Factors effecting aluminum speciation in drinking water by laboratory research

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Abstract

Effects of aluminum on water distribution system and human health mainly attribute to its speciation in drinking water. Laboratory experiments were performed to investigate factors that may influence aluminum speciation in water supply system. The concentration of soluble aluminum and its transformation among other aluminum species were mainly controlled by kinetics processes of related reactions. Total aluminum concentration had a notable effect on the concentrations of mononuclear and soluble aluminum in the first 4 day; then its effect became weak. At pH above 7.50, both fluoride and orthophosphate had little effect on aluminum speciation; while, when the solution pH was below 7.50, the concentrations of mononuclear and soluble aluminum were proportional to the concentration of fluoride and inversely proportional to the concentration of orthophosphate. Both mononuclear and polynuclear silicic acids could complex with mononuclear aluminum by forming soluble aluminosilicates. In addition, the adding sequence of orthophosphate and aluminum into drinking water would also affect the distribution of aluminum species in the first 4 day. In order to minimize aluminum bioavailability in drinking water, it was suggested that orthophosphate should be added prior to coagulant process, and that the concentrations of fluoride and silicic acids should be controlled below 2.0 and 25 mg/L, respectively, prior to the treatment. The solution pH in coagulation and filtration processes should be controlled in the range of 6.50–7.50.

Key words: aluminum speciation; drinking water; fluorometric method; orthophosphate; silicic acid

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Introduction

With the development of modern industry, acid precipitation is becoming more and more common. This has led to a large amount of aluminum dissolving from soil to natural waters (Christopher and Carl, 1979; Gerhard et al., 2002), which may increase the concentration of residual aluminum in drinking waters. Moreover, aluminum salts are widely used as coagulants in drinking water treatment processes. Although they are effective for removing turbidity, Al-based coagulants may result in high Al concentration in treated water. Driscoll and Letterman (1988) reported that approximately 11% of the aluminum input remained in the treated water. Surveys of residual aluminum in drinking waters in China (Cui et al., 2002), the USA (Miller et al., 1984; Letterman and Driscoll, 1988), and Europe (Sollars et al., 1989) also showed that aluminum salts used in coagulation could increase residual aluminum concentrations notably.

High aluminum concentrations in drinking water distribution systems can cause high turbidity (Srinivasan et al., 1999; Kvech and Edwards, 2001), pipe wall deposition

(Fuge et al., 1992; Shea, 1993; Lauer and Lohman, 1994; Kriewall et al., 1996; Havics, 2001), and disinfection inhibition (Letterman and Driscoll, 1988). Health problems such as neurological disorders have also been associated with the presence of aluminum in drinking water. In order to control the content of residual aluminum in drinking water, various strategies have been proposed. Letterman and Driscoll (1988) found that low pH coagulation (6.50–7.00) combined with efficient filtration could reduce residual aluminum concentration. The presence of silicate salts could promote coagulation efficiency and reduce the residual aluminum concentration effectively (Snoeyink et al., 2003).

Most research works were aimed to control the concentration of total residual aluminum in drinking water. However, its adverse effects on drinking water distribution system and human health are always related to specific aluminum species. High concentrations of suspended aluminum (Sus-Al) might reduce disinfection effects and water transport capacity. Soluble aluminum (Sol-Al) might be highly cytotoxic to both plant and animal life, as it can be directly assimilated. Meanwhile, soluble aluminum in various forms also present different toxicities. Al^{3+} is known to be severely neurotoxic and was thus considered

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to be associated with the formation of neurofibrillary tangles and senile plaques in brain. However, when Al^{3+} reacts with other ions, its toxicity will be reduced. It showed that silicic acid interacted with aqueous Al^{3+} could significantly reduce the bioavailability of the latter by inhibiting the gastro-intestinal adsorption process (Birchall, 1994). Most organic compounded aluminum also showed weak biotoxicity. Polynuclear aluminum (Pol-Al) was less toxic to human body, as the dissociation process to simple species was slow. Accordingly, the evaluation of aluminum influence on drinking water distribution system and human health should consider its speciation.

As mentioned above, previous studies have drawn a conclusion on how water quality affected the concentration of residual aluminum in drinking water treatment processes. However, neither in drinking water treatment processes nor in its distribution systems, aluminum speciation has received much attention. In present work, laboratory experiments were conducted to study the effects of pH, temperature, and the concentrations of total aluminum (Tol-Al) and inorganic anions on aluminum speciation.

1 Materials and methods

Laboratory experiments were carried out to study aluminum speciation applying polyaluminium chloride (PAC) synthetic water over a period of 4 to 7 day. The reaction time was selected basing on the longest water retention time in drinking water distribution system. In most experiments, the concentration of Tol-Al was controlled at about 0.20 mg/L, the limit value of Tol-Al according to the "Standards for Drinking Water Quality" (GB 5749-2006 of China). Factors studied in the experiment include pH, temperature, Tol-Al, fluoride, orthophosphate, and silicate acid. The levels of each factor were selected basing on water quality standards and their typical concentrations in most drinking waters, as shown in Table 1.

It was found by Taylor et al. (1997) that silicic acid mainly existed in monomeric form when its concentration was lower than 120 mg/L, and mainly existed in polymeric form when its concentration was above 180 mg/L at pH below 9.0. Polynuclear silicic acid would transform into mononuclear silicic acid at low concentrations, while the process was much slow. In the experiments mononuclear silicic acid storage solution (50 mg/L) and polynuclear silicic acid storage solution (5000 mg/L) needed to be freshly prepared. In order to reduce their influence on

solution pH, both storage solutions were prepared with the same pH value as the synthetic water.

In the experiments, all solutions were stored in polyethylene bottles. Instruments, reagents, and distilled deionized water were stored at target temperatures for 24 hr prior to the experiments. Synthetic waters were prepared by adding 32.50 mL of 50.00 mg/L (calculated as Al) freshly prepared PAC solution and 16.00 mL of 1.00 mol/L $NaHCO_3$ storage solution to 8.0 L distilled deionized water, with the final aluminum concentration 0.2 mg/L and alkalinity about 100.00 mg/L (calculated as $CaCO_3$). Then equal volumes of the solution were added to 6 uniform 1.5 L reactors, and then inorganic ions were added to the reactors, as shown in Table 1. Finally, 0.10 mol/L NaOH and 0.10 mol/L HNO_3 were used to adjust solution pH. After preparation, synthetic waters were airproofed and restored at target temperatures. In order to maintain the solution pH, nitrogen gas protection was adopted in the experiments. Reagent grade chemicals were used except where noted. PAC, with an OH/Al molar ratio of 2.2, was provided by a local factory in powder form containing 30% Al_2O_3.

In order to determine the concentration of aluminum species, approximately 25–30 mL water sample, were collected from each reactor. One subsample was digested using reagent grade HNO_3 at pH 1.0 for 24 hr to analyze the concentration of Tol-Al. The other subsample was filtered through 0.22 μm polycarbonate filters. Then part of the filtrate was digested in HNO_3 with pH 1.0 for 24 hr to analyze the concentration of Sol-Al, and part of the filtrate was used to determine the concentration of Mon-Al. The concentration of Sus-Al and Pol-Al could be obtained by following Eqs. (1) and (2):

$$C_{Sus-Al} = C_{Tol-Al} - C_{Sol-Al} \tag{1}$$

$$C_{Pol-Al} = C_{Sol-Al} - C_{Mon-Al} \tag{2}$$

Aluminum concentration was determined by applying 8-HQ fluorometric method described by Lian et al. (2004). The minimum detection limit for the method ranged from 0.006 to 0.015 mg/L, where the accuracy (% recovery) was 91.7%–106.1%, the precision (relative standard deviation, $n = 6$) was 0.68%, and the linearity range was 0.027–0.27 mg/L. Aluminum species were identified using a spectrofluorometer (Model F-2500, Hitachi High-Technologies Corporation, Japan).

Table 1 Water quality and the levels selected in the experiments

Parameters	Selected levels					
	1	2	3	4	5	6
pH	6.50	7.00	7.50	8.00	8.50	9.00
Temperature (K)	278.15	283.15	288.15	293.15	298.15	–
Tol-Al (Al) (mg/L)	0.10	0.20	0.30	0.50	1.00	2.00
F (mg/L)	0.00	0.40	0.80	1.00	1.20	1.50
PO_4^{3-} (mg/L)	0.0	0.6	1.2	1.8	2.4	3.0
SiO_2 (mg/L)	0.0	5.0	10.0	15.0	20.0	25.0

2 Results

2.1 Effects of pH and temperature on aluminum speciation

At the same water temperature, the concentration of OH$^-$ increased with increasing pH value. Accordingly, more OH$^-$ would coordinate with aluminum at higher solution pH, leading to aluminum transformed among different species. Sus-Al was the major species at pH below 7.50 (Fig. 1a), while its proportion to Tol-Al decreased notably as solution pH increased from 6.50 to 9.00. At initial time, the concentrations of Sus-Al were 0.169, 0.117, and 0.046 mg/L at pH 6.50, 7.50, and 8.50, respectively, accounting for 74.4%, 51.5%, and 20.2% of the Tol-Al. At pH above 7.50, Sol-Al became the major species. At initial time, mononuclear aluminum (Mon-Al) concentration increased from 0.023 to 0.182 mg/L and Sol-Al concentration increased from 0.058 to 0.202 mg/L, as solution pH increased from 6.50 to 9.00. The concentration of Pol-Al varied little with solution pH, at about 0.02–0.04 mg/L, and on day 4 it decreased below 0.01 mg/L (Fig. 1b).

The variation of each aluminum species with reaction time was also affected by solution pH (Fig. 1c, d). At pH 8.00, the concentrations of Mon-Al and Sol-Al varied little with reaction time. However, at low pH (< 7.50), both the concentrations of Mon-Al and Sol-Al decreased with reaction time; and at high pH (> 8.00), they increased notably with reaction time. Generally, the pH values of

drinking water varied between 7.00 and 8.50. Based on those results, to our findings, aluminum species would be relatively stable in water distribution system considering the solution pH alone.

The effects of temperature on aluminum speciation were studied at pH 8.00 (Fig. 2). At beginning, as the second major species at temperatures below 278.15 K, the concentration of Sus-Al was 0.09 mg/L. However, when the water temperature increased to 298.15 K, its concentration gradually decreased to 0.03 mg/L. Meanwhile, the concentration of Sol-Al increased from 0.119 to 0.168 mg/L, and Mon-Al appeared a trend similar to Sol-Al. The concentration of Pol-Al varied little with water temperature at pH 8.00. The effects of temperature on aluminum speciation might be attributed to its effects on reaction constants. At constant solution pH, when water temperature increased from 278.15 to 298.15 K, the chemical equilibrium would be broken, which leaded to amorphous Al(OH)$_3$(s) transforming to Mon-Al (Van Benschotem et al., 1994). During the experiment, the concentrations of Mon-Al and Sol-Al varied little with reaction time increasing.

2.2 Effects of Tol-Al concentration on aluminum speciation

In the experiment, pH and water temperature were controlled at 7.50 and 293.15 K, respectively. The concentration of Sus-Al increased from 0.023 to 1.555 mg/L, with the increase in concentration of Tol-Al at initial
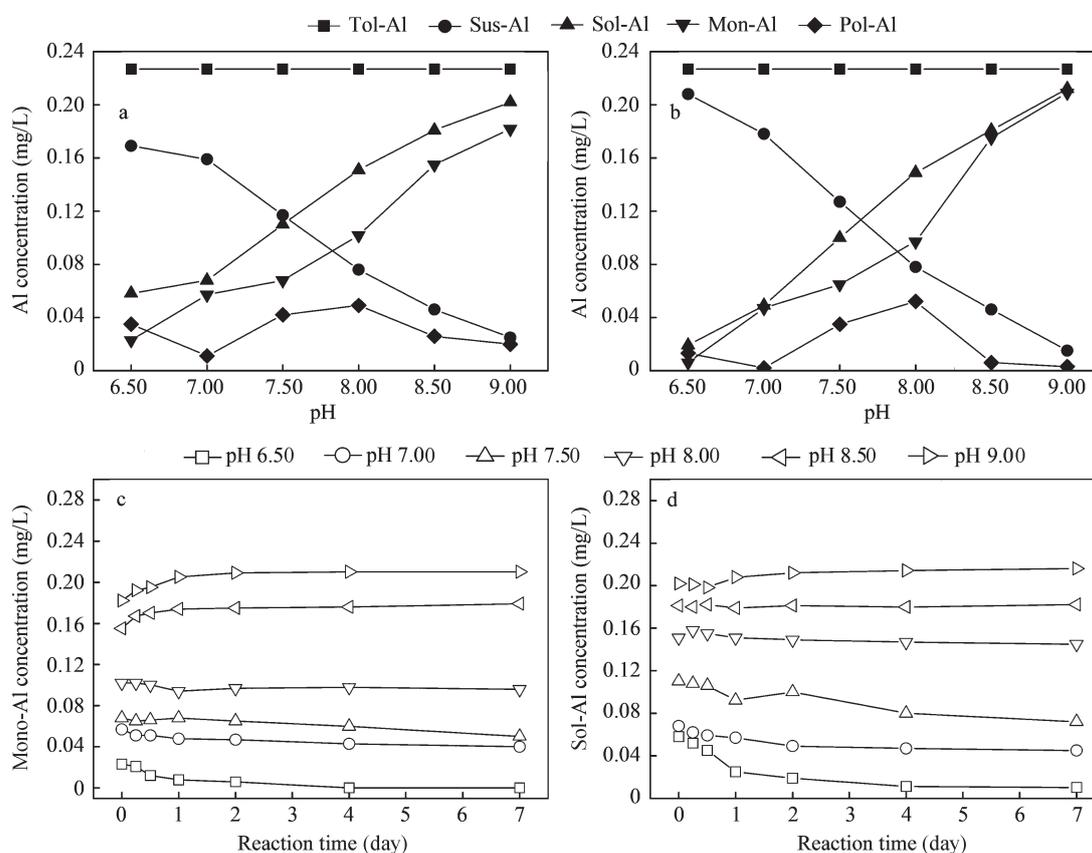

Fig. 1 Effects of solution pH on aluminum speciation at 293.15 K. (a) aluminum speciation variation at initial time; (b) aluminum speciation variation on day 4; (c) Mon-Al variation; (d) Sol-Al variation.
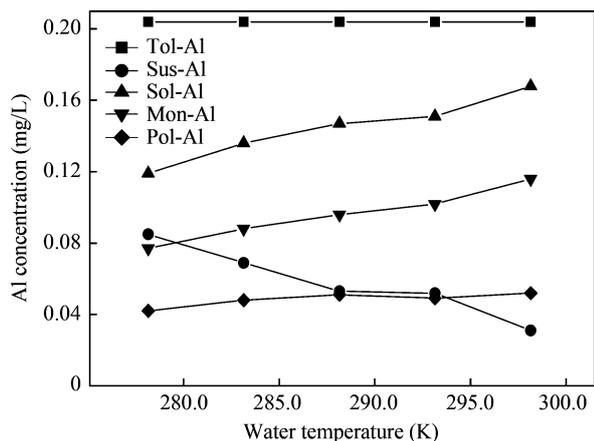
Fig. 2 Effects of water temperature on aluminum speciation at initial time.

time (Fig. 3a). The ratio of Sus-Al to Tol-Al increased significantly from 23.0% to 78.0%. Both Mon-Al and Pol-Al also showed increasing trends, while the increments were less than that of Sus-Al (Fig. 3b). When the concentration of Tol-Al increased from 0.1 to 2.00 mg/L, Mon-Al increased from 0.032 to 0.157 mg/L, and Sol-Al increased from 0.099 to 0.479 mg/L at initial time. However, the concentrations of Sol-Al, Mon-Al, and Pol-Al varied little on the 4th day. Mon-Al varied from 0.05 to 0.07 mg/L and Sol-Al varied from 0.08 to 0.16 mg/L in solutions with Tol-Al varied from 0.1 to 2.0 mg/L. It indicated that

these species were in a supersaturated state at initial time. As the reaction time increasing, both Mon-Al and Pol-Al transformed to Sus-Al gradually. On day 4, the reaction reached stabilization (Fig. 3c, d).

2.3 Effects of fluoride concentration on aluminum speciation

Fluoride exists universally in drinking water, and may compete with OH^- by forming stable aluminum fluoride complexes in moderately acidic conditions (Pommerenk and Schafran, 2005). In the experiments, aluminum species varied little with the concentration of fluoride increasing at pH 7.50 (Fig. 4d), indicating that fluoride could not compete with OH^- at moderately basic conditions, and existed mainly in ionic state (F^-).

Further studies were conducted at low pH levels (Fig. 4 a–c). It was found that the effects of fluoride became significant as solution pH decreasing. At pH 6.50, the concentration of Mon-Al increased from 0.023 to 0.106 mg/L as fluoride increased from 0 to 1.5 mg/L; and Sol-Al increased from 0.058 to 0.120 mg/L in the same condition. However, as solution pH increased from 6.50 to 7.00, the increment of aluminum concentration decreased notably with fluoride addition. Comparing experimental data with the thermodynamic calculation results (Fig. 4e), it could be concluded that the pH value, at which aluminum presented the minimum solubility, varied from 6.50 to 7.00 as the concentration of fluoride increasing. At pH 6.50,
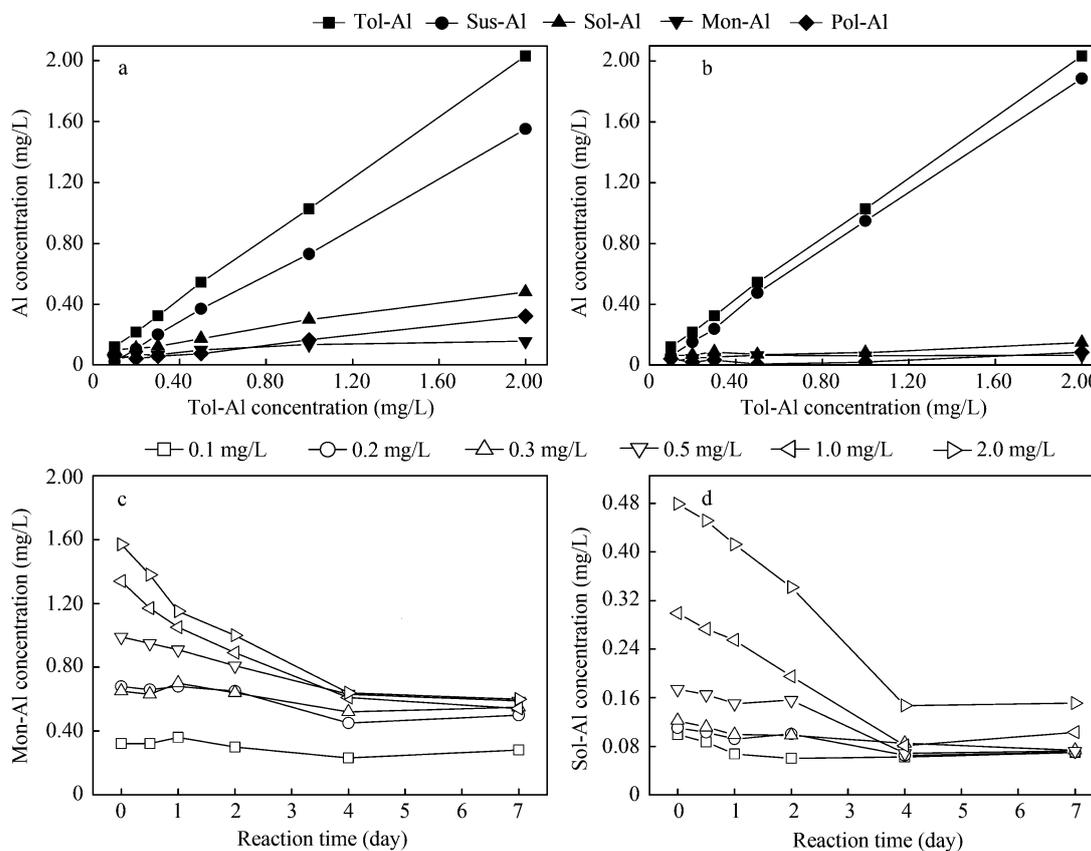

Fig. 3 Effects of Tol-Al concentration on aluminum speciation at 293.15 K. (a, b) aluminum speciation versus Tol-Al concentration at initial time (a) and on day 4 (b); (c) Mon-Al variation with reaction time at different Tol-Al concentrations; (d) Sol-Al variation with reaction time at different Tol-Al concentrations.
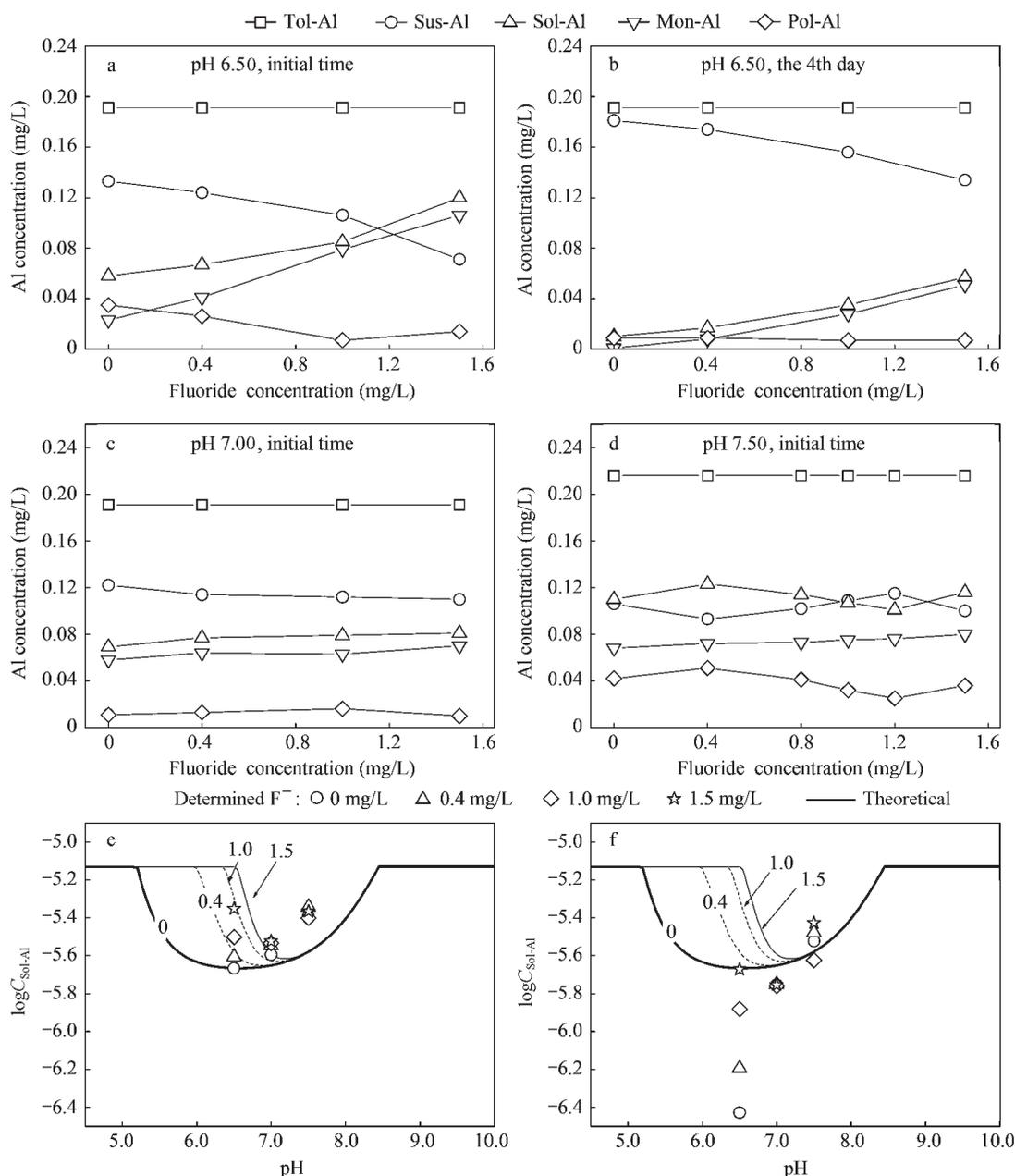
Fig. 4　Effects of fluoride concentration on aluminum speciation at 293.15 K. (a) at initial time and pH 6.50; (b) at day 4 and pH 6.50; (c) at initial time and pH 7.00; (d) at day 4 and pH 7.00; (e, f) comparison between determined data and theoretical value of Sol-Al at initial time (e) and on day 4 (f).

experimental data match well with the calculated value at initial time. However, as the reaction time increasing, the concentration of Sol-Al decreased below the theoretic value (Fig. 4f), indicating that the compounds formed between fluoride and aluminum were not stable at low pH and that solids with low solubility might be precipitated from solution. At initial time, the most of the fluoride existed as aluminum fluoride complexes, inducing the concentration of Sol-Al increased significantly. However, with prolonging reaction time, the formation of amorphous $Al(OH)_3(s)$ and other solids led to a decrease in free aluminum ions and provided more adsorption sites, which promoted the decomposition of aluminum fluoride complexes into Al^{3+} and F^- until chemical equilibriums were reached.

To confirm whether aluminum settlements were more

apt to form than aluminum fluoride complexes at pH 6.50–7.00, comparative experiments were conducted by adding PAC prior to fluoride during synthetic water preparation. It was found that aluminum speciation had little relation to fluoride concentration at pH 7.00. As the concentration of fluoride increasing, the increments of Mon-Al and Sol-Al were much smaller compared with the values obtained by adding fluoride prior to PAC at pH 6.50.

2.4 Effects of orthophosphate concentration on aluminum speciation

In general, orthophosphate salts were added to drinking water after filtration to control pipe-line corrosion. It was found that the effects of orthophosphate on aluminum species distribution varied at different solution pH values. At pH 6.50, when the concentration of orthophosphate

increased from 0 to 3.0 mg/L, the concentration of Sus-Al increased from 0.163 to 0.193 mg/L at initial time (Fig. 5a). Meanwhile, the concentration of Sol-Al decreased sharply from 0.035 to 0.0027 mg/L and the concentration of Mon-Al decreased from 0.01 to 0.004 mg/L, indicating that orthophosphate had a notable effect on aluminum species distribution at pH 6.50. While at pH 7.00 (Fig. 5c), the concentration of Sus-Al increased by 0.013 mg/L in solution with 3.00 mg/L orthophosphate, which is much less than the value of 0.03 mg/L at pH 6.50. Similarly, the concentrations of Sol-Al and Mon-Al also decreased less in the presence of orthophosphate at pH 7.00. When solution pH was above 7.5, orthophosphate had little

effect on aluminum species distribution (Fig. 5d), which consisted with the conclusion drawn by thermodynamic calculation (Fig. 5f). Therefore, under general drinking water pH range (7.00–8.50), the effect of orthophosphate on aluminum species distribution was not significant.

At pH values below 7.50, Mon-Al and Pol-Al transformed to Sus-Al as reaction time increasing. As shown in Fig. 5c, d, aluminum species distribution varied little in solutions with different amounts of orthophosphate on the 4th day, which indicated that the effects of orthophosphate on aluminum species distribution became weak as the reaction time increasing. It was known that orthophosphate could react with aluminum by forming $AlPO_4$ sediment, and its
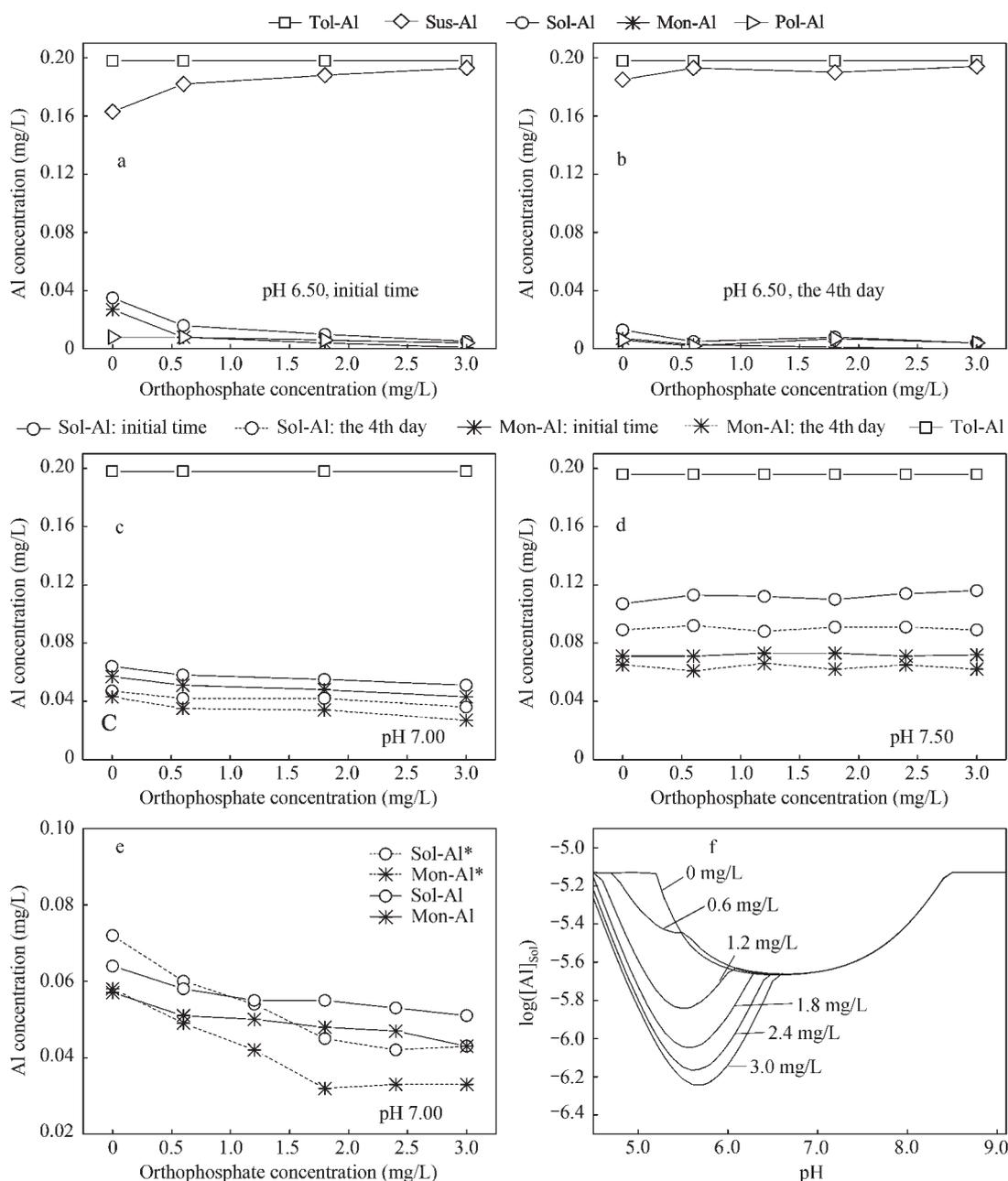

Fig. 5 Effects of orthophosphate concentration on aluminum speciation at 293.15 K. (a, b) aluminum speciation versus at initial time (a) and on day 4 (b) at 6.50; (c, d) Sol-Al and Mon-Al variation at initial time and pH 7.0 (c) and on day 4 and pH 7.50 (d); (e) Sol-Al and Mon-Al variation with orthophosphate concentration at pH 7.00. Sol-Al* and Mon-Al* represented the concentrations wherein orthophosphate added prior to PAC; (f) theoretical value of Sol-Al variation with solution pH at different concentrations of orthophosphate.

solubility varied with solution pH as shown in Fig. 5f. At pH below 6.80 the solubility of $AlPO_4$ was lower than that of amorphous $Al(OH)_3$ sediment, the final reaction product would be $AlPO_4$. Accordingly, the concentration of orthophosphate would control the concentration of Sol-Al in solution. However, at pH above 6.80, the solubility of $AlPO_4$ was higher than that of amorphous $Al(OH)_3$ sediment, the final reaction product would be amorphous $Al(OH)_3$ sediment. Thus, orthophosphate would not affect the final aluminum species distribution between pH 7.00 and 7.50.

2.5 Effects of silicic acid concentration on aluminum speciation

In natural waters, soluble silicic acids exist prevalently in the form of $Si(OH)_4$ (State Environmental Protection Administration of China, 1989). And polynuclear silicic acids are generally added to drinking water in the coagulation process. In the experiments, both kinds of silicic acids were studied at concentration range 0–25 mg/L (calculated as SiO_2); water temperature was controlled at 293.15 K.

Silicic acid, both in mononuclear and polynuclear forms, affected aluminum speciation at pH 7.50. At initial time, Mon-Al concentration increased from 0.068 to 0.140 mg/L and Sol-Al concentration increased from 0.109 mg/L to 0.172 mg/L, as mononuclear silicic acid increased from 0 to 25 mg/L (Fig. 6a). Meanwhile, the concentration of Sus-Al decreased from 0.116 to 0.053 mg/L. Compared with mononuclear silicic acid, polynuclear silicic acid could also increase the concentration of Sol-Al (Fig. 6d). However, Mon-Al decreased from 0.122 to 0.036 mg/L significantly, as polynuclear silicic acid increased from 0 to 25 mg/L.

As reaction time increasing, both Mon-Al and Sol-Al decreased slightly in solutions with mononuclear silicic acid, except at 25 mg/L (Fig. 6b, c). However, the concentration of Sol-Al increased slightly in solution with polynuclear silicic acid above 15 mg/L (Fig. 6f). It could be concluded that polynuclear silicic acid was more effective than mononuclear silicic acid in combining Mon-Al by forming soluble complex compounds (Taylor et al., 1997). The binding ability between analytical reagent and Mon-Al was weaker than that between polynuclear silicic acid and Mon-Al, leading to Mon-Al that had been combined by polynuclear silicic acid could not be determined accurately. The determined value of Mon-Al concentration represented the part that had not been combined by polynuclear silicic acid. In fact, the increment of Pol-Al represents the part of Mon-Al that had been combined by polynuclear silicic.

3 Discussion

Drinking water treated by PAC contained large proportions of suspended, colloidal, and polynuclear aluminum, especially at pH below 8.00 (Berkowitz et al., 2005). As reaction time increasing, aluminum speciation varied little at pH 7.50–8.00, which might be connected with the character of aluminum speciation in drinking water. The

suspended and colloidal aluminum, which were treated as Sus-Al in the experiments, are particles and colloids preformed in coagulant preparation. Their dissolution rates were much slower than those of amorphous aluminum sediments in the research period. Accordingly, sand filtration followed by activated carbon adsorption or micro-filtration would remove most of the Sus-Al and keep Tol-Al at a low level in drinking water. At pH 6.5–7.5, Sus-Al was the major species; and as reaction time increasing, both Mon-Al and Pol-Al would transform to Sus-Al gradually. Therefore, maintaining low pH values during drinking water coagulation process would be a potential method to enhance water quality and reduce the concentration of Sol-Al.

In our survey of a northeastern Chinese city (sample number 117), the average drinking water pH was 7.00. Tol-Al, Sus-Al, Sol-Al, and Mon-Al average concentrations were 0.170, 0.123, 0.047, and 0.030 mg/L, respectively. Sus-Al was the major species, accounting for 72.3% of Tol-Al. Therefore, maintaining a neutral environmental solution pH would reduce the concentrations of Sol-Al and Mon-Al. However, in the drinking water distribution process, solution pH was generally controlled at about 8.00 or higher to protect against pipe-line corrosion. In this condition, aluminum existed primarily in Mon-Al form. In order to reduce its toxicity to human body, the introduction of silicic acids was suggested. By forming soluble aluminosilicate salts, aluminum toxicity could be notably reduced. However, orthophosphate salts had little aids in reducing Mon-Al concentration at pH above 7.0. In addition, controlling the concentration of Tol-Al in drinking water could also reduce its toxicity.

Both fluoride and orthophosphate had significant effect on aluminum speciation at low pH values (Figs. 4 and 5). High fluoride concentration in source water would increase the concentration of Mon-Al significantly in treated water, when it was treated by aluminum-based coagulants. Adding orthophosphate salts to drinking water would be helpful for Mon-Al and Sol-Al control in treated water. Meanwhile, the adding sequence between aluminum and orthophosphate could also influence the concentrations of Mon-Al and Sol-Al at low pH (6.50–7.00), as shown in Fig. 5e. Aluminum was generally added to source water as coagulant prior to orthophosphate salts in water treatment processes. Both Sol-Al and Mon-Al decreased much more significantly when orthophosphate salt was added before coagulation than that in the opposite sequence. Accordingly, reducing fluoride concentration and introducing orthophosphate salts before coagulation would be helpful in reducing the total residual aluminum concentration in drinking water.

Because silicic acid would increase the concentrations of Sol-Al and Mon-Al notably, it would be helpful to apply other kinds of coagulant aids to substitute for activated silicic acid in the coagulant process. However, reports have shown that the existence of mononuclear and polynuclear silicic acids could significantly reduce aluminum bioavailability by forming soluble coordination compounds. The newly formed soluble aluminosilicate salts were not easily
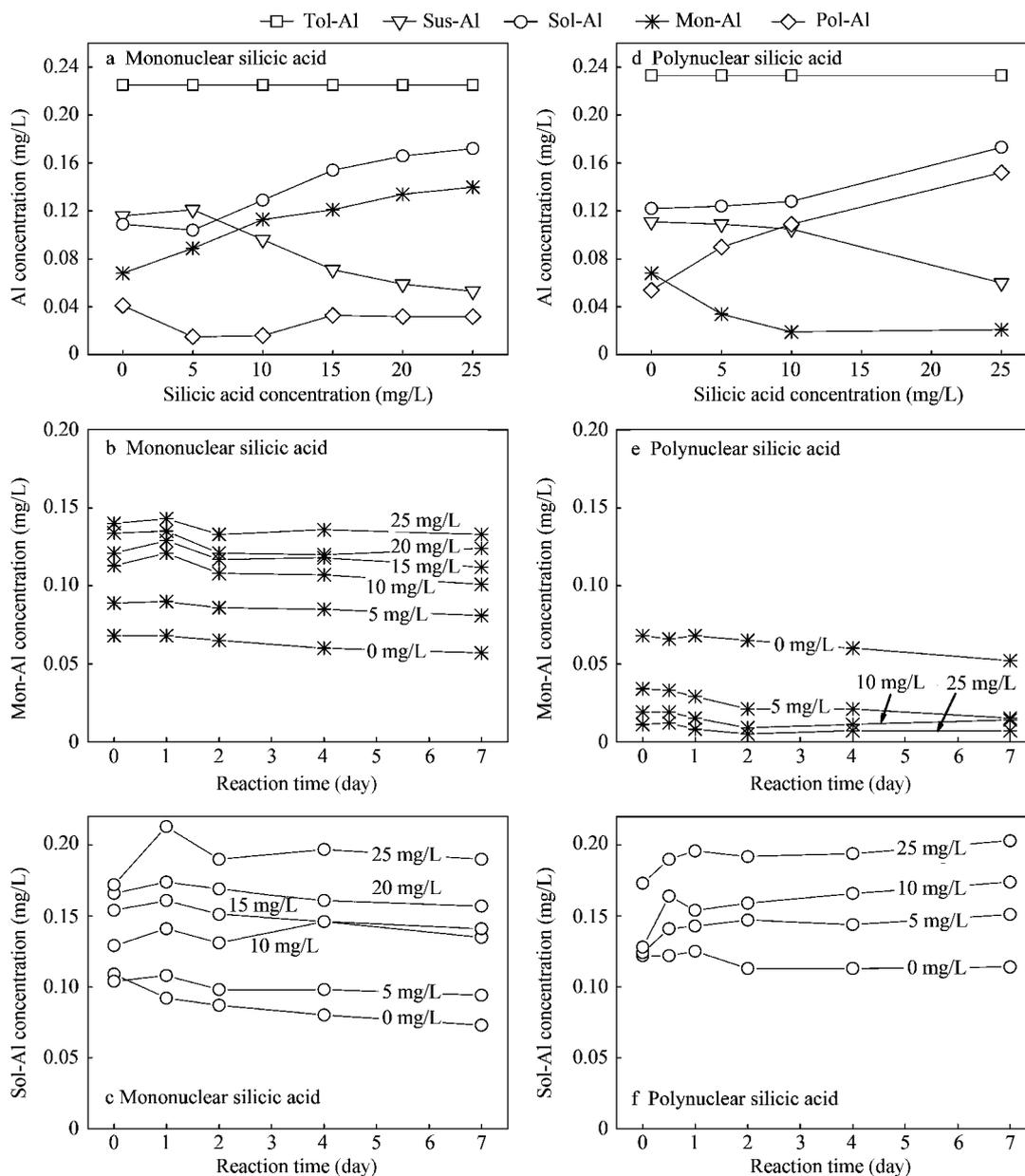
Fig. 6 Effects of silicic acid concentration on aluminum speciation at 293.15 K. (a, d) aluminum species distribution; (b, e) Mon-Al concentration variation; (c, f) Sol-Al concentration variation.

adsorbed by the gastro-intestinal system.

4 Conclusions

Both Mon-Al and Sol-Al are proportional to solution pH range 6.50–9.00. Low coagulation and filtration pH values (6.50–7.50) are recommended to improve treated water quality and decrease the concentrations of Mon-Al and Sol-Al. Meanwhile, at pH 7.50–8.00, aluminum speciation varies little as reaction time increase. The effects of water temperature on aluminum speciation are mainly dependent on its effects on the reaction constants. Low water temperature would be helpful in controlling the content of Mon-Al.

Tol-Al has significant influence on the concentrations of Mon-Al and Sol-Al at initial time; while as the reaction time increase, the effects of Tol-Al become weak generally.

The stability time is about 4 days, which is not favorable to Mon-Al and Sol-Al control since the water retention time in most distribution systems is less than 4 days. Controlling the concentration of Tol-Al will be the most direct method to reduce Mon-Al and Sol-Al concentrations in drinking water.

At pH > 7.50, fluoride has little influence on aluminum speciation. While at pH < 7.50, both Sol-Al and Mon-Al are positively correlated with fluoride concentration. However, the existence of fluoride will not prevent the transformation process from Sol-Al to Sus-Al as the reaction time increasing. In order to reduce the concentrations of Mon-Al and Sol-Al in treated water, fluoride concentration is suggested being controlled below 2.0 mg/L. Similar to fluoride, orthophosphate has little influence on aluminum speciation at pH above 7.50. At pH below 6.80, orthophosphate can effectively reduce Mon-Al concen-

tration by forming aluminum orthophosphate settlement. Meanwhile, the adding sequence of orthophosphate and PAC in drinking water treatment will also affect aluminum speciation; adding orthophosphate prior to PAC is recommended.

Silicic acid could react with Mon-Al by forming soluble aluminosilicate, inducing a significant increase in Mon-Al and Sol-Al concentration. The bonding ability between polynuclear silicic acid and Mon-Al is stronger than that between mononuclear silicic acid and Mon-Al. Consequently, applying other kinds of coagulant aids to substitute for activated silicic acid would be helpful in Mon-Al and Sol-Al control.

Acknowledgments

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