



## Characteristic of a novel composite inorganic polymer coagulant–PFAC prepared by hydrochloric pickle liquor

Wei Lan<sup>a,b</sup>, Huiqin Qiu<sup>a,\*</sup>, Jie Zhang<sup>a</sup>, Yanjing Yu<sup>a</sup>, Kailiang Yang<sup>a</sup>, Zhongzhe Liu<sup>a</sup>, Guoji Ding<sup>a</sup>

<sup>a</sup> School of Environmental and Chemical Engineering, Shanghai University, Shanghai 200072, PR China

<sup>b</sup> Department of Water Conservancy Management, Shanghai Water Authority, Shanghai 200003, PR China

### ARTICLE INFO

#### Article history:

Received 29 September 2007

Received in revised form 5 May 2008

Accepted 6 May 2008

Available online 15 May 2008

#### Keywords:

Hydrochloric pickle liquor

Polyferric aluminum chloride (PFAC)

Coagulation performance

Stability

### ABSTRACT

A composite inorganic polymer coagulant, polyferric aluminum chloride (PFAC) was prepared by using hydrochloric pickle liquor and calcium aluminate as main materials. The optimum conditions for preparing PFAC with the hydrochloric pickle liquor and the calcium aluminate were studied. The coagulation performance of PFAC was investigated by studying the turbidity, COD, total phosphate (TP) and NH<sub>3</sub>-N removal efficiency in municipal sewage treatment. Results indicated that the effective composition, basicity (simplified as  $B$ ,  $B = [\text{OH}]/(3[\text{Fe}_T + \text{Al}_T]) \times 100\%$ ), coagulation performance and stability of PFAC were affected by calcium aluminate dosage, reaction time, reaction temperature and stabilizing agents. The COD and turbidity removal efficiency of PFAC was better than that of PFS and FeCl<sub>3</sub>, and the TP and NH<sub>3</sub>-N removal efficiency of PFAC was much better than that of PFS, FeCl<sub>3</sub> and PAC. PFAC not only possessed a good coagulation performance, but also had good stability when stored.

© 2008 Elsevier B.V. All rights reserved.

### 1. Introduction

Pickling is a method used in sheet mills or metal fabricating plants to remove oxides and scale from the surface or parts of the metal by passing them through an acid bath. During the pickling, the scale oxides dissolve and form iron(II) sulphate/chloride, and repeated pickling in the same liquor leads to the generation of pickle liquor containing residual acids and salts. Due to the corrosive nature and the presence of high amount of dissolved iron(II), the pickle liquor is specifically listed as hazardous waste in many countries [1]. Currently, several processes are available for the treatment of hydrochloric pickle liquor. The traditional procedure of neutralization of acids in pickle liquor generates large volume of wastes and causes high acid consumption in steel pickling. New methods used for recycling hydrochloric acids and salts include ion exchange [2], ion exchange combined with membrane electrolysis [3], membrane distillation [4,5], electrodialysis [6,7] or combination of these methods [8]. However, none of these methods makes iron and/or hydrochloric acids recovery economically attractive.

Coagulation is a common process in water treatment for destabilizing dissolved and colloid impurities and producing large floc aggregates which can be removed from the water in subsequent clarification/filtration processes. Fe(III) salts and Al(III) salts are the most commonly used coagulants in water treatment. Poly-

aluminum ferric chloride (PAFC) has been developed by partially neutralizing AlCl<sub>3</sub> and FeCl<sub>3</sub>, and it has been widely used [9,10]. However, methods for preparing stable and low-cost PAFC have not yet been well characterized. Recently, PAFC has been synthesized by using industrial wastes (such as blast furnace dust, gangue and bauxite) as materials and using reagent hydrochloric acid or industrial hydrochloric acid as solvents, which can reduce the cost for preparing coagulant to some extent [11–13]. If we replace the reagent hydrochloric acid or industrial hydrochloric acid with hydrochloric pickle liquor, it may not only further reduce the cost for preparing coagulant, but also provide a feasible way to recycle the salts and residual acids in the hydrochloric pickle liquor.

The purpose of this work is to examine the preparation route from hydrochloric pickle liquor and calcium aluminate to polyferric aluminum chloride (PFAC). To prepare PFAC, the dissolved ferrous iron(II) in spent pickle liquor was oxidized by sodium chlorate, and the residual acid was neutralized with calcium aluminate. PFAC was then synthesized by hydrolysis and copolymerization of the resultant iron(III) and Al(III) under basic condition. The composition of PFAC was analyzed, and the coagulant performance of PFAC in the municipal sewage treatment was investigated as well.

### 2. Materials and methods

#### 2.1. Preparation of PFAC

PFAC was prepared in the laboratory by using hydrochloric pickle liquor (FeCl<sub>2</sub> content = 6.6%, HCl content = 9.9%, specific

\* Corresponding author. Tel.: +86 21 5633 3424; fax: +86 21 5633 3052.  
E-mail address: [huiqin.qiu@sina.com](mailto:huiqin.qiu@sina.com) (H. Qiu).

**Table 1**  
Composition of calcium aluminate (%)

Al <sub>2</sub> O <sub>3</sub>	57.0
CaO	27.8
Fe <sub>2</sub> O <sub>3</sub>	4.2
SiO <sub>2</sub>	9.8
TiO <sub>2</sub>	0.5
MgO	0.1
Other	0.6

gravity = 1.157, Baosteel Group Shanghai Ergang Co., China) and calcium aluminate (Henan Northern Naihuo Co., China) as materials. The composition of calcium aluminate is shown in Table 1. An aliquot of 300 mL of pickle liquor was added to a three-necked flask and placed in an electric heat constant temperature water bath (Shenzhen Guohua electronic Co., China) at atmospheric pressures. 7.26 g of sodium chlorate (chemically pure) were gradually introduced into the three-necked flask to oxidate the Fe(II) in the pickle liquor to Fe(III). Temperatures were varied from 50 to 70 °C for optimization purposes. After oxidation, varied amounts of calcium aluminate were added to the mixture according to the weight/volume ratio of calcium aluminate/pickle liquor (W/V, 1:20 to 7:60) in order to optimize for the effect of dosage. The mixture was stirred to dissolve the aluminum oxide and to induce Al(III) to copolymerize with Fe(III). Finally, the mixture was filtrated and stabilized using different stabilizing agents (H<sub>3</sub>PO<sub>4</sub>, Na<sub>3</sub>PO<sub>4</sub>, NaH<sub>2</sub>O<sub>4</sub> and Na<sub>2</sub>SO<sub>4</sub>).

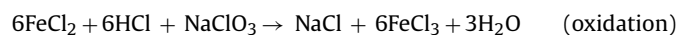
## 2.2. Jar test procedure

Coagulation tests were performed by using synthetic water or actual municipal sewage. All coagulation tests were conducted in 1.0 L beakers using a six paddle gang stirrer (Shanghai Changsi Co., China). Turbidity was the key factor in testing the performance of coagulants in many Refs. [14–16], so we just considered the turbidity removal efficiency of PFAC in the treatment of synthetic water to get the optimum preparation conditions. The synthetic water was prepared by adding 2.0 g kaolin (Chemical pure) to 10.0 L distilled water, the pH value and turbidity of the synthetic water were 7.0 and 120 NTU, respectively. The actual municipal sewage was taken from the primary sedimentation tank of Quyang sewage treatment plant (Shanghai, China). The original pH value, turbidity, COD, TP and NH<sub>3</sub>-N of the municipal sewage were 7.7, 125 NTU, 291 mg/L, 6.0 mg/L and 37.2 mg/L respectively. An aliquot of 700 mL of test water was placed in a beaker and stirred rapidly at 200 rpm for 2 min after adding the coagulant, followed by slow stir at 60 rpm for 10 min and sedimentation for 20 min. After sedimentation, 2.0 cm below the surface of test water, a supernatant sample was taken for turbidity measurement by using 2100P portable turbidimeter (HACH, American), for COD, TP and NH<sub>3</sub>-N measurement by using HI 83214 multi-parameter photometer (HANNA, Italy) [17].

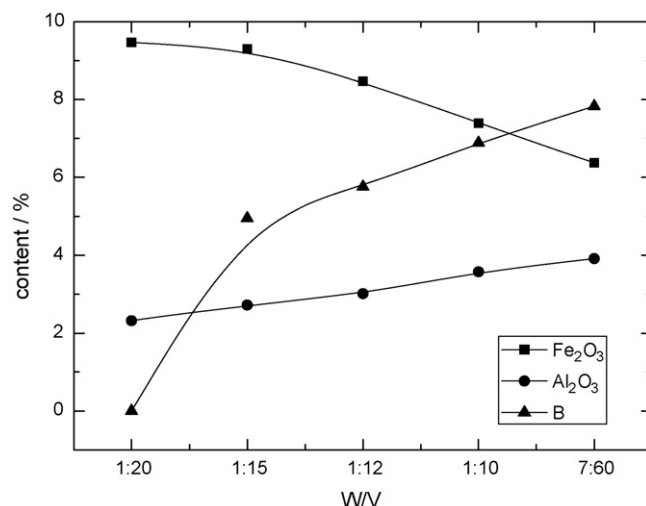
## 3. Results and discussion

### 3.1. Preparation of PFAC

The preparation of PFAC began with the oxidation of Fe(II) in pickle liquor containing hydrochloric acid and ferrous chloride.

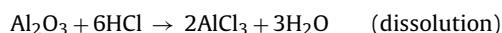


When the calcium aluminate was added to the hydrochloric pickle liquor, Al<sub>2</sub>O<sub>3</sub> in calcium aluminate reacted with the residual hydrochloric acid in hydrochloric pickle liquor, and the dissolution

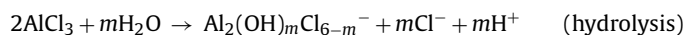
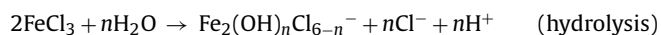


**Fig. 1.** Effect of W/V on effective composition and B value of PFAC.

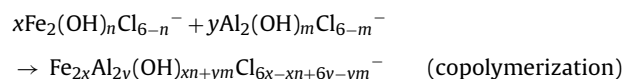
would occur.



With the increase of calcium aluminate, the residual hydrochloric acid was gradually neutralized and the basicity of the mixture gradually increased, and the hydrolyzation of Fe(III) and Al(III) would occur.



With the polymerization of hydroxyl, the copolymerization of the hydrolysis complexes formed by the Fe(III) and the Al(III) would occur.



As shown in Table 1, calcium aluminate was mainly made up of metal oxides, exhibiting alkaline character. It could neutralize the hydrochloric acid in the hydrochloric pickle liquor and then adjust the basicity of the solution, the dosage of calcium aluminate not only affected the Al<sub>2</sub>O<sub>3</sub> content of PFAC, but also influenced the B content of PFAC. Reaction time affected the Al<sub>2</sub>O<sub>3</sub> dissolution rate of calcium aluminate and the reaction extent between calcium aluminate and pickle liquor. Reaction temperature affected the Al<sub>2</sub>O<sub>3</sub> dissolution rate of calcium aluminate and the reaction speed. Therefore, dosage of calcium aluminate, reaction time and reaction temperature were considered as the three main factors in the preparation of PFAC.

#### 3.1.1. W/V

In order to investigate the effect of the dosage of calcium aluminate, W/V was identified as the weight/volume ratio of calcium aluminate/pickle liquor, and the unit of W/V was gram/milliliter. To study the effect of W/V, the volume of pickle liquor was kept constant at 300 mL, the reaction temperature was kept constant at 60 °C and the reaction time was kept constant at 1.5 h, while varying the dosage of calcium aluminate. Results are shown in Figs. 1 and 2.

Fig. 1 shows that with the increase of W/V, the Al<sub>2</sub>O<sub>3</sub> content and the B of PFAC increased, and the Fe<sub>2</sub>O<sub>3</sub> content decreased. The calcium aluminate was basic, therefore the B of PFAC increased with the increasing dosage of calcium aluminate. However, the increase

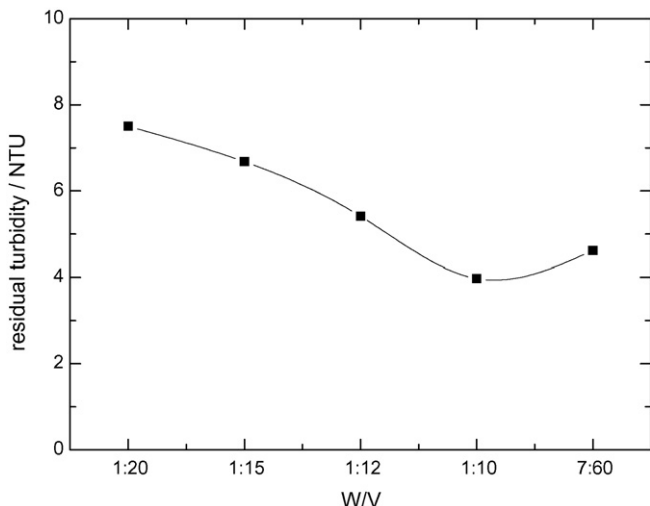


Fig. 2. Effect of W/V on the coagulation performance of PFAC.

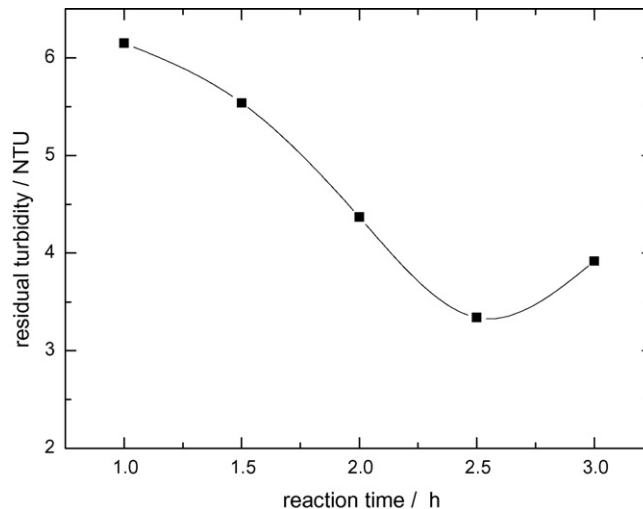


Fig. 4. Effect of reaction time on the coagulation performance of PFAC.

of  $B$  might lead to the hydrolyzation and sedimentation of  $\text{Fe(III)}$ , and the  $\text{Fe}_2\text{O}_3$  content decreased with the increasing dosage of calcium aluminate.

Fig. 2 shows that after dosing PFAC coagulant, the residual turbidity of the synthetic water decreased with the increase of  $W/V$  (from 1:20 to 1:10), followed by increased after  $W/V$  higher than 1:10. Generally, the higher the  $B$  of the coagulant, the higher the polymerization extent formed by the hydrolyzing  $\text{Fe(III)}$  and/or  $\text{Al(III)}$ . The higher the polymerization extent, the stronger the charge neutralizing and adsorption-bridging capacity of the coagulant. The stronger the charge neutralizing and adsorption-bridging capacity of the coagulant, the better the coagulation performance of the coagulant. However, if the  $B$  of PFAC was too high,  $\text{Fe(III)}$  in the PFAC product might easily hydrolyze and sedimentate, leading to the decrease of  $\text{Fe}_2\text{O}_3$  content of PFAC and decreasing the coagulation performance and stability of PFAC. For the above reasons, 1:10 was considered as the optimum  $W/V$ .

### 3.1.2. Reaction time

In order to study the effect of reaction time, the  $W/V$  was kept constant at 1:10, reaction temperature was kept constant at  $60^\circ\text{C}$ , while varying the reaction time. Results are shown in Figs. 3 and 4.

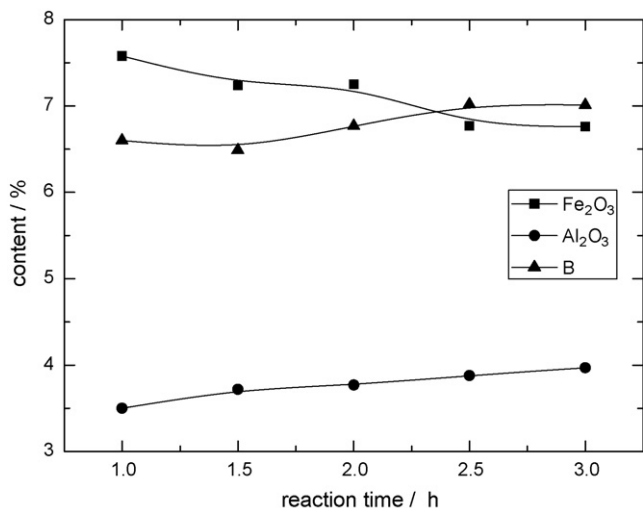


Fig. 3. Effect of reaction time on effective composition and  $B$  value of PFAC.

Fig. 3 shows that with the increase in reaction time, the  $\text{Al}_2\text{O}_3$  content and the  $B$  of PFAC slowly increased, while the  $\text{Fe}_2\text{O}_3$  content of PFAC decreased. The  $B$  content of PFAC increased with increased of reaction time, which may lead to the hydrolyzation and sedimentation of  $\text{Fe(III)}$  in the product and a decrease of the  $\text{Fe}_2\text{O}_3$  content. Fig. 4 shows that after dosing PFAC coagulant, the residual turbidity of the synthetic water decreased with the increase of reaction time (from 1.0 h to 2.5 h), followed by increase with the reaction time higher than 2.5 h. In addition, the shorter the reaction time, the lower the utilization rate of calcium aluminate. If the reaction time is very long, the effective compositions of PFAC might decrease and more energy will be needed, which might prevent the industrial preparation and application of PFAC. For the above reasons, 2.5 h was considered as the optimum reaction time.

### 3.1.3. Reaction temperature

In order to study the effect of reaction temperature, the  $W/V$  was kept constant at 1:10 and reaction time was kept constant at 2.5 h, while varying the reaction temperature.

Fig. 5 shows that with the increase of reaction temperature, the  $\text{Al}_2\text{O}_3$  content and the  $B$  of PFAC increased, while the  $\text{Fe}_2\text{O}_3$  content of PFAC drastically decreased. The higher the reaction tempera-

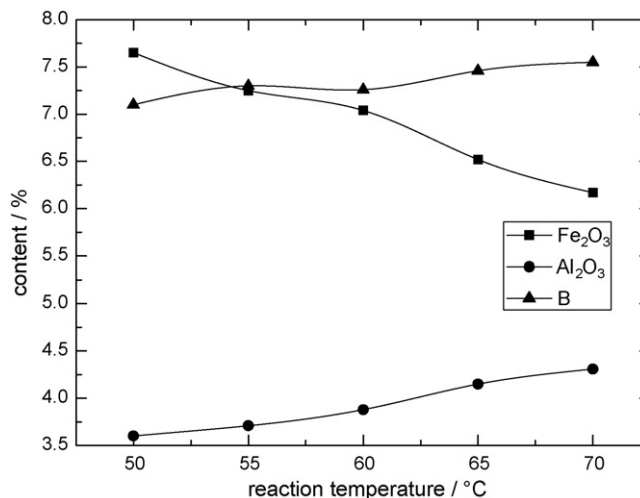


Fig. 5. Effect of reaction temperature on effective composition and  $B$  value of PFAC.

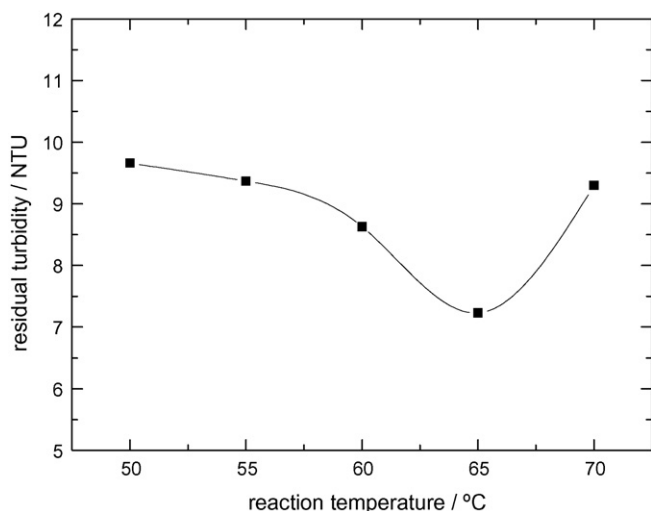


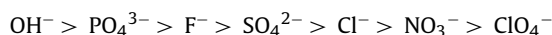
Fig. 6. Effect of reaction temperature on the coagulation performance of PFAC.

ture, the higher the reaction extent between calcium aluminate and hydrochloric pickle liquor which led to the increase of  $B$ . The increase of  $B$  might lead to the hydrolyzation and sedimentation of Fe(III) in PFAC product, therefore the  $\text{Fe}_2\text{O}_3$  content of PFAC decreased with the increase of reaction temperature. As a result, the performance and stability of PFAC decreased. Furthermore, with the increase of reaction temperature, more energy was needed for preparing PFAC, which might prevent the industrial preparation and application of PFAC.

The coagulation performance of different PFAC products prepared at different reaction temperatures is shown in Fig. 6. It could be seen that after dosing PFAC coagulants prepared at different temperatures, the residual turbidity of the synthetic water decreased with the increase of reaction temperature (from 50 °C to 65 °C), followed by increase after the reaction temperature was higher than 65 °C. The turbidity removal efficiency of PFAC prepared at 65 °C was the best. For the above reasons, 65 °C was considered as the optimum reaction temperature.

### 3.2. Stability of PFAC

High quality coagulants should not only give a good coagulation performance, but also have good stability when stored. The PFAC prepared under the above optimum conditions gave a good coagulation performance, but its stability was poor. After 15 days, Fe(III) in the PFAC product was partly sedimentated and the PFAC product became cloudy. Erik Hogfeldt proved that Fe(III) had a strong affinity with  $\text{OH}^-$ , the affinity of Fe(III) with different anions was on the decrease [18]:



Anions such as  $\text{PO}_4^{3-}$ ,  $\text{F}^-$  and  $\text{SO}_4^{2-}$ , could prevent the combination of Fe(III) and  $\text{OH}^-$  and then prevent the hydrolyzation and sedimentation of Fe(III), so anions could extend the storage time of PFAC products. Therefore, it was necessary to investigate the effect of different anions on the stability of PFAC products. Phosphoric acid, sodium phosphate, sodium dihydrogen phosphate and sodium sulfate were evaluated as the stabilizing agents.

Fig. 7 shows that under the same S/M of 0.06, the stabilizing effect of  $\text{Na}_3\text{PO}_4$  was the best, while the stability effect of  $\text{Na}_2\text{SO}_4$  was the worst. Counter ions such as  $\text{PO}_4^{3-}$  and  $\text{H}_2\text{PO}_4^-$  could play a special protective role in polynuclear hydroxy complex formed by the hydrolyzation of Fe(III).  $\text{PO}_4^{3-}$  and  $\text{H}_2\text{PO}_4^-$  could

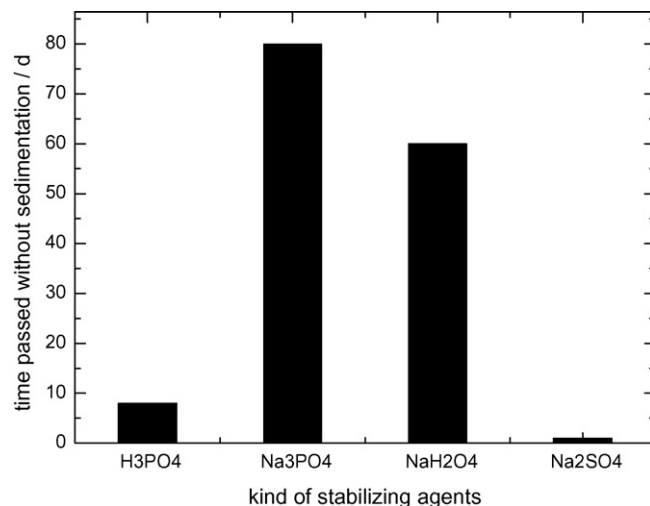


Fig. 7. Effect of kind of stabilizing agents on the stability of PFAC.

replace some hydroxy group of the polynuclear hydroxy complex and then weaken the strong affinity of  $\text{Fe}^{3+}$  and  $\text{OH}^-$ , therefore,  $\text{PO}_4^{3-}$  and  $\text{H}_2\text{PO}_4^-$  could prevent the hydrolyzation and sedimentation of Fe(III).  $\text{SO}_4^{2-}$  could also play the same role in preventing the hydrolyzation and sedimentation of Fe(III), but its effect was very weak [19]. For the above reasons,  $\text{Na}_3\text{PO}_4$  was chosen as the optimum stabilizing agent. The effect of its dosages on the stability and performance of PFAC was investigated. Results are shown in Figs. 8–10.

Fig. 8 shows that the storage time of PFAC increased with the increase of S/M. When the S/M reached 0.10, the storage time of PFAC was extended to more than 5 months. Fig. 9 shows that with the increase of S/M, the  $\text{Al}_2\text{O}_3$  content and the  $\text{Fe}_2\text{O}_3$  content of PFAC slightly decreased, while the  $B$  of PFAC increased drastically. As a strong-alkali weak-acid salt,  $\text{Na}_3\text{PO}_4$  could alter the pH of PFAC product, therefore it could increase the  $B$  of PFAC. Fig. 10 shows that after dosing PFAC coagulants, the residual turbidity of the synthetic water decreased with the increase of S/M (from 0 to 0.10), followed by increased with the S/M higher than 0.10. The  $\text{Na}_3\text{PO}_4$  stabilizing agent could strengthen the polymerization extent of PFAC and then promote the coagulation performance of PFAC. But the higher the S/M, the higher the consumption of the  $\text{Na}_3\text{PO}_4$  stabilizing agent and the higher cost of preparing PFAC, which might prevent the

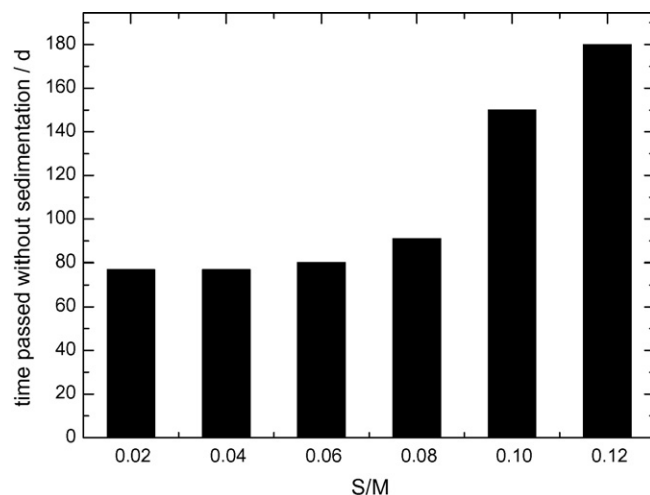


Fig. 8. Effect of S/M on the stability of PFAC.

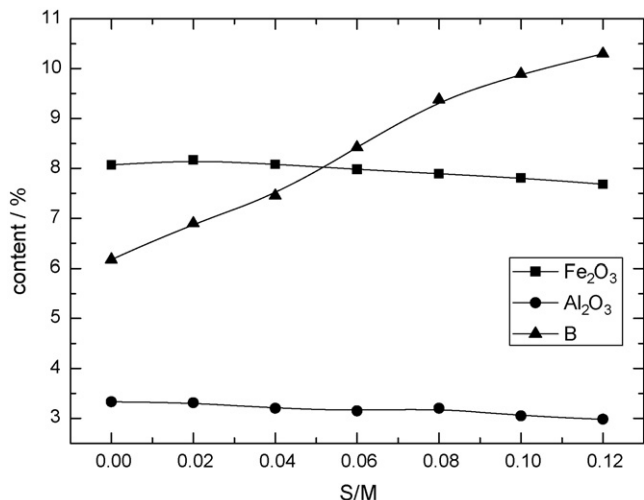


Fig. 9. Effect of S/M on effective composition and B value of PFAC.

industrial preparation and application of PFAC. Therefore, S/M of 0.10 was chosen as the optimum dosage of Na<sub>3</sub>PO<sub>4</sub> stabilizing agent.

### 3.3. Coagulation performance of PFAC

In order to investigate the coagulation performance of PFAC, it was applied in the treatment of actual municipal sewage and compared with three market coagulants (PAC, PFS and FeCl<sub>3</sub>).

As shown in Figs. 11 and 12, the turbidity and COD removal rates of PFAC were evidently higher than that of PFS and FeCl<sub>3</sub> but somewhat lower than that of PAC in the treatment of municipal sewage. In the range of dosage researched, compared with PFS and FeCl<sub>3</sub>, PFAC could promote the turbidity and COD removal rates from 0.9% to 2.1% and from 2.9% to 9%, respectively.

With the COD removal rate of 71.6%, the dosages of PFAC, PFS and PAC coagulants were 6.29 mg/L, 10.00 mg/L, 3.45 mg/L, respectively (the highest COD removal rate of FeCl<sub>3</sub> was 64.4%), and the cost of using PFAC, PFS and PAC was  $3.07 \times 10^{-6}$  \$/L,  $5.64 \times 10^{-6}$  \$/L,  $3.89 \times 10^{-6}$  \$/L respectively. The treatment cost of PFAC was much less than that of PFS and PAC, therefore it has a good potential for commercial applications.

It could be seen in Figs. 13 and 14 that the total phosphate and NH<sub>3</sub>-N removal rates of PFAC were much higher than that of PFS,

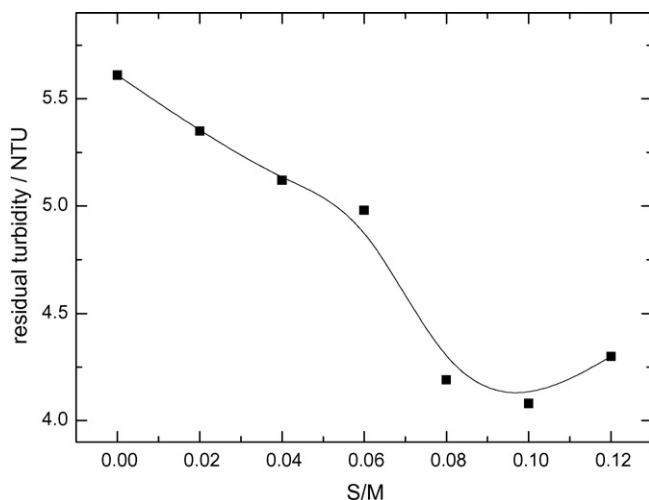


Fig. 10. Effect of S/M on the coagulation performance of PFAC.

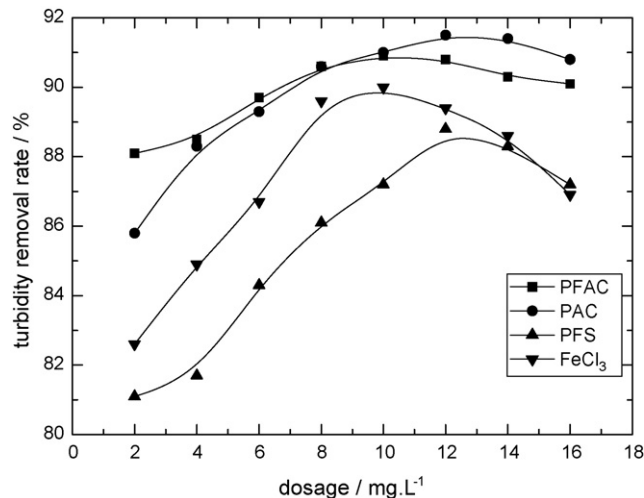


Fig. 11. Effect of coagulant dosage on the turbidity removal rate.

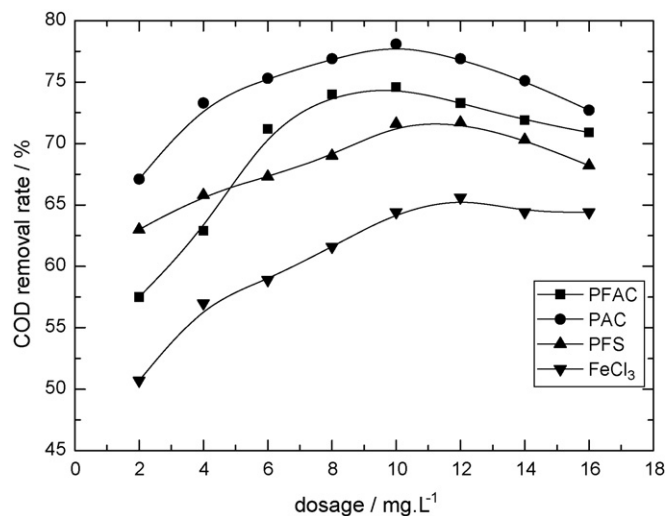


Fig. 12. Effect of coagulant dosage on the COD removal rate.

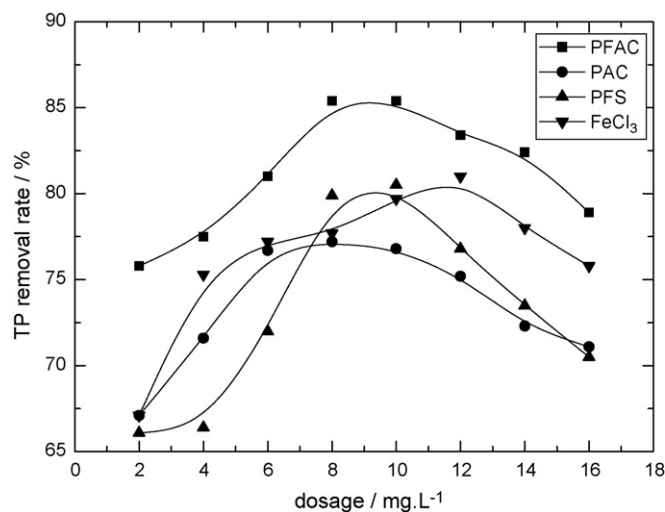


Fig. 13. Effect of coagulant dosage on the TP removal rate.

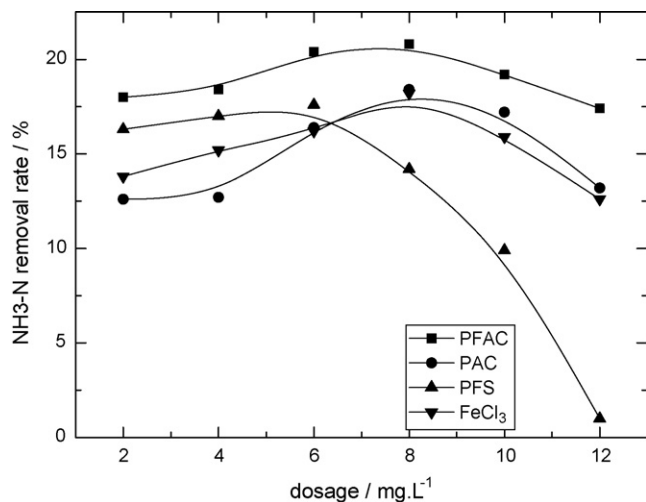


Fig. 14. Effect of coagulant dosage on the NH<sub>3</sub>-N removal rate.

FeCl<sub>3</sub> and PAC. PFAC could promote the total phosphate and NH<sub>3</sub>-N removal rates from 4.4% to 8.2% and from 2.4% to 3.2%, respectively. Therefore, the coagulation performance of PFAC prepared by hydrochloride pickle liquor was considerable in the treatment of municipal sewage.

#### 4. Conclusion

Polyferric aluminum chloride prepared by hydrochloric pickle liquor, not only gave a good coagulation performance, but also had good stability when stored (without sedimentation or gelatinization after 5 months). The COD and turbidity removal efficiency of PFAC was better than that of PFS and FeCl<sub>3</sub> in the treatment of municipal sewage, and the TP and NH<sub>3</sub>-N removal efficiency of PFAC was much better than that of PFS, FeCl<sub>3</sub> and PAC.

Preparing PFAC by using hydrochloride pickle liquor as material, it provided a feasible way to recycle hydrochloride pickle liquor and a process to prepare coagulant with low-cost material, and also gain good economic, social and environmental benefits.

#### Acknowledgements

This work was supported by the National High-tech Research and Development Program of China (863 Program) (Nos.

2005AA60101005, 2007AA06Z347), the China National Natural Science Foundation-funded Project (No. 20677038) and the Innovation Action Plan of Shanghai Science and Technology Commission (No. 072312002).

#### References

- [1] A. López-Delgado, F.J. Alguacil, F.A. López, Recovery of iron from bio-oxidized sulphuric pickling waste water by precipitation as basic sulphates, *Hydrometallurgy* 45 (1997) 97–112.
- [2] E. Marañón, F. Suarez, F. Alonso, et al., Preliminary study of iron removal from hydrochloric pickling liquor by ion exchange, *Ind. Eng. Chem. Res.* 38 (1999) 2782–2786.
- [3] G. Csicsovszki, T. Kékesi, T.I. Török, Selective recovery of Zn and Fe from spent pickling solutions by the combination of anion exchange and membrane electrowinning techniques, *Hydrometallurgy* 77 (2005) 19–28.
- [4] M. Tomaszewska, M. Grypta, A.W. Morawski, Recovery of hydrochloric acid from metal pickling solutions by membrane distillation, *Sep. Purif. Technol.* 22–23 (2001) 591–600.
- [5] M. Tomaszewska, M. Grypta, A.W. Morawski, The influence of salt in solutions on hydrochloric acid recovery by membrane distillation, *Sep. Purif. Technol.* 14 (1998) 183–188.
- [6] K. Urano, T. Ase, Y. Naito, Recovery of acid from wastewater by electrodialysis, *Desalination* 51 (2) (1984) 213–226.
- [7] E. Paquay, A.M. Clarinval, A. Delvaux, et al., Applications of electrodialysis for acid pickling wastewater treatment, *Chem. Eng. J.* 79 (3) (2000) 197–201.
- [8] J. Wiśniewski, G. Wiśniewska, Water and acid recovery from the rinse after metal etching operations, *Hydrometallurgy* 53 (1999) 105–119.
- [9] P. Rengasamy, J.M. Oades, Interaction of monomeric and polymeric species of metal ions with clay surface. IV. Mixed system of aluminum (III) and iron (III), *Aust. J. Soil Res.* 17 (1979) 141–153.
- [10] Z. Chunlu, M. Wenlin, L. Zhenru, et al., Study of copolymerization of Al(III) and Fe(III) liquid, *Environ. Chem.* 15 (1) (1996) 36–40.
- [11] L. Shanping, Z. Yanli, L. Fubo, Preparing composite inorganic polymer flocculant PAFC by reusing steel mill waste, *Environ. Chem.* 24 (2) (2005) 168–170.
- [12] G. Baoyu, Y. Hui, Y. Qinyan, et al., Study on the preparation of polyaluminum ferric chloride from Gangue, *Environ. Sci.* 17 (4) (1996) 62–66.
- [13] Z. Zhanmei, Z. Huaili, C. Chunyan, Preparation and application of polymerize aluminum ferrum chloride with bauxite, *Techn. Equip. Environ. Pollut. Control* 7 (6) (2006) 52–55.
- [14] B.Y. Gao, H.H. Hahn, E. Hoffmann, Evaluation of aluminum-silicate polymer composite as a coagulant for water treatment, *Water Res.* 36 (2002) 3573–3581.
- [15] T.-K. Liu, E.S.K. Chian, Effect of base addition rate on the preparation of partially neutralized ferric chloride solutions, *J. Colloid Interf. Sci.* 284 (2005) 542–547.
- [16] J. Duan, J. Gregory, The influence of silicic acid on aluminium hydroxide precipitation and flocculation by aluminium salts, *J. Inorg. Biochem.* 69 (1998) 193–201.
- [17] APHA, AWWA, WEF, Standard Method for the Examination of Water and Wastewater, 20th ed., Washington DC, USA, 1998.
- [18] E. Hogfeldt, Stability Constants of Metal-Ion Complexes. Part A. Inorganic Ligands, Pergamon Press, New York, 1982.
- [19] T. Baozhen, T. Hongxiao, The hydrolyzed and other chemical characters of ferric chloride solution containing phosphate, *Environ. Chem.* 14 (4) (1995) 329–337.