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## Deposition behavior of residual aluminum in drinking water distribution system: Effect of aluminum speciation

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### ABSTRACT

Finished drinking water usually contains some residual aluminum. The deposition of residual aluminum in distribution systems and potential release back to the drinking water could significantly influence the water quality at consumer taps. A preliminary analysis of aluminum content in cast iron pipe corrosion scales and loose deposits demonstrated that aluminum deposition on distribution pipe surfaces could be excessive for water treated by aluminum coagulants including polyaluminum chloride (PACl). In this work, the deposition features of different aluminum species in PACl were investigated by simulated coil-pipe test, batch reactor test and quartz crystal microbalance with dissipation monitoring. The deposition amount of non-polymeric aluminum species was the least, and its deposition layer was soft and hydrated, which indicated the possible formation of amorphous  $\text{Al}(\text{OH})_3$ .  $\text{Al}_{13}$  had the highest deposition tendency, and the deposition layer was rigid and much less hydrated, which indicated that the deposited aluminum might possess regular structure and self-aggregation of  $\text{Al}_{13}$  could be the main deposition mechanism. While for  $\text{Al}_{30}$ , its deposition was relatively slower and deposited aluminum amount was relatively less compared with  $\text{Al}_{13}$ . However, the total deposited mass of  $\text{Al}_{30}$  was much higher than that of  $\text{Al}_{13}$ , which was attributed to the deposition of particulate aluminum matters with much higher hydration state. Compared with stationary condition, stirring could significantly enhance the deposition process, while the effect of pH on deposition was relatively weak in the near neutral range of 6.7 to 8.7.

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### Introduction

Aluminum is one of the common metallic elements in natural water, and its concentration in drinking water is generally restricted by regulations all over the world. However, the factors influencing the concentration of aluminum in tap water are very complicated. Meeting the drinking water quality standards of aluminum is a great challenge for many

water utilities. Based on one investigation of drinking water quality in some cities of China, aluminum concentration collected from tap water could significantly exceed the standard limit of 0.2 mg/L (according to Chinese standards for drinking water quality, GB 5749-2006) (Li et al., 2013). Elevated levels of aluminum have been associated with health problems. For example, bone disorders were testified to be related with aluminum among dialysis dependent patients

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(Dahl et al., 2014). Additionally, aluminum accumulated in humans may cause severe nervous system diseases, such as Alzheimer's disease, amyotrophic lateral sclerosis and Parkinson's dementia (Dzulfakar et al., 2011).

The source of aluminum in finished drinking water may originate from raw water or be introduced from Al-based coagulants (Snoeyink et al., 2003). Previous investigations on residual aluminum conducted in China, the United States and the Europe, showed that aluminum salts used for coagulation could increase residual aluminum content significantly (Wang et al., 2010b). Polyaluminum chloride (PACl), has been widely applied as coagulants in drinking water treatment (Lin et al., 2014) due to its many advantages, such as being effective within a broader pH range, rapid aggregation and sedimentation of its flocs (Gao et al., 2002).

Various aluminum hydrolysis products exist in PACl, including  $\text{Al}^{3+}$ ,  $\text{Al}(\text{OH})^{2+}$ ,  $\text{Al}_2(\text{OH})_2^{4+}$ ,  $\text{Al}_3(\text{OH})_4^{5+}$ ,  $\text{AlO}_4\text{Al}_{12}(\text{OH})_{24}(\text{H}_2\text{O})_{12}^{7+}$  (Keggin- $\text{Al}_{13}$  species),  $[\text{Al}_{30}\text{O}_8(\text{OH})_{56}(\text{H}_2\text{O})_{24}]^{18+}$  ( $\text{Al}_{30}$  polymer) and  $\text{Al}(\text{OH})_3$  (Xu et al., 2011). The coagulation efficiency of PACl is highly related to its aluminum speciation, and  $\text{Al}_{13}$  is claimed as the most efficient species for coagulation in PACl (Hu et al., 2006). The detrimental effects of aluminum on humans are also highly related to its specific species (Wang et al., 2010a). As dissolved aluminum could be assimilated immediately, it shows high cytotoxicity to both animal and plant (Szatanik-Kloc and Jozefaciuk, 2007). The mononuclear aluminum form is known to be severely neurotoxic because it can accelerate the formation of neurofibrillary tangles and senile plaques in the brain. While, the toxicity of polynuclear aluminum is relatively small as a result of its slow dissociation process (Wang et al., 2010c). However, it is currently not clear on the transformation and fate of residual aluminum with different speciation in finished water from the treatment plant to the consumer taps.

It has been reported that aluminum ubiquitously existed in corrosion scales and loose deposits within cast iron pipes, lead pipes, plastic pipes and cement-mortar lined pipes (Snoeyink et al., 2003). The aluminum accumulated in pipe scale and sediments may release back into bulk water once the water chemistry or hydraulic condition changes, which could result in relatively high aluminum content in tap water. Furthermore, severe deposition of aluminum might weaken disinfection efficiency, increase turbidity and interfere water transport capacity.

In order to control residual aluminum concentration in drinking water, many studies were conducted and different measures were proposed. However, most previous research focused on the optimization of coagulant dosage and coagulation condition, the investigation of aluminum transformation and deposition in drinking water distribution system was relatively rare. To our knowledge, study on the migration and transformation of different aluminum species in pipe distribution process has not been reported.

The main objective of this work was to understand the deposition features of different aluminum species on pipe surface, particularly on inert interface. Aluminum content in real distribution pipe corrosion scales and loose deposits, obtained from two cities with different source water types, was firstly analyzed. The deposition behavior of different aluminum species was systematically investigated using both simulated coil-pipe test and batch reactor test. QCM-D (Quartz

Crystal Microbalance with Dissipation Monitoring) technique was applied to explore the characteristics of deposition layer formed between different aluminum species and inert interface. The influence of hydraulic condition (stagnant vs. stirring) and pH (within neutral range) on the deposition behavior of different aluminum species were also observed. The deposition mechanism of different aluminum species was discussed.

## 1. Materials and methods

### 1.1. Analysis of aluminum content in real water distribution pipe scales and deposits

Cast iron pipe and plastic pipe sections were obtained from a northern and a southern city of China. Pipe corrosion scales and loose deposits were collected, respectively, where available. Totally 76 samples were collected from the northern city, among them 67 samples were hard iron corrosion scales, 9 samples were surficial loose deposits. Fifteen samples were collected from the southern city, 6 hard iron corrosion scale samples and 9 surficial loose deposit samples (including 3 from plastic pipes). Aluminum content in these two type scales were analyzed using XRF (X-ray Fluorescence, Advant' XP, Thermo Electron, Ecublens, Switzerland) and the pretreatment procedures were performed according to the report of Yang et al. (2012).

### 1.2. Preparation of stock solutions with different aluminum species

Three stock solutions  $\text{Al}_0$ ,  $\text{Al}_{13}$  and  $\text{Al}_{30}$  with different aluminum speciation were prepared as follows. Their total aluminum concentration and speciation are provided in Table 1. All reagents used were of analytical grade. Milli-Q water was used to rinse all reactors and to prepare all solutions.

For  $\text{Al}_0$  solution, a measured amount of  $\text{AlCl}_3$  was directly dissolved into 100 mL milli-Q water. Such solution was dominated by monomeric aluminum and some oligomeric aluminum species. For  $\text{Al}_{13}$ , firstly, PACl with basicity ( $\text{OH}/\text{Al}$ , molar ratio) 2.0 was prepared by base titration method. 0.5 mol/L NaOH was slowly titrated into a predetermined amount of  $\text{AlCl}_3$  solution under magnetic stirring. The obtained PACl solution was mixed with  $\text{Na}_2\text{SO}_4$ , settled for 24 hr, then filtered to obtain the precipitate. Then 0.1 mol/L  $\text{BaCl}_2$  was added to the precipitate, rapid stirring for 3 hr. The supernatant was filtered by 0.45  $\mu\text{m}$  membranes after 10 min settlement to obtain purified  $\text{Al}_{13}$  polymer (Xu et al., 2014). For  $\text{Al}_{30}$  solution, 1.0 mol/L  $\text{AlCl}_3$  solution with a measured amount of  $\text{Na}_2\text{CO}_3$  was mixed to obtain a molar ratio of  $\text{Na}_2\text{CO}_3/\text{Al}^{3+} = 1.15$ , then heated at 80°C till the precipitation dissolved. After settling for 12 hr the

**Table 1 – Al concentration and speciation of stock solutions.**

	Concentration ( $10^{-3}$ mol/L)	$\text{Al}_a$ (%)	$\text{Al}_b$ (%)	$\text{Al}_c$ (%)
$\text{Al}_0$	50.00	92.68	7.06	0.26
$\text{Al}_{13}$	21.62	13.10	83.35	3.55
$\text{Al}_{30}$	28.67	7.07	7.05	85.88

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