.

Lab-scale experiments were conducted by adding 0.5 L greywater to 1 L beakers. Triplicate samples were analyzed under each experimental condition. The pH of the samples was adjusted to predetermined pH with dropwise addition of 1 mol/L HCl or 1 mol/L KOH. The samples were shaken at 200 r/min using a jar tester (MY3000-6B, Meiyu Co., China) for 10 min. The suspension was left undisturbed for 20 min. After the settling, the final pH of the supernatant was measured. The treated water was filtered through 0.45 μm glass fiber filters. The analysis was performed immediately.

Table 1 – Greywater characteristics after treatment with the different systems at optimum conditions.

	DGW			LGW		
	Raw	70 mg/L Fe(VI) 50 mg/L Al(III) pH = 6.5 ± 0.5	90 mg/L Fe(VI) 60 mg/L Al(III) pH = 6.5 ± 0.5	Raw	25 mg/L Fe(VI) 25 mg/L Al(II) pH = 6.5 ± 0.5	30 mg/L Fe(V) 30 mg/L Al(II) pH = 6.5 ± 0.5
COD (mg/L)	385 ± 50	10 ± 2.0	3 ± 1.2	151.5 ± 50	3.0 ± 2.2	1.5 ± 1.2
TOC (mg/L)	55 ± 7	7.6 ± 0.7	2.6 ± 0.2	19.5 ± 5	3.1 ± 0.4	1.6 ± 0.4
Turbidity (NTU)	42 ± 9	3.3 ± 0.3	2.0 ± 0.2	36.5 ± 8	0.5 ± 0.3	1.9 ± 0.3
pH	7.4 ± 0.3	7 ± 0.5	7 ± 0.5	7.15 ± 0.2	7 ± 0.5	7 ± 0.5
TN (mg/L)	15.5 ± 3.0	12.1 ± 1.6	8.1 ± 1.2	8.2 ± 0.5	6.3 ± 0.6	5.7 ± 0.5
PO ₄ ³⁻ -P (mg/L)	1.45 ± 0.1	0.038 ± 0.02	0.026 ± 0.01	0.45 ± 0.2	0.014 ± 0.005	0.01 ± 0.005
NH ₃ -N (mg/L)	1.05 ± 0.3	nd	nd	0.75 ± 0.5	nd	nd
MPN (cells/mL)	7.57 × 10 ⁴ –6.0 × 10 ⁶	nd	nd	4.25 × 10 ⁴ –1.35 × 10 ⁶	nd	nd

COD: chemical oxygen demand; TOC: total organic carbon; NTU: nephelometric turbidity units; TN: total nitrogen; DGW: dark greywater; LGW: light greywater; nd: not detectable; MPN: most probable number. Detection limit of NH₃-N was 0.01 mg/L.

To determine the optimum pH for the total organic carbon (TOC) removal, the experiments were performed as a function of pH from 3 to 11 for LGW, where the Fe(VI) dose was set at 15 and 35 mg/L. Subsequently, the TOC removal was evaluated at various Fe(VI) dose from 5 to 85 mg/L with LGW at the optimum pH. As the Fe(III)–Al(III) oxy/hydroxides precipitates with larger surface area may be able to remove organic compound at a lower dose than the Fe(III) oxy/hydroxides precipitates with smaller surface area, this study carried the following experiment to get better results at greywater recycling. Purification greywater experiments were conducted using Fe(VI) and Al(III) (AlCl₃, industrial grade) salts, the Fe(VI) concentrations in mixed solutions were set at three concentration levels, i.e., 20, 25 and 30 mg/L. The pH was subsequently adjusted to the desired pH by dropwise addition of 1 mol/L HCl or 1 mol/L KOH. After mixing for 1 min, Al(III) chloride salts were added in varied concentrations ranging from 2.5 to 30 mg/L with LGW. The suspension was left undisturbed for 20 min. The final pH was measured and samples were collected for subsequent measurements.

Similar batch experiments of using Fe(VI) were performed to evaluate the TOC removal on DGW. The doses of Fe(VI) were set from 10 to 160 mg/L at pH = 6.5 ± 0.5. Then purification experiments were also conducted for DGW by using Fe(VI) and Al(III) (AlCl₃, industrial grade) salts, the Fe(VI) concentrations in mixed solutions were set at three concentration levels, i.e., 50, 70 and 90 mg/L. The doses of Al(III) chloride salts were varied from 5 to 60 mg/L.

1.4. Analytical methods

Samples of raw and treated greywater were taken, preserved and analyzed in accordance with Standard Methods for the Examination of Water and Wastewater (APHA, 2005), which determined parameters include pH, ammonia (NH₃-N), Total N, PO₄³⁻-P and chemical oxygen demand (COD). Turbidity was measured using a turbidity meter (2100N, Hach, USA). TOC was measured by a TOC analyzer (TOC-5000A, Shimadzu, Japan).

As a biological parameter, viable counts of cultivable heterotrophic bacteria were determined. For obtaining the most probable numbers (MPNs) of bacteria, the standard serial dilution technique was applied. Bacterial growth was detected

in a liquid growth medium (tryptone 6 g/L, yeast extract 3 g/L; pH 7.2 ± 0.2) after 72 hr incubation at 22 ± 1°C. The MPN values were calculated as described in previous publications (Garthright and Blodgett, 2003; Gombos et al., 2013). The removal efficiency (%) of each component was calculated as concentration differences between raw and treated samples.

2. Results and discussion

2.1. Removal of TOC for LGW by Fe(VI) and Fe(VI) + Al(III)

2.1.1. Effect of pH

Effect of pH on TOC removal from LGW is shown in Fig. 1. With the Fe(VI) dose of 15 and 35 mg/L, TOC removal efficiency varied with pH. The removal efficiency of organic matter might be divided into three sections within the pH range of 3.0–11.0. The removal efficiency was very low when the pH was below 5.0. A steep increase in TOC elimination was expressed at pH > 5.0. The optimum pH for TOC removal by Fe(VI) was observed at pH 6.5, at which 77% and 56% was observed by using 35 and 15 mg/L Fe(VI), respectively. As the

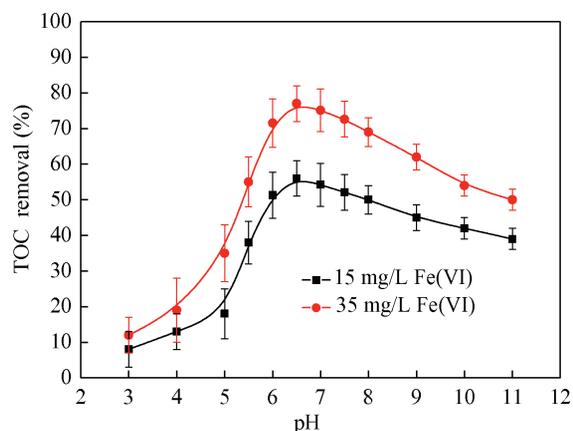


Fig. 1 – Effect of pH on total organic carbon (TOC) removal efficiency for light greywater. Data are presented as means and standard deviations, N = 3.

pH further increased from 6.5 to 11, the TOC removal was continuously reduced.

The ferrate has a higher oxidation potential at low pH than that in the alkaline condition. When $\text{pH} < 5$, Fe(VI) was rapidly decomposed, which was unable to fully contact with pollutants, the oxidation of pollutants by Fe(VI) was not complete. In addition, Fe(III) oxy/hydroxides precipitates could not form at this acidic solution. When pH ranged from 5 to 6.5, Fe(VI) possessed strong oxidation capacity, since Fe(VI) showed high protonation degree with strong oxidation capacity. At pH 9.0–10.0, Fe(VI) was relatively stable, especially at $\text{pH} = 9.3$ (Li et al., 2008). However, the oxidizing capacity of Fe(VI) was weak and the coagulation of Fe(III) oxy/hydroxides played a main role. Therefore, the optimal water pH in this study was determined by the comprehensive effect of stability and oxidizing capacity of Fe(VI). In the following study, the pH of Fe(VI) solution was adjusted to 6.5 ± 0.5 at which Fe(VI) showed best performance for TOC removal.

2.1.2. Performance of organic matter removal by Fe(VI)

Fig. 2 presents TOC removal efficiency as a function of varied Fe(VI) concentrations for LGW sample. In the case of LGW samples, the TOC removal efficiency varied from 30.1% to 94.2% as a function of Fe(VI) concentrations (5–85 Fe(VI) mg/L). The TOC removal efficiency increased sharply with increasing Fe(VI) dose from 5 to 50 mg/L, then slowly increased with increasing Fe(VI) dose. The optimum dosage of Fe(VI) reducing TOC to less than 3 mg/L is 50 mg/L when $\text{pH} = 6.5$. The outstanding performance in reduction of TOC/COD with Fe(VI) is consistent with previous studies, where, a range of organic contaminants including alcohol, carboxylic compounds, amino-acids, phenol, organic nitrogen compounds, aliphatic sulfur, nitrosamine compounds, recalcitrant organics, thiourea, chlorine oxyanions, and hydrazine compounds can be removed efficiently by using Fe(VI) (Jiang et al., 2006). These results can be attributed to the high oxidation efficiency of Fe(VI) in reducing TOC. In addition, the percentage oxidation of these compounds strongly depends on the dose of ferrate(VI); and an excess of ferrate dose was proved to be more effective in reducing organic concentration (Jiang and Lloyd, 2002).

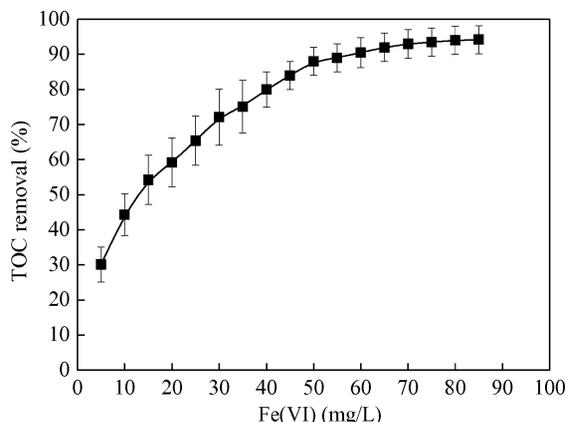


Fig. 2 – Removal efficiency of TOC for light greywater as a function of the Fe(VI) addition. Data are presented as means and standard deviations, $N = 3$.

According to the literature, a secondary effluent having a TOC of 12 mg/L was treated with 20 mg/L Fe(VI) and 35% of the TOC was removed. The results didn't include the efficiency of filtration process (Jiang and Lloyd, 2002). In the present study, after filtration, the TOC removal efficiency reached 59% for LGW (TOC = 19.5 mg/L) treated with 20 mg/L Fe(VI). It indicated that the TOC removal efficiency is much greater when the ferrate coagulation capacity was taken into account.

2.1.3. Performance of organic matter removal by Fe(VI) + Al(III)

The organic removal efficiency by Fe(VI)/Al(III) salts is shown in Fig. 3. The amounts of Fe(VI) required by the Fe(VI)/Al(III) were much lower than that of Fe(VI) alone to achieve the same effects on reducing the polluted matter in greywater. Fig. 3 presents the optimum dosages of Fe(VI) and Al(III) as 25 mg/L and 25 mg/L, respectively. To achieve the same level of organic removal, approximately half of Fe(VI) dosage was sufficient in Fe(VI)/Al(III) system compared to that of Fe(VI) alone according to our measured data. In addition, in combination systems, at all three Fe(VI) concentrations used, Al(III)/Fe(VI) was found more efficient than Fe(VI) alone in decreasing TOC (Fig. 3). By comparing Figs. 2 and 3, organic compound concentrations of solution decreased sharply with increasing Al concentrations in the Fe(VI)/Al(III) system. This result could be attributed to the smaller particle size of the Fe(III)–Al(III) oxy/hydroxide precipitates leading to higher surface area and more adsorption sites available for organic compound than that of Fe(III) oxy/hydroxide precipitates. This observation was confirmed by the determination of the surface areas of the precipitates (Jain et al., 2009).

Fig. 4 compares the performance of Fe(VI) at a dose of 25 mg/L with that of Fe(VI)/Al(III) at doses of 25 mg/L/25 mg/L for the LGW treatment involved in the study. It can be seen that Fe(VI) treatment at a dose of 25 mg/L allows obtaining a lower removal efficiency than that in the Fe(VI)/Al(III) treatment for all of the parameters. Concentration of total N (8.2 mg/L) and $\text{PO}_4^{3-}\text{-P}$ (0.45 mg/L) was reduced to 7.4 mg/L and 0.05 mg/L (removal 9.6% and 88.9%) by using Fe(VI), respectively. Whereas, the corresponding total N and $\text{PO}_4^{3-}\text{-P}$ removal was 23.1% and 96.9% by using Fe(VI)/Al(III), respectively. The

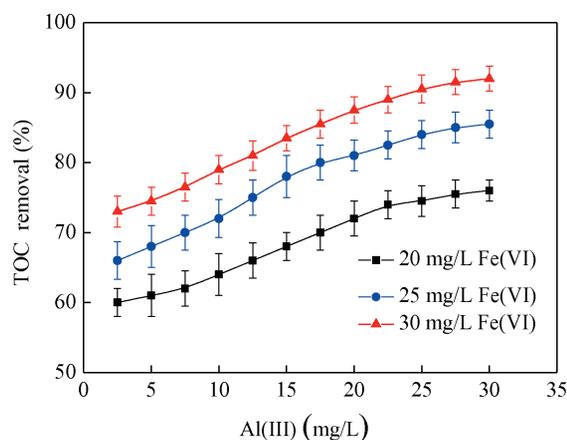


Fig. 3 – Removal efficiency of TOC as a function of the Fe(VI)/Al(III) addition for light greywater. Data are presented as means and standard deviations, $N = 3$.

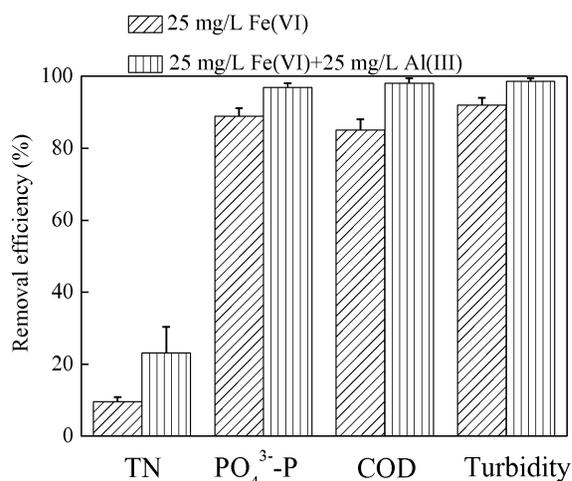


Fig. 4 – Light greywater treatment with Fe(VI) addition vs. Fe(VI)/Al(VI) addition. Data are presented as means and standard deviations, N = 3.

COD value (151.5 mg/L) was reduced to 22.7 mg/L (removal 85%) by using Fe(VI). Whereas, by using Fe(VI)/Al(III), the corresponding COD removal was 98%. Turbidity of LGW was reduced from 36.5 to 2.9 and 0.5 nephelometric turbidity unit (NTU) (removal 92% and 98.6%) by using Fe(VI) and Fe(VI)/Al(III), respectively. In total, Fe(VI)/Al(III) was proved to be more effective than Fe(VI) alone. Therefore, further tests were focused on Fe(VI)/Al(III), as described in the following paragraphs.

2.2. Removal of TOC for DGW by Fe(VI) and Fe(VI) + Al(III)

The Al(III) and Fe(VI) systems were proved to be efficient for treating LGW for reuse. However, it is worthwhile to note that the LGW strength was very low and it is perhaps not too surprising that the systems were capable of removing sufficient materials to meet the standard. In order to make

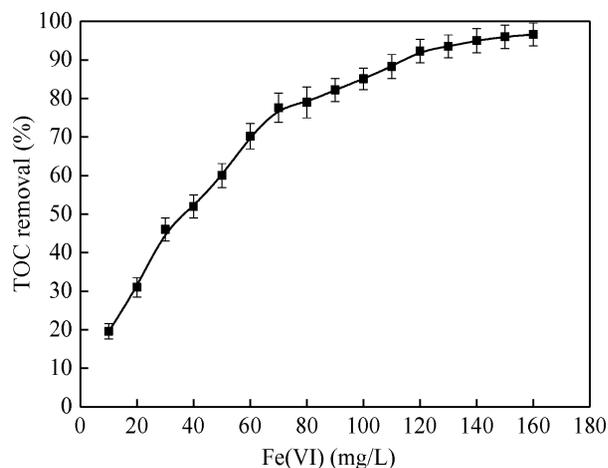


Fig. 5 – Removal efficiency of TOC for dark greywater as a function of the Fe(VI) addition. Data are presented as means and standard deviations, N = 3.

Fe(VI) adapt the change of DGW water quality, the optimum technological condition should be determined again as above. Fig. 5 presents TOC removal efficiency as a function of higher Fe(VI) concentration for DGW samples. The TOC removal efficiency varied between 19.6% and 96.6% with varying Fe(VI) concentrations (10–160 mg/L). The optimum dosage of Fe(VI) reducing TOC of DGW to less than 3 mg/L is 140 mg/L, which is much higher than that of LGW.

In combination systems, TOC of the DGW greywater was reduced from 55 to 7.6 mg/L and 2.6 mg/L (removal 86.2% and 95.2%) by using 70 mg/L Fe(VI)/50 mg/L Al(III) and 90 mg/L Fe(VI)/60 mg/L Al(III), respectively (Fig. 6). Table 1 shows the removal efficiency of TOC, COD, turbidity, total nitrogen (TN), NH₃-N, PO₄³⁻⁻-P, and the MPN values of heterotrophic bacteria from greywater by Fe(VI) and Al(III). In the case of DGW samples, the turbidity was reduced from 42 NTU to 3.3 and 2.0 NTU (removal 92.1% and 95.2%) by using 70 mg/L Fe(VI)/50 mg/L Al(III) and 90 mg/L Fe(VI)/60 mg/L Al(III), respectively. COD was reduced from 385 to 10 and 3 mg/L (removal 97.4% and 99.2%) by using 70 mg/L Fe(VI)/50 mg/L Al(III) and 90 mg/L Fe(VI)/60 mg/L Al(III), respectively. Concentration of ammonia N (1.05 mg/L), total N (15.5 mg/L) and PO₄³⁻⁻-P (1.45 mg/L) was reduced to <0.01 mg/L, 12.1 mg/L and 0.038 mg/L (removal >99.0%, 21.9% and 97.4%), respectively, by using 70 mg/L Fe(VI)/50 mg/L Al(III). Whereas, by using 90 mg/L Fe(VI)/60 mg/L Al(III), the corresponding ammonia N, total N and PO₄³⁻⁻-P removal efficiency was >99.0%, 47.7% and 98%, respectively. MPN values of heterotrophic bacteria were reduced from 7.57 × 10⁴–6.0 × 10⁶ cells/mL to nd. by using 70 mg/L Fe(VI)/50 mg/L Al(III) and similar results were also observed by using 90 mg/L Fe(VI)/60 mg/L Al(III). For both LGW and DGW greywater, an excess of ferrate dose was proved to be more effective in reducing organic concentration.

2.3. The cost of greywater treatment by using Fe(VI) + Al(III) and application prospect

The laboratory studies demonstrated that the effluent of the system could meet most of the standards for wastewater reuse in terms of pH, turbidity, COD, TOC, TN, PO₄³⁻⁻-P and

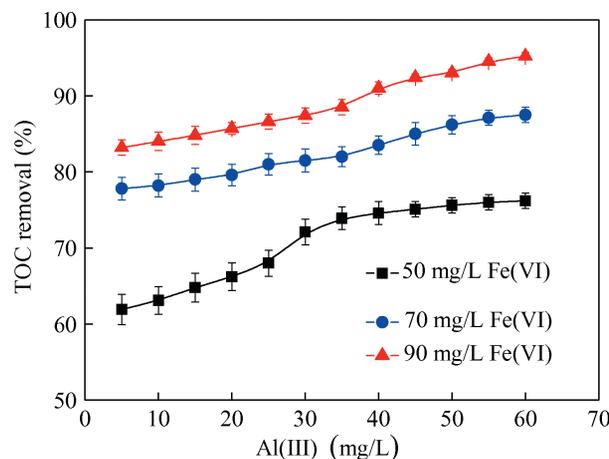


Fig. 6 – Removal efficiency of TOC for dark greywater as a function of the Fe(VI)/Al(VI). Data are presented as means and standard deviations, N = 3.

ammonia N. The addition of Fe(VI) and Al(III) can effectively purify greywater. Summarizing the results presented above, 25–90 mg/L Fe(VI) and 25–60 mg/L Al(III) would be enough to satisfy some of the reuse standards for greywater recycling. Thus, Fe(VI) dosage of 25–90 mg/L was considered potentially feasible for further engineering application, and subjected to the following cost analysis for applying ferrate(VI).

Based on the Chinese retail prices of AlCl_3 , KClO_4 , $\text{Fe}(\text{NO}_3)_3$ as 2100–2600, 17,000–18,000, 6500–6900 RMB per ton, respectively. The cost of AlCl_3 is 0.008–0.024 \$ for treating per ton of water. The cost of self-made Fe(VI) solution (concentration of $\text{Fe}(\text{VI}) \geq 2 \text{ mol/L}$) is about 1000 \$/ton (including reactants and their preparation, conditioning of the pellets, workforce and energy consumption). When the Fe(VI) dosing ranged from 25 to 90 mg/L, the cost for implementing Fe(VI) in greywater treatment process was calculated to be 0.06–0.224 \$/ton of water. Therefore, it would be 0.068–0.224 \$ for treating per ton of water by using Fe(VI)/Al(III), which is economically acceptable. In addition, compared with other methods, Fe(VI) has advantages in low dosage, no toxic intermediates generation (Ma and Liu, 2002), applicable to a wide range of water quality, and enhancing the follow-up processes.

3. Conclusion

Greywater reuse is a potential method to reduce potable water consumption in buildings and, therefore, to reduce wastewater discharged to public sewage systems and treatment plants. This study showed that TOC were both less than 3 mg/L for LGW and DGW after treatment by Fe(VI)/Al(III) salt at $\text{pH} = 6.5 \pm 0.5$, respectively, which meet USEPA criteria for wastewater reuse. Other water quality indicators such as PO_4^{3-} -P, turbidity, TN, ammonia N, heterotrophic bacteria treated by Fe(VI)/Al(III) salt also satisfied the Guidelines for Water Reuse Report of USEPA. On the basis of our result, it can be stated that Fe(VI) and Al(III) process is a promising solution in greywater treatment for disinfection and organic removal, although full-scale studies are still needed. This method is more flexible, which can adapt the change of water quality through regulating the dose of ferrate(VI) and AlCl_3 . Moreover, on-line batch producing ferrate(VI) with chemical method reduces the cost of using ferrate(VI). It is possible to apply the generated ferrate(VI) directly for greywater treatment. Thus, this method was considered potentially feasible for further engineering application. The cost of using Fe(VI) (at the dosage of 25–90 mg/L) and Al(III) (at the dosage of 25–60 mg/L) in the greywater treatment process was estimated to be 0.068–0.224 \$ per ton water being treated. As far as application of Fe(VI) is concerned, this study put forward a better way not only from cost of producing Fe(VI) but also from efficiency of water treatment.

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REFERENCES

- Alfiya, Y., Gross, A., Sklarz, M., Friedler, E., 2013. Reliability of on-site greywater treatment systems in Mediterranean and arid environments — a case study. *Water Sci. Technol.* 67, 1389–1395.
- APHA, 2005. *Standard Methods for the Examination of Water and Wastewater*. 21st ed. American Public Health Association, Washington, DC.
- Birks, R., Hills, S., 2007. Characterisation of indicator organisms and pathogens in domestic greywater for recycling. *Environ. Monit. Assess.* 129, 61–69.
- Cho, M., Lee, Y., Choi, W., Chung, H., Yoon, J., 2006. Study on Fe(VI) species as a disinfectant: quantitative evaluation and modeling for inactivating *Escherichia coli*. *Water Res.* 40, 3580–3586.
- Diaper, C., Jefferson, B., Parsons, S.A., Judd, S.J., 2001. Water-recycling technologies in the UK. *J. Chart. Inst. Water Environ. Manag.* 15, 282–286.
- Ding, L., Li, X.Z., Lee, S.C., 2013. Kinetics of CH3S-reaction with in situ ferrate(VI) in aqueous alkaline solution. *Chemosphere* 92, 1301–1306.
- Friedler, E., Kovalio, R., Galil, N.I., 2005. On-site greywater treatment and reuse in multi-storey buildings. *Water Sci. Technol.* 51, 187–194.
- Garthright, W.E., Blodgett, R.J., 2003. FDA's preferred MPN methods for standard, large or unusual tests, with a spreadsheet. *Food Microbiol.* 20 (4), 439–445.
- Ghernaout, D., Naceur, M.W., 2011. Ferrate(VI): in situ generation and water treatment — a review. *Desalin. Water Treat.* 30, 319–332.
- Gombos, E., Barkacs, K., Felföldi, T., Vertes, C., Mako, M., Palko, G., Zaray, G., 2013. Removal of organic matters in wastewater treatment by ferrate (VI)-technology. *Microchem. J.* 107, 115–120.
- Hasan, M.M., Shafiquzzaman, M., Nakajima, J., Ahmed, A.K.T., Azam, M.S., 2015. Application of a low cost ceramic filter to a membrane bioreactor for greywater treatment. *Water Environ. Res.* 87, 233–241.
- Jain, A., Sharma, V.K., Mbuya, O.S., 2009. Removal of arsenite by Fe(VI), Fe(VI)/Fe(III), and Fe(VI)/Al(III) salts: effect of pH and anions. *J. Hazard. Mater.* 169, 339–344.
- Jiang, J.Q., 2007. Research progress in the use of ferrate(VI) for the environmental remediation. *J. Hazard. Mater.* 146, 617–623.
- Jiang, J.Q., 2014. Advances in the development and application of ferrate(VI) for water and wastewater treatment. *J. Chem. Technol. Biotechnol.* 89, 165–177.
- Jiang, J.Q., Lloyd, B., 2002. Progress in the development and use of ferrate(VI) salt as an oxidant and coagulant for water and wastewater treatment. *Water Res.* 36, 1397–1408.
- Jiang, J.Q., Wang, S., 2003. Enhanced coagulation with potassium ferrate(VI) for removing humic substances. *Environ. Eng. Sci.* 20, 627–633.
- Jiang, J.Q., Lloyd, B., Grigore, L., 2001. Preparation and evaluation of potassium ferrate as an oxidant and coagulant for potable water treatment. *Environ. Eng. Sci.* 18, 323–328.
- Jiang, J.Q., Wang, S., Panagouloupoulos, A., 2006. The exploration of potassium ferrate(VI) as a disinfectant/coagulant in water and wastewater treatment. *Chemosphere* 63, 212–219.
- Jiang, J.Q., Wang, S., Panagouloupoulos, A., 2007. The role of potassium ferrate(VI) in the inactivation of *Escherichia coli* and in the reduction of COD for water remediation. *Desalination* 210, 266–273.
- Johnson, M.D., Sharma, K.D., 1999. Kinetics and mechanism of the reduction of ferrate by one-electron reductants. *Inorg. Chim. Acta* 293, 229–233.
- Lee, Y., Um, I.H., Yoon, J., 2003. Arsenic(III) oxidation by iron(VI) (ferrate) and subsequent removal of arsenic(V) by iron(III) coagulation. *Environ. Sci. Technol.* 37, 5750–5756.

- Lee, Y., Zimmermann, S.G., Kieu, A.T., von Gunten, U., 2009. Ferrate (Fe(VI)) application for municipal wastewater treatment: a novel process for simultaneous micropollutant oxidation and phosphate removal. *Environ. Sci. Technol.* 43, 3831–3838.
- Li, C., Li, X.Z., Graham, N., Gao, N.Y., 2008. The aqueous degradation of bisphenol A and steroid estrogens by ferrate. *Water Res.* 42, 109–120.
- Li, G., Wang, N., Liu, B., Zhang, X., 2009. Decolorization of azo dye Orange II by ferrate(VI)–hypochlorite liquid mixture, potassium ferrate(VI) and potassium permanganate. *Desalination* 249, 936–941.
- Ma, J., Liu, W., 2002. Effectiveness of ferrate (VI) preoxidation in enhancing the coagulation of surface waters. *Water Res.* 36, 4959–4962.
- Nolde, E., 2000. Greywater reuse systems for toilet flushing in multi-storey buildings — over ten year experience in Berlin. *Urban Water* 1, 275–284.
- Organization, WHO, 2006. WHO guidelines for the safe use of wastewater, excreta and greywater. http://www.who.int/water_sanitation_health/wastewater/gsuweg4/en/.
- Santala, E., Uotila, J., Zaitsev, G., Alasiurua, R., Tikka, R., Tengvall, J., 1998. Microbiological Greywater Treatment and Recycling in an Apartment Building. Proc. of the 2nd International Advanced Wastewater Treatment, Recycling and Reuse, 14–16th September, Milan, Italy.
- Sharma, V.K., 2002. Potassium ferrate(VI): an environmentally friendly oxidant. *Adv. Environ. Res.* 6, 143–156.
- Sharma, V.K., 2007. Disinfection performance of Fe(VI) in water and wastewater: a review. *Water Sci. Technol.* 55, 225–232.
- Sharma, V.K., 2010. Oxidation of nitrogen-containing pollutants by novel ferrate(VI) technology: a review. *J. Environ. Sci. Health Part A Tox. Hazard. Subst. Environ. Eng.* 45, 645–667.
- Sharma, V.K., 2011. Oxidation of inorganic contaminants by ferrates (VI, V, and IV)-kinetics and mechanisms: a review. *J. Environ. Manag.* 92, 1051–1073.
- Sharma, V.K., 2013. Ferrate(VI) and ferrate(V) oxidation of organic compounds: kinetics and mechanism. *Coord. Chem. Rev.* 257, 495–510.
- Song, Y., Ma, J., 2013. Development of Ferrate(VI) Salt as an Oxidant and Coagulant for Water and Wastewater Treatment. In: Xu, N., Tian, L., Dai, F. (Eds.), *Sustainable Cities Development and Environment Protection Vols. Pts 1-3*, pp. 658–661.
- Surendran, S., Wheatley, A.D., 1998. Grey-water reclamation for non-potable re-use. *Water Environ. J.* 12, 406–413.
- Teh, X.Y., Poh, P.E., Gouwanda, D., Chong, M.N., 2015. Decentralized light greywater treatment using aerobic digestion and hydrogen peroxide disinfection for non-potable reuse. *J. Clean. Prod.* 99, 305–311.
- Tiwari, D., Yang, J.-K., Lee, S.-M., 2005. Applications of ferrate(vi) in the treatment of wastewaters. *Environ. Eng. Res.* 10, 1–13.
- USEPA, 2004. Guidelines for water reuse report. <http://water.epa.gov/aboutow/owm/upload/Water-Reuse-Guidelines-625r04108.pdf>.
- Yuan, B.L., Qu, J.H., Fu, M.L., 2002. Removal of cyanobacterial microcystin-LR by ferrate oxidation-coagulation. *Toxicol.* 40, 1129–1134.