

# Effect of Al(III) speciation on coagulation of highly turbid water

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## Abstract

In Taiwan, the turbidity of raw water for fresh water treatments can sometimes reach as high as 40000 NTU due to intensive rainfall, especially in typhoon seasons. In response, water works often apply large quantities of coagulants such as polyaluminium chloride (PACl). In this study, simulated and natural highly turbid water was coagulated with two PACls, a commercial product (PACl-1) and a laboratory product (PACl-E). The Al species distributions of PACl-1 and PACl-E under various pH conditions were determined, and the corresponding coagulation efficiency was evaluated. The PACl-E has a wider range of operational pH, while the efficiency of PACl-1 peaks at around neutral pH. For simulated water up to 5000 NTU, the PACl-E was superior to PACl-1 at low dosage and in the pH range studied. Similar results were discovered with natural water, except that when the turbidity was extremely high, the coagulation efficiency of PACl-E decreased significantly due to the presence of large amounts of organic matter. The coagulation of PACl-E was closely related to the content of polycationic aluminium ( $Al_{13}$ ) while that of PACl-1 was dictated by the amount of  $Al_3$ . The sludge from PACl-E coagulation had better dewaterability when the optimum dosage was applied. The experimental results suggest that for natural water up to 5000 NTU, PACl containing high  $Al_{13}$  species is recommended for coagulation. In cases when the water contains high organic matter, efficient coagulation depends upon enmeshment by amorphous aluminium hydroxide.

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## 1. Introduction

During typhoon seasons in Taiwan, excessive upstream erosion has substantially raised the turbidity of surface waters. Consequently, the raw water turbidity for water purification is extremely high. Turbidity of the raw water at the pump station can reach over tens of thousands NTU, which can significantly handicap the water treatment plant. Researchers have explored strategies such as full-scale sludge blanket, dissolved air floatation (DAF) and two-stage filtration to treat highly turbid water (Lin et al., 2004; Kwon et al., 2004; Di Bernardo and Di Bernardo, 2006). Unfortunately, the applicable range of

turbidity was substantially lower than what we have encountered in Taiwan. The highly turbid water also occurs annually in the arid zone of western China, where water treatment plants draw raw water from the Yellow River. The river water contains extremely high concentration of solids, which is carried into water treatment plants such as the Langchou water treatment plant. In response, several large pre-sedimentation tanks have been constructed prior to upstream of the conventional water treatment plant. After the sedimentation of great amounts of suspended particles, the supernatant was then delivered to the treatment plant. The coagulant was replaced by polyacrylamide in the coagulation process to enhance turbidity removal (Fang and Fu, 2004). Because of land scarcity in Taiwan, expansion of the sedimentation tank is beyond reach. Most waterworks in Taiwan generously apply polymers such as polyaluminium chloride (PACl)

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in hopes of bringing down the high turbidity. In the existing facility, however, performance was limited.

Poor performance of PACl has been blamed on its low content of critical hydrolysis products, especially polycationic aluminium ( $\text{Al}_{13}$ ).  $\text{Al}_{13}$  is a highly charged polymeric aluminium species,  $\text{Al}_{13}\text{O}_4(\text{OH})_{24}^{7+}$ , which is an effective agent for charge neutralization (Wang and Hsu, 1994). Studies have also confirmed that  $\text{Al}_{13}$  is the key to efficient coagulation of particulates and natural organic matter (NOM) by PACl (Wang et al., 2002; Gao et al., 2005; Hu et al., 2006). PACl is an intermediate product during hydrolysis, polymerization and precipitation of aluminium salts. There are different ways to produce PACl. Researchers have shown that the distribution of Al species in PACl varies according to the physical and chemical conditions during PACl production (Bertsch, 1987; Bottero et al., 1987; Klopogge et al., 1992; Qu and Liu, 2004; Hu et al., 2005). A PACl with over 70%  $\text{Al}_{13}$  has been produced by an electrolysis process (Liu et al., 1999a). PACl containing over 90%  $\text{Al}_{13}$  has also been prepared by sulfate precipitation and nitrate metathesis ( $\text{SO}_4^{2-}/\text{Ba}^{2+}$  separation method) from a pre-hydrolyzed PACl solution produced by alkaline titration method (Shi et al., 2007). Similar results can be achieved through separation and purification by ultra-filtration (Huang et al., 2006). In addition, the pH of the solution has a profound effect on Al speciation of PACl during coagulation processes, depending mainly on its  $\text{OH}^-/\text{Al}$  ratio (Wang et al., 2004). Studies have shown that treatment of highly turbid water is affected by rapid mixing and humic acid content (Kan et al., 2002; Annadurai et al., 2004). However, there has been limited research investigating the effects of Al speciation on coagulation for highly turbid water treatments. To determine the effectiveness and optimum conditions of PACl on treating highly turbid water, the Al speciation of PACl in relation to the mechanism of coagulation must be examined thoroughly. In this study, experiments were carried out to investigate the effects of PACl speciation on coagulation efficiency for waters of different levels of turbidity. One commercial and one laboratory PACl product were applied to a range of highly turbid waters.

## 2. Materials and methods

### 2.1. Highly turbid water samples

The sediment for the preparation of the highly turbid water samples was collected from the Shihmen Reservoir located in Taoyuan County, Taiwan. The sediment was mixed with tap water and aerated for 1 h, followed by settling for 30 min. After settling, the supernatant was used as the stock solution, to which deionized water was added to prepare the highly turbid water samples with the desired turbidity. Natural highly turbid water was also collected from rivers of northern, central, and southern Taiwan during heavy rainfalls. The suspension was also settled for

30 min, and the supernatant was collected as the natural highly turbid water sample.

Dissolved organic carbon (DOC) was measured by total organic carbon analyzer (TOC-5000A, Shimadzu, Japan). Particle size distribution and zeta-potential were characterized by particle size analyzer (Mastersizer 2000, Malvern, UK) and laser zeta analyzer (Zetasizer nano ZS, Malvern, UK), respectively. The characteristics of the two natural water (Natural 1 and Natural 2) and one simulated water (Simulated) samples are listed in Table 1. The table indicates that the properties of the simulated water were quite similar to those of the two natural waters, except that the turbidity as well as DOC and volatile solids of Natural 2 were substantially higher.

### 2.2. Coagulants

Two PACl products were used in this study. Reagent-grade PACl ( $\text{Al}_2\text{O}_3 = 10\%$ ) was purchased from Showa Chemicals Inc., designated as PACl-1. Another PACl ( $\text{Al}_2\text{O}_3 = 33\%$ ) named PACl-E, for the electrochemical process by which it is made, was received as a gift from the Research Center for Eco-environmental Sciences, Chinese Academy of Sciences in China. The  $\text{OH}^-/\text{Al}$  ratios,  $\gamma$ , of PACl-1 and PACl-E were 1.4 and 2.1, respectively. Working solutions containing  $1000 \text{ mg l}^{-1}$  as Al were freshly prepared from the solutions before each test. Aluminium concentration was analyzed by inductive coupled plasma atomic emission spectrometry (ICPAES) (JY24, Jobin-Yvon, France).

### 2.3. Jar tests

Standard jar tests were conducted in one-liter beakers to evaluate coagulation efficiencies. An initial rapid mixing was conducted at 200 rpm ( $G = 350 \text{ s}^{-1}$ ) for one min followed by a slow mixing at 30 rpm ( $G = 25 \text{ s}^{-1}$ ) for 20 min. Zeta-potential was measured immediately without dilution after the rapid mixing. The suspension was left undisturbed for 20 min. After settling, the residual turbidity (RT) of the supernatant was measured.

Table 1  
Characteristics of simulated and natural highly turbid water

Parameters	Stock water samples		
	Simulated	Natural 1	Natural 2
Turbidity (NTU)	–	300–7500	35000
Turbidity after 30 min settling (NTU)	4500–5300	200–6500	10000
pH	8.1	7.6–8.2	8.1
Zeta-potential (mV)	–22 to –18	–21.9 to –13.8	–18.1
Size ( $\mu\text{m}$ )	0.8–2.2	0.5–2.3	2.0
Alkalinity ( $\text{mg l}^{-1}$ as $\text{CaCO}_3$ )	100–120	70–200	191
DOC ( $\text{mg l}^{-1}$ )	2.0–3.0	2.1–2.7	8.5
VS of TS (%)	4.5	1.2–3.7	8.3

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