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Hydrolysis of polyaluminum chloride prior to coagulation: Effects on coagulation behavior and implications for improving coagulation performance

Zhongguo Zhang^{1,*}, Jun Wang^{2,3,*}, Dan Liu¹, Jiuyi Li⁴, Xiaolin Wang⁵, Boyu Song¹, Bing Yue^{1,6}, Kehui Zhao^{1,6}, Yun Song^{1,6}

1. Key Laboratory of Energy-Water Conservation and Wastewater Resources Recovery (Environmental Protection Research Institute of Light Industry), China National Light Industry, Beijing 100089, China

2. University of Chinese Academy of Sciences, Beijing 100049, China

3. Beijing Key Laboratory of Industrial Wastewater Treatment and Resource Recovery, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China

4. Department of Civil and Environmental Engineering, Beijing Key Laboratory of Aqueous Typical Pollutants Control and Water Quality Safeguard, School of Civil Engineering, Beijing Jiaotong University, Beijing 100044, China

5. Beijing Water Authority, Beijing 100038, China

6. Beijing Key Laboratory of Industrial Land Contamination and Remediation, Environmental Protection Research Institute of Light Industry, Beijing Academy of Science and Technology, Beijing 100089, China

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ABSTRACT

The effects of polyaluminum chloride (PACl) hydrolysis prior to coagulation on both the coagulation zone and coagulation performance of a kaolin suspension were investigated by a novel jar test named the “reversed coagulation test”. The tests showed that PACl hydrolysis prior to coagulation decreased the performance of charge neutralization coagulation in the case of short-time slow mixing (10 min; $G = 15 \text{ sec}^{-1}$) and increased the optimal dosage for charge neutralization and sweep coagulation. Moreover, the hydrolysis time had insignificant effects on the size and zeta potential of PACl precipitates and the residual turbidity of the raw water. However, PACl hydrolysis prior to coagulation and the size of PACl precipitates had a negligible effect on the performance of sweep coagulation. The results imply that, in practice, preparing a PACl solution with deionized water, rather than tap water or the outlet water from a wastewater treatment unit, can significantly save PACl consumption and improve the performance of charge neutralization coagulation, while preparing the PACl solution with tap or outlet water would not affect the performance of sweep coagulation. In addition, the optimal rapid mixing intensity appears to be determined by a balance between the degree of coagulant hydrolysis before contacting the primary particles and the average size of flocs in the rapid mixing period. These results provide new insights into the role of PACl hydrolysis and will be useful for improving coagulation efficiency.

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* Corresponding authors. E-mail: cn.zhang@163.com (Zhongguo Zhang), junwang@rcees.ac.cn (Jun Wang).

Introduction

Coagulation is an important process in the pretreatment of water and wastewater. Charge neutralization (Yu et al., 2010; Zhao et al., 2011b) and sweep coagulation (Zhao et al., 2011b) are the most important coagulation mechanisms in coagulation processes using metal salt coagulants. Charge neutralization coagulation occurs through mutual aggregation between destabilized particles when coagulants reduce or eliminate the electrostatic repulsion between particles (Ghernaout and Ghernaout, 2012). In sweep coagulation, the particles in solution are removed by entrapment within a suspended coagulant precipitate (Ghernaout and Ghernaout, 2012). The dominant mechanism depends on solution pH (Vepsalainen et al., 2012) and coagulant dosage (Yu et al., 2010), and appears to be determined by the diphasic equilibrium state between soluble metal species and hydroxide precipitation (Duan and Gregory, 2003).

The charge neutralization mechanisms of conventional hydrolyzing salt coagulants primarily include adsorption of soluble, polymeric metal-hydroxide species (namely, adsorption-charge neutralization) (O'Melia and Stumm, 1967), surface precipitation (Farley et al., 1985), and precipitation-charge neutralization (Dentel, 1988; Dentel and Gossett, 1988). Adsorption-charge neutralization usually takes place under acidic conditions at very low coagulant concentrations (White, 1980). In most cases, surface precipitation or precipitation-charge neutralization play a more important role (Benjamin, 1983; Farley et al., 1985). According to the precipitation-charge neutralization mechanism proposed by Dentel and Gossett (1988), the zeta potential (ZP) of aggregate, $\zeta_{1,2}$, can be given as

$$\zeta_{1,2} = \frac{\zeta_1 A_1 + \zeta_2 A_2}{A_1 + A_2}, \quad (1)$$

where, ζ is ZP, A is the surface area at the shear plane, and 1 and 2 are precipitated aluminum or ferric hydroxide and original particles, respectively. The results predicted by Eq. (1) agreed with the data from closely controlled jar-test experiments involving the addition of aluminum salts (aluminum sulfate) to synthetic suspensions, over a wide range of pH, coagulant dosage, and particle concentrations (Dentel and Gossett, 1988). This implies that the precipitated hydroxide on particles plays a dominant role in charge neutralization.

Based on the above, it can be concluded that the precipitates formed by coagulant hydrolysis are of great importance in both charge neutralization coagulation and sweep coagulation for hydrolyzing coagulants such as aluminum sulfate and ferric chloride.

The most common pre-hydrolyzed coagulant, polyaluminum chloride (PACl), is being used widely in coagulation-flocculation processes because it has benefits over conventional hydrolyzing aluminum or iron salts, including better performance at low temperature (Van Benschoten and Edzwald, 1990; Yu et al., 2007), lower aluminum residuals (Yang et al., 2011), lower sludge volume (Aguilar et al., 2002), a smaller effect on the pH of raw water (Duan and Gregory, 2003; Gregory and Dupont, 2001), and more rapid flocculation (Gregory and Dupont, 2001). However, the effects of PACl hydrolysis on coagulation behaviors, especially the hydrolysis occurring before it is mixed with raw water

and the precipitates of PACl, are still not very clear, although we do know that there are significant differences between alum and PACl in morphology, precipitates and coagulation mechanisms (Hu et al., 2006; Van Benschoten and Edzwald, 1990; Wu et al., 2007). In practice, hydrolysis prior to coagulation usually occurs in water and wastewater treatment engineering, such as during the process of preparing a PACl stock solution before coagulation or in the case of an insufficient dispersion of PACl during the rapid mixing period.

To explore the effects of PACl hydrolysis prior to coagulation, especially the formation of PACl precipitates, on coagulation zones (i.e., stabilization, charge neutralization coagulation, restabilization and sweep coagulation zones) and coagulation performance, a series of jar tests, in particular a novel "reversed coagulation test", were conducted using a kaolin suspension as a model raw water. The implications of the experimental results for the improvement of coagulation performance are discussed. This work will be helpful for improving our understanding of the role of PACl hydrolysis and optimizing coagulation performance.

1. Materials and methods

1.1. Preparation of PACl stock solution

A commercial powder PACl (Nanning Chem. Eng. Co. Ltd., Nanning, China), with a basicity (OH/Al) of 1.35 and an Al_2O_3 content of 30% (wt), was dispersed into deionized water to prepare a concentrated stock solution of 2 mol/L Al. A fresh coagulant solution of 0.1 mol/L Al was prepared from the stock solution one day before each set of experiments, to avoid aging phenomena and improve reproducibility. Deionized water was used to prepare the PACl solution.

1.2. Synthetic kaolin suspension

Kaolin was used as a model suspension. A stock suspension of kaolin was prepared by dispersing 10 g of kaolin in powder form (Xudong Chemical Plant, Beijing, China; chemical grade) into 5 L of tap water to a concentration of 2 g/L, followed by rapid mixing for 24 hr. The experimental suspensions were prepared by diluting the stock suspension to 100 mg/L, with a corresponding turbidity of 68 NTU (Nephelometric Turbidity Unit). Tap water was used for diluting the stock kaolin suspension. The size distribution of the kaolin suspension, measured by a Mastersizer 2000 (Malvern Instruments Ltd., Worcestershire, UK), was in the range of 1.18 to 28.3 μm , with a mean volume diameter of 7.28 μm , as reported in a previous study (Zhang et al., 2013). The characteristics of the tap water are as following: SO_4^{2-} 36.5 mg/L, Cl^- 13.4 mg/L, NO_3^- 9.83 mg/L, total hardness as CaCO_3 224 mg/L, alkalinity as CaCO_3 145 mg/L, COD_{Mn} 1.3 mg/L, pH 7.2, $\text{Fe} < 0.05$ mg/L, and electrical conductivity 370 $\mu\text{S}/\text{cm}$ (Zhang et al., 2013). It should be noted that under alkaline conditions, the hardness of the tap water can cause the destabilization of kaolin particles to a certain extent during preparation of a kaolin suspension (Liu and Wei, 2012). The ability of Ca^{2+} and Mg^{2+} to destabilize kaolin particles, however, is far lower than that of Al^{3+} (Liu and Wei, 2012). Moreover, the kaolin suspensions used in all the

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