

## Study on coagulation property of metal-polysilicate coagulants in low turbidity water treatment\*

YANG Hai-yan (杨海燕)<sup>†</sup>, CUI Fu-yi (崔福义), ZHAO Qing-liang (赵庆良), MA Chao (马超)

(School of Municipal & Environment Engineering, Harbin Institute of Technology, Harbin 150090, China)

<sup>†</sup>E-mail: haiyang\_hw@sina.com

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**Abstract:** In order to remove the low turbidity present in surface water, a novel metal-polysilicate coagulant was used to treat the raw water taken from Tanjiang River in Guangdong Province. This study on the effects of Al/Fe molar ratio on the performance of a complex compound formed by polysilicic acid, aluminium and ferric salt (PAFS) showed that PAFS with Al/Fe ratio of 10:3 seemed to have the best coagulation performance in removing turbidity and color. Experimental results showed that under the conditions of polymerization time of 15 d, sedimentation time of 12 min, and pH of 6–8, PAFS with Al/Fe molar ratio of 10:3 had the best coagulation efficiency and lowest residual Al concentration. The turbidity decreased from 23.8 NTU to 3.23 NTU and the residual Al concentration was only 0.165 mg/L in the product water. It could be speculated that colloidal impurities and particulate Al were removed by adsorption bridging and electrical neutralization of long chain inorganic polymer coagulants.

**Key words:** Potable water treatment, Metal-polysilicate coagulant, Coagulation property, Residual aluminum

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### INTRODUCTION

Coagulation-flocculation is a major step in the potable water treatment for removal of colloidal impurity. Due to the increase of water pollution, the standards of drinking water supply and wastewater discharge are becoming more and more stringent, and chemical coagulants that are more efficient in water treatment are more and more urgently in demand.

As well known, the residual Al in potable water comes from three main sources: natural water-body, coagulants used in the water treatment process and dissolution from water pipes. In the

past decade, more and more attention has been paid to aluminum toxicity to human health (Crapper, 1973; Jiang and Liang, 1999; Cui *et al.*, 2002). A new kind of water treatment coagulant, the inorganic macromolecule coagulant, was recently developed. Polysilicate coagulant combined with ferric salt and aluminium salt work especially well in treating low temperature and turbidity water. Moreover, polysilicate has several advantages such as facileness, abundance and nontoxicity, so comprehensive studies on polysilicate were conducted by researchers all over the world (Hasegawa *et al.*, 1990; 1991; Luan and Song, 1997; Gao *et al.*, 2000).

This paper focuses on the effects of metal-polysilicate coagulants used for treating Southern China raw water at ambient temperature and low turbidity. Results of a coagulation test for

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determination of turbidity, color and residual Al, were applied to find the relationship between the coagulation performance and residual Al. The experimental results may provide information for practical applications in the future.

## MATERIALS AND METHODS

### Chemical reagents

Some kind of commercially available soluble glass (SS/CS/SDHT3883, SINOSI, China), rough aluminum sulfate coagulant (GW Aluminum Corporation of China), ferric chloride with 99%  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ , aluminon and other analytical reagents were used.

### Preparation of metal-polysilicate coagulants

Under magnetic stirring and pH control using an acidity meter (PHS-25, Shanghai REX Instrument Factory, China), soluble glass was diluted with diluent sulphuric acid to half of the original concentration. After a certain time, a definite concentration of  $\text{Al}_2(\text{SO}_4)_3$  or  $\text{FeCl}_3$  solution was added into the soluble glass. After a certain coagulation time, the metal-polysilicate coagulant (PAFS) was ready for use.

### Jar test methods

The raw water used for this study was taken from a river in Guangdong Province so that the experimental results might guide the local applications of the process. The raw water characteristics are summarized in Table 1.

The detailed experiment procedure was as follows. First, six liters of raw water were poured

into six beakers. Then, the coagulant solution was added into all the beakers simultaneously. Immediately after that, the solutions were rapidly mixed for 2 minutes at 150 rev/min, gently stirred for 10 minutes at 60 rev/min using jar tester (SC656, Hubei Meiyu, China) and then allowed to settle for half an hour. Finally, water samples were drawn from 2–3 cm below the water surface and determined for their residual Al (using 722 raster spectrophotometer, Shanghai Optical Instrument, China), turbidities (using WGZ-100, Shanghai Precision Instrument, China), colors and pH values.

## RESULT AND DISCUSSION

To assess the coagulant performance of PAFS, the coagulant dosages were measured in terms of their metal content.

### Effects of Al/Fe ratio on PAFS performance and residual Al

#### 1. Effects of Al/Fe ratio on PAFS performance

Raw water with turbidity of 16.2 NTU and color of 37 degree was used for coagulation test at different PAFS dosages. The experimental results are shown in Fig.1 and Fig.2 showing that PAFS with  $\text{Al/Fe} > 1$  had better performance than PAFS with  $\text{Al/Fe} < 1$  in treating low turbidity water. This meant that Fe(III) had significant influence on the coagulation during the course of preparation of metal-polysilicate coagulants. At a lower dosage of PAFS, turbidity removal decreased with increasing Fe content in the coagulants. Under condition of small Fe amount ( $\text{Al/Fe} = 10:3$ ) present in PAFS, PAFS performed best in turbidity removal. At a higher dosage of PAFS, PAFS containing only ferric salt performed better in removing turbidity than PAFS with  $\text{Al/Fe} < 1$ . In addition, all of the mentioned PAFS coagulants showed excellent coagulation performance at relatively higher dosages.

As shown in Fig.2, color removal decreased at a lower dosage of PAFS as the Fe content increased. Under the condition of small Fe contents ( $\text{Al/Fe} = 10:3$ ), PAFS yielded the best result. Due to the presence of Fe in PAFS, the color increased if PAFS

**Table 1 Raw water characteristics**

Parameters	Unit	Value
Temperature	°C	25–34
Turbidity	NTU	11–40
Color	Degree	30–50
pH	Unitless	6.5–7.5
Permanganate index	mg $\text{O}_2/\text{L}$	4.0–6.0
Al	mg/L	0.049–0.078

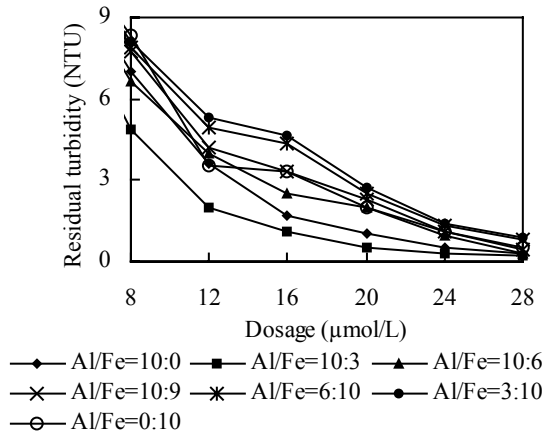


Fig.1 Turbidity removal by PAFS under different Al/Fe ratios

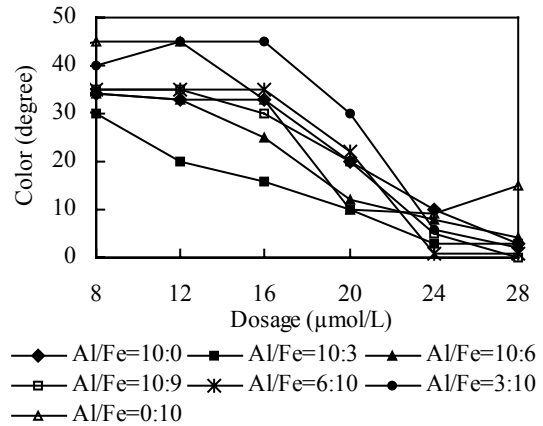


Fig.2 Color removal by PAFS under different Al/Fe ratios

contained only ferric salt at a high dosage of 28 μmol/L. All of the mentioned PAFS coagulants exhibited excellent performance in removing color.

### 2. Effects of Al/Fe ratio on residual Al

Residual Al in the clarified water treated by PAFS under different Al/Fe ratios is shown in Fig.3. With increased coagulant dosages, the concentration of residual Al in the clarified water increased initially and decreased subsequently, while the residual turbidity decreases linearly. The smaller the Al/Fe molar ratio was, the lower was the concentration of residual Al. Moreover, the residual Al concentration in the water treated by PAFS (Al/Fe=0) was much less than that in the water treated by other PAFS (different Al/Fe ratios). This was due to the following two reasons:

(1) Single-nucleus or multi-nucleus hydroxyl chelate compound generated from dissolved aluminum hydrolysis carried positive charges and was encapsulated in suspended granules during electrical neutralization interactions. So the residual Al concentration, and turbidity present in the clarified water was higher under low coagulation dosages.

(2) In contrast, the concentration of residual Al and the turbidity decreased under high coagulant dosages, because the aluminum hydroxide amount was reduced due to the interactions between polysilicic acid and various species of hydrolyzed aluminum. And PAFS is a long chain inorganic polymer coagulant that removes colloidal impurities and hydrolytical byproducts in the

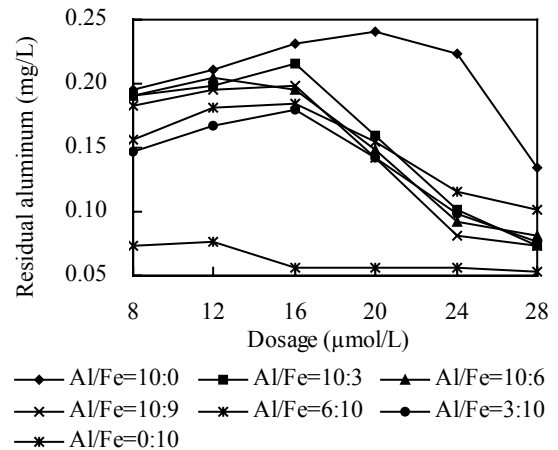


Fig.3 Residual Al in the clarified water under different Al/Fe ratios

form of particulate Al in water by adsorption-bridging and sweep-coagulation mechanisms.

It can be noted from the above results that PAFS with Al/Fe ratio of 10:3 can remove turbidity and color effectively from low turbidity water and result in a lower concentration of residual Al in the clarified water. So the following sections focus on investigating PAFS performance under the influence of other factors, keeping the Al/Fe ratio at 10:3.

### Effects of polymerization time on turbidity removal and residual Al

To investigate the effect of polymerization time on turbidity removal and on the residual Al in

the treated water, a test was conducted to treat the raw water with turbidity of 12 NTU and Al of 0.069 mg/L. The experimental results are shown in Fig.4 and Fig.5. Polymerization time of 1, 15 and 30 days was adopted respectively for the study.

Fig.4 and Fig.5 show that the effect of polymerization time on PAFS performance and residual Al are not very obvious. As for turbidity removal (Fig.4), the polymerization time of 30 days showed indistinct polymerized jelly because coagulant activity decreased when the polysilicic acid with long-chain structure changed into silica gel. However, the residual Al changed slightly (Fig.5) because the long polymerization time (i.e. 30d) changed PAFS with long-chain into polysilicic acid and amphoteric hydroxide which consisted mainly of particulate Al. The residual Al in water existed mostly in dissolved form after the particulate Al was removed.

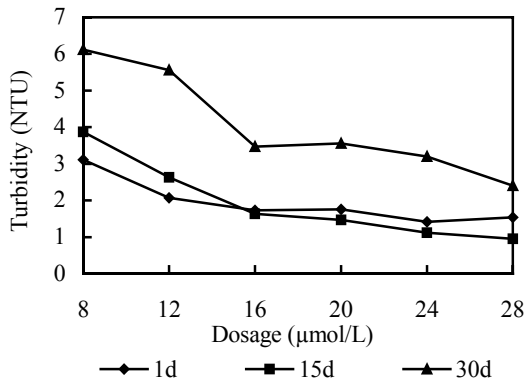


Fig.4 The effects of polymerization time on turbidity removal

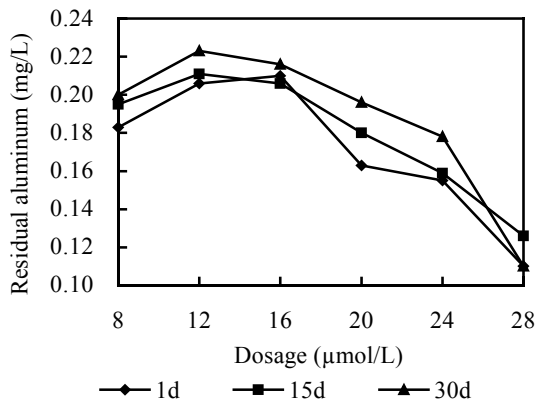


Fig.5 The effects of polymerization time on residual Al

### Relationship between effective coagulation area of PAFS and residual Al

The increase in coagulant dosage is often used to solve problems caused by the sudden change in raw water's quality. Unfortunately, due to the affects of zeta potential, a high dosage is sometimes not so ideal as expected. Therefore, the relationship between coagulation and residual Al was studied in treating water with turbidity of 18.6 NTU and Al concentration of 0.078 mg/L. The result is shown in Fig.6.

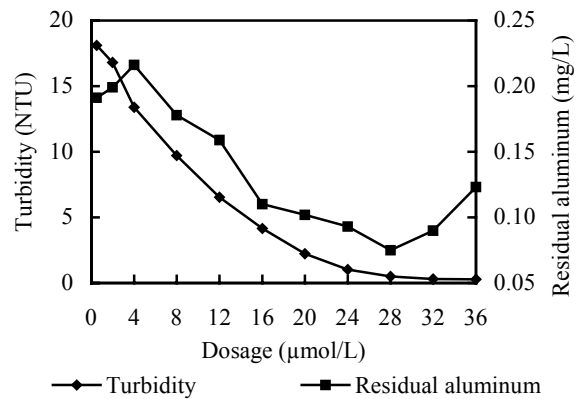


Fig.6 The effects of the PAFS dosage (Al/Fe=10:3) on turbidity and residual Al

With increased PAFS dosage, the treated water's turbidity decreased gradually and never bounced back, while most other coagulants at high dosage might even increase the turbidity. Even under the condition that the dosage was less than 28 μmol/L, the residual Al also decreased along with the turbidity. This can be explained as follows: the Al in water existed only in the dissolved and particulate forms and was encapsulated in suspended impurities, resulting in a similar trend for the turbidity and the residual Al. When the dosage was less than 28 μmol/L, Al particulates were adsorbed and swept by PAFS and only a small part of them were left in the water. Whereas, residual Al increase was mainly caused by the dissolved Al brought in with PAFS.

### Effects of pH on turbidity removal and residual Al

It is known that the pH is one of the main

factors that influence the gelation time. To investigate pH influence on the coagulation performance and residual Al, a test was performed to treat the raw water with turbidity of 26 NTU, pH of 6.8 and Al concentration of 0.069 mg/L.

Fig.7 shows that both the turbidity and the residual Al in water decrease initially while the pH increases. Afterwards, with pH increased, both the turbidity and the residual Al begin to increase. There exists an optimal range of pH for turbidity removal. This test yielded evidence that PAFS had the best coagulation performance in the pH range of 5.5 to 8 and minimized at pH of 6. Moreover, pH also had considerable effects on the residual Al. Residual Al concentration is relatively high in water with pH<6 or >9. This is due to the following facts: (a) the shift of aluminum hydrolysis equilibrium increases the dissolved Al; (b) not all of the Al particulates are adsorbed by long-chain polysilicate and then removed by sedimentation. The residual Al concentration was relatively low in the pH range of 6 to 9, and minimized at pH 7. To sum up, the optimal pH range for PAFS was from 6 to 8.

**Effects of sedimentation time on flocculation**

Previous study phase revealed that the floc formed by PAFS had such advantages as short formation time, large size, and easy sedimentation. To investigate the effects of sedimentation time on flocculation, we conducted a test wherein the raw water with turbidity of 23.8 NTU and Al concentra-

tion of 0.078 mg/L was treated under the coagulant dosage of 16 μmol/L. The results are shown in Fig.8. Obviously, PAFS took only 8 minutes to remove the turbidity, while other traditional coagulants required 20 minutes. The flocs were large, solid and settled easily which fully revealed the good performance of this kind of coagulant's macromolecule property including fast electrical neutralization and fast adsorption bridging. After settling for 12 min, the residual Al remained at 0.165 mg/L. The settlement time for removing the residual Al was almost equal to that for removing the turbidity, which proved that residual Al exists mainly in particulate form. Fortunately, as the turbidity present in water is linked to the residual Al concentration, this method can be applied to remove both the turbidity and the residual Al in water.

**CONCLUSION**

PAFS is the complex compound formed by polysilicic acid, aluminium salt and ferric salt through copolymerization. Based on the study of the coagulation for reducing the residual aluminium in purifying low turbidity water with PAFS, the following conclusions can be drawn:

Under condition of a low dosage, PAFS's performance in removing the turbidity and color declined slightly while the Fe content increased. But under high dosage, PAFS showed excellent per-

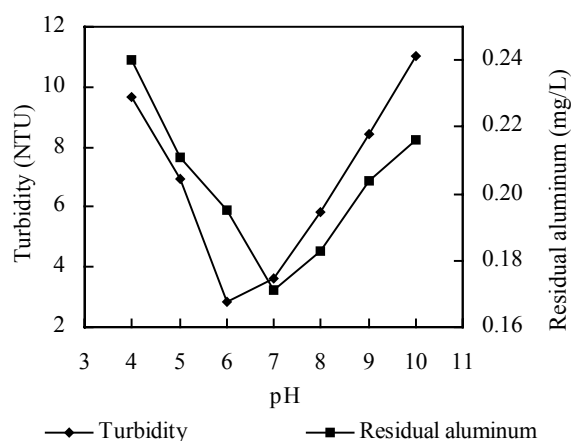


Fig.7 The effects of pH on turbidity removal and residual Al

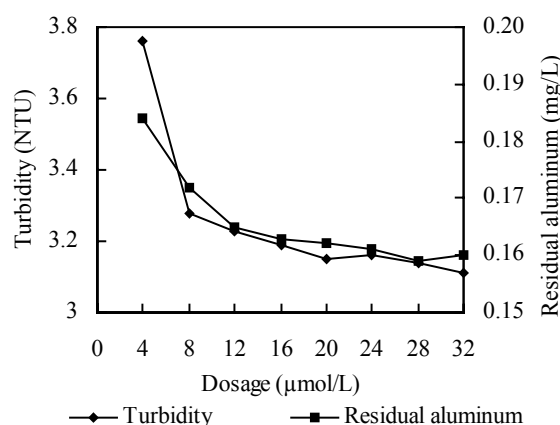


Fig.8 The effects of sedimentation time on turbidity removal and residual Al

formance in removing both. The Al/Fe molar ratio affected the PAFS's performance to some extent, and the optimal Al/Fe molar ratio was 10:3.

Polymerization time had some influence on the PAFS's performance but less influence on the residual Al.

Under high dosage, the phenomenon of colloid restabilization did not appear during the whole experimental period, while the residual Al content increased somewhat.

In terms of coagulation performance and residual Al, the most optimal pH range of PAFS was found to be 6 to 8.

The flocs formed by PAFS were large and easily settled within 12 minutes. PAFS had better coagulation property and require less sedimentation time compared to other coagulants.

The residual Al in the clarified water treated by PAFS was low because the interactions between polysilicic acid and various species of hydrolyzed aluminum reduced hydrolytical byproduct in the form of particulate Al, and inorganic polymer coagulant with long-chain structure removed further the colloidal impurities by adsorption-bridging and sweep-coagulation.

## References

- Crapper, D.R., 1973. Brain Aluminum distribution in Alzheimer's disease and experimental neurofibrillary degeneration. *Science*, **180**:511-513.
- Cui, F.Y., Zhang, X.Y., Feng, Q., Zhou, B., Zhou, F.T., 2002. Correlation between turbidity and aluminum in conventional water treatment. *Journal of Harbin University of Civil Engineering and Architecture*, **35**(3):52-55 (in Chinese with English abstract).
- Gao, B.Y., Yue, Q.Y., Wang, Z.S., Tang, H.X., 2000. Study on the species distribution of polyaluminum silicate chloride (PASC). *Environmental Chemistry*, **19**(1):1-12 (in Chinese with English abstract).
- Hasegawa, T., Onitsuka, T., Suzuki, M., et al., 1990. New polysilicic acid coagulation and their properties. *Water Supply*, **8**(3-4):152-161.
- Hasegawa, T., Hashimoto, K., Onitsuka, T., et al., 1991. Characteristics of metal-polysilicate coagulation. *Water Science and Technology*, **23**(7):1713-1722.
- Jiang, S.J., Liang, J.J., 1999. Hazard and control of residual aluminum in drinking water. *Journal of Chongqing Jianzhu University*, **21**(6):27-30 (in Chinese with English abstract).
- Luan, Z.K., Song, Y.H., 1997. Preparation and flocculation of polysilicate-metals (PSMS) flocculants. *Environmental Chemistry*, **16**(6):534-539 (in Chinese with English abstract).

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