Effects of advanced oxidation pretreatment on residual aluminum control in high humic acid water purification

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Abstract

Due to the formation of disinfection by-products and high concentrations of Al residue in drinking water purification, humic substances are a major component of organic matter in natural waters and have therefore received a great deal of attention in recent years. We investigated the effects of advanced oxidation pretreatment methods usually applied for removing dissolved organic matters on residual Al control. Results showed that the presence of humic acid increased residual Al concentration notably. With 15 mg/L of humic acid in raw water, the concentrations of soluble aluminum and total aluminum in the treated water were close to the quantity of Al addition. After increasing coagulant dosage from 12 to 120 mg/L, the total-Al in the treated water was controlled to below 0.2 mg/L. Purification systems with ozonation, chlorination, or potassium permanganate oxidation pretreatment units had little effects on residual Al control; while UV radiation decreased Al concentration notably. Combined with ozonation, the effects of UV radiation were enhanced. Optimal dosages were 0.5 mg O_3/mg C and 3 hr for raw water with 15 mg/L of humic acid. Under UV light radiation, the combined forces or bonds that existed among humic acid molecules were destroyed; adsorption sites increased positively with radiation time, which promoted adsorption of humic acid onto polymeric aluminum and $Al(OH)_3(s)$. This work provides a new solution for humic acid coagulation and residual Al control for raw water with humic acid purification.

Key words: advanced oxidation pretreatment; drinking water; humic acid; residual aluminum; water purification

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Introduction

Aluminum salts are widely used as coagulating agents in drinking water treatment. Although they are effective in removing turbidity, aluminum-based coagulants may result in a high concentration of Al residue. Driscoll et al. (1987) reported that about 11% of the Al input remains in treated water, while a survey of 380 plants found that the concentration of total aluminum ranges from 0.003 to 1.6 mg/L (Kopp, 1969). Surveys conducted in China (Cui et al., 2002), the United States (Miller et al., 1984; Letterman and Driscoll, 1988), and Europe (Sollars et al., 1989) also showed that the Al in drinking water coagulated by aluminum salts is notably high, especially in water with soluble organic matter.

The occurrence of Al in treated water is an undesirable consequence of treatment practice (Driscoll et al., 1987; Van Benschoten and Edzwald, 1990) and may result in a variety of water supply problems. Aluminum flocs and sediments interfere with the disinfection effects by en-

meshing and protecting micro-organisms (Hoff, 1978). The deposition of aluminum in the pipeline decreases water carrying capacity, and even thin aluminum coating result in a significant pressure drop (Hudson, 1966). Aluminum may play an active role in the pathogenesis of critical neuropathologic lesions in Alzheimer's disease and other related disorders (Exley, 2005; Perl and Moalem, 2006; Walton, 2006). A survey of 88 districts in England and Wales, found that the rate of Alzheimer's disease was 1.5 times higher in districts where the mean Al concentration exceeded 0.11 mg/L, than in districts where Al was less than 0.01 mg/L (Martyn et al., 1989).

Effectiveness of general drinking water treatment units available for Al removal has been previously summarized (Srinivasan et al., 1999). Processes such as coagulation, sedimentation and filtration as well as lime softening are moderately effective for Al control. Disinfection and anion ion exchange units are, however, ineffective for Al control. By maintaining low coagulating pH (6.50–7.00) and effective sand filtration, residual Al can be controlled below 0.1 mg/L (Letterman and Driscoll, 1988). However,

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this is not efficient for raw water with soluble organic matter, especially humic acid, as most Al exists in organic combined form (Jelel, 1986; Van Benschoten and Edzwald, 1990; Browne and Discoll, 1993; Weng et al., 2002). Thus, effective Al control methods are still not available for high humic acid water purification.

Advanced oxidation pretreatment (AOP) is considered to be one of the best available technologies for the removal of color, tastes, and mineral compounds (Gunten, 2003; Hijnen et al., 2006). However, the application of AOP will damage or change the structure of humic acid and thus affect its combination with Al. The goal of the present study was to investigate the effects of AOP including ozonation, UV radiation, chlorination, and potassium permanganate oxidation on residual Al control, and to optimize the treatment process for raw water with high humic acid concentration. Experimental results will provide a new solution for residue Al control in drinking water treatment processes.

1 Materials and methods

1.1 Raw water

All experiments were conducted in the lab. Experimental water was prepared from the civil drinking water of Xi'an, China. Total organic carbon (TOC) in the raw water was adjusted by adding humic acid (Fluka humic acid, Sigma-Aldrich, Japan) stock solution, which was prepared by dissolving humic acid in basic condition and filtering it through a 0.45 μm polycarbonate membrane. The filtrate was analyzed by a TOC analyzer (Dohrmann Model DC 80 USA), and stored at 4°C. Turbidity was adjusted by kaolin (BSF ASP170, USA) suspended solution after 24 hr immersion. Solution pH was adjusted by the addition of 0.10 mol/L of NaOH or 0.10 mg/L of HNO$_3$.

Besides TOC and turbidity, other water quality parameters such as pH, water temperature, alkalinity, fluoride, silicic acid, and orthophosphate can affect coagulation and filtration efficiency, thus leading to variation of Al

in treated water (Browne and Discoll, 1993; Weng et al., 2002; Wang et al., 2010). To observe the effects of AOP on residual Al control, the water quality parameters were selected according to general values in the drinking water, as shown in Table 1.

1.2 Advanced oxdation pretreatment

A glass reactor was employed for ozonation (Fig. 1). Ozone was supplied by an ozone generator (Model SG-01A, Sumitomo, Japan) and was introduced at the bottom of the reactor through a bubble diffuser. The solution was mixed by a magnetic stirrer. Treated water was withdrawn from the reactor at different ozone dosages for the following treatment processes.

UV radiation was conducted in a stainless-steel batch reactor (Fig. 1). A 450-W high-pressure mercury-vapor lamp was inserted into the hollow quartz located at the center of the reactor. A water-cooling loop was used to prevent the lamp from overheating and to maintain the water at room temperature. Three liters of aqueous solution was added into the reactor. Two aliquots of 20 mL were used to measure initial humic acid concentration. Treated water of 500 mL was withdrawn from the reactor at various time intervals.

The raw water was chlorinated with hypochlorite solution in a glass reactor, and mixed by a magnetic stirrer. The reaction was completed by Na$_2$S$_2$O$_3$.

The raw water was oxidized with potassium permanganate in a glass reactor, and mixed by a magnetic stirrer.

Table 1   Quality of the raw water used in the experiment.

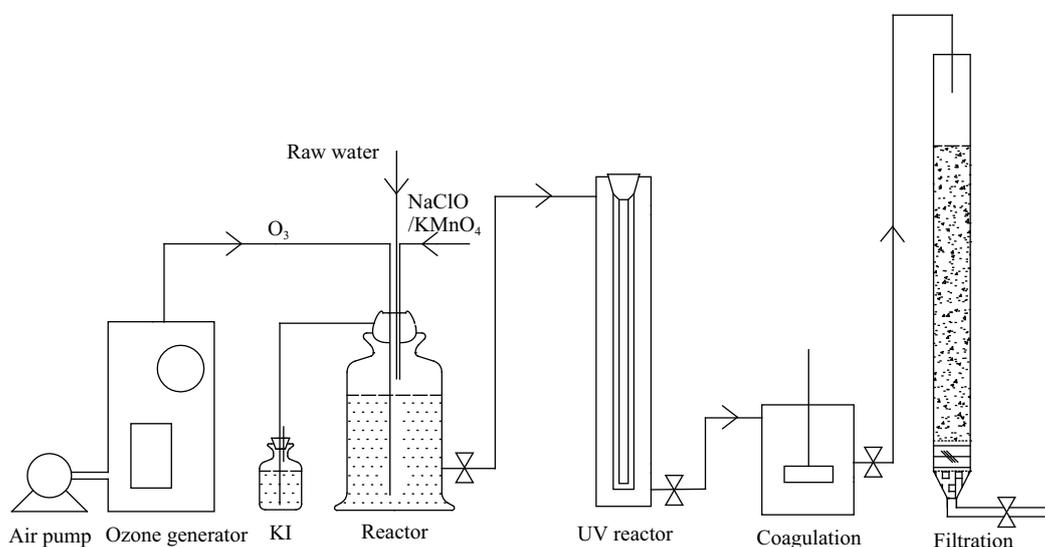| Parameter	Description	Parameters	Description
Temperature	20.0°C	Soluble-Al	0.111 mg/L
pH	7.50	Silicic acid	4.41 mg/L
Turbidity	5.0 NTU	Phosphate (as P)	ND
Alkalinity (as CaCO$_3$)	98.7 mg/L	Fluoride (as F)	0.36 mg/L
TOC	15.0 mg/L	Calcium (as Ca)	20–25 mg/L
Soluble TOC	14.0 mg/L	Magnesium (as Mg)	8–12 mg/L
Total Al	0.130 mg/L		

TOC: total organic carbon.

Fig. 1   Flow chart of the raw water purification with advanced oxidation pretreatment.

1.3 Coagulation and filtration

Raw water was coagulated on a program-controlled JJ-4A jar tester with six beakers (Guo-Hua Electrics Co., China). Certain amounts of polyaluminum chloride (PACl) were added to the solution. The coagulation procedure involved rapid mixing at 100 r/min for 2 min, followed by slow stirring at 30 r/min for 30 min, and a settling period of 1 hr. Samples were taken from the surface water.

Coagulant effluent water was then filtered by a sand column with 100 mm in inner diameter and 2500 mm in height (Fig. 1). The bottom section was supported by a gravel layer 200 mm in depth. Silica sand (with grain size 0.5–1.2 mm, depth 700 mm, and porosity 43%) was used as a single layer media. Filtration rate was adjusted by a control value, at 8–10 m/hr.

1.4 Analytical methods

Aluminum concentration was determined by Chrome Azurol S Spectrophotometry (GB/T5750.6-2006), applying a spectrophotometer (UV-1650PC, Shimadzu, Japan). One subsample was digested in pH 1.0 with reagent grade HNO_3 for 24 hr to analyze the concentration of total-Al. The other subsample was filtered through a 0.45-μm polycarbonate filter, with the filtrate then digested with HNO_3 to analyze the concentration of soluble-Al. The minimum detection limit was 0.20 μg/L, accuracies (% recovery) were 94%–106%, precision (relative standard deviation, $n = 6$) was less than 5%, and linearity ranges were 0.005–0.25 mg/L.

Other water quality parameters, such as pH determined by a pH meter (Model 828, Thermo Electron Corporation, USA), turbidity determined by a turbidity meter (Model PCCHECKIT, Lovibond), and conductivity determined by an electrical conductivity meter (Model DDS-320, Da-Pu Equipment Corporation, USA) were analyzed according to GB/T 5750.4-2006. Orthophosphate was determined by molybdenum-antimony anti-spectrophotometry according to GB/T 5750.5-2006. Silicic acid concentration was determined by Si-Mo spectrophotometry. The concentrations of calcium and magnesium were determined by applying an IRIS intrepid-II ICP meter (Thermo Elemental, USA).

To observe the structure of humic acid before and after oxidiation, a three dimension fluorescence spectrum was recorded with a fluorescence spectrometer (JASCO FP-6500, Japan). The excitation and emission wavelengths were 270–470 nm and 380–580 nm, with a slit band-width of 5 and 2 nm, respectively. Experimental data were processed with SURFER computer software (Golden Software, USA). Average particle size and zeta potential of humic acid colloids were determined by a zeta potential meter (MacrotechNichion ZC-2000, Japan).

2 Results

2.1 Effects of enhanced coagulation on residual Al control

Traditional coagulation, sedimentation, and filtration treatment processes were effective in residual Al control for low humic acid water purification. Without humic acid, both soluble-Al and total-Al were maintained below 0.2 mg/L (Fig. 2). The presence of humic acid increased Al concentration notably. When the concentration of humic acid was 5.0 mg/L, soluble-Al in treated water was 0.170 mg/L. When humic acid was increased from 5 to 10 mg/L, soluble-Al concentration increased to 0.82 mg/L and total-Al was 1.3 mg/L, close to the quantity of Al addition. When humic acid was increased to 15 mg/L, the concentrations of soluble-Al and total-Al were 1.8 and 2.1 mg/L, respectively. Most Al existed in the supernatant, and was difficult to remove by filtration.

To ensure treated water quality, enhanced coagulation is usually adopted in organic polluted water purification. We fixed humic acid at 15 mg/L to determine the effects of coagulant dosage on residual Al concentration, as shown in Fig. 3. When coagulant dosage was less than 20 mg/L (as PACl), the dosage used in traditional drinking water treatment plants, residual Al concentration increased positively with coagulant dosage, and most of the Al remained in the treated water. When the coagulant dosage was above 20 mg/L, aluminum concentration was controlled gradually. The critical value was higher for water with higher humic acid concentration. When the coagulant dosage was increased from 20 to 120 mg/L, total-Al in the treated water decreased to 0.19 mg/L, meeting the drinking water quality requirements of China.

With 140 mg/L PACl addition, soluble-Al and total-Al in treated water were 0.034 and 0.033 mg/L respectively, much lower than those in water coagulated with 120 mg/L PACl. When the coagulant dosage was increased to more than 150 mg/L, residual Al in the treated water stabilized at 0.01–0.03 mg/L, indicating that not only the Al added in coagulation, but also the Al in the raw water was removed. However, the dosage used in enhanced coagulation was about 10 times higher than that used in traditional coagulation.

2.2 Effects of advanced oxidation on residual Al control

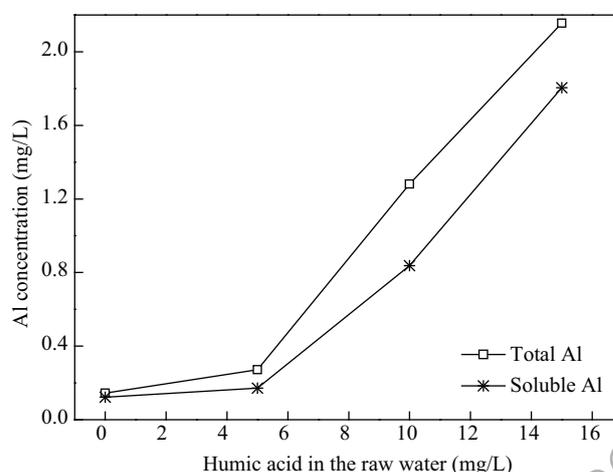Raw water was pretreated with advanced oxidation methods, including UV radiation, ozonation, chlorination,

Fig. 2 Effects of humic acid on residual Al concentration in traditional drinking water treatment system.
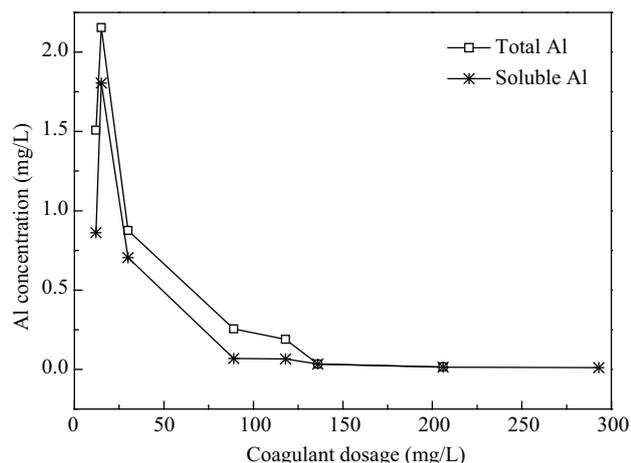
Fig. 3 Effects of enhanced coagulation on residual Al concentration, with 15 mg/L humic acid in the raw water.

or potassium permanganate oxidation. The effluent was then coagulated with 12 mg/L of PACl and filtrated. Results showed that Al residue in the treated water varied notably in systems applying different oxidation methods (Fig. 4). Ozonation, chlorination, and potassium permanganate oxidation pretreatment had little effect on residual Al control. Although part of the soluble-Al was removed, the concentrations of total-Al were still high (about 1.9 mg/L) and close to the value obtained without pretreatment.

Being different from others, systems with UV radiation decreased residual Al concentration notably. After 3 hr radiation, soluble-Al and total-Al in the treated water were 0.16 and 0.24 mg/L respectively, much lower than those in water without pretreatment. Adjusting radiation time, we observed variations of soluble-Al and total-Al in the treated water (Fig. 5a). Both soluble-Al and total-Al decreased notably with radiation time. To maintain total-Al in the filtered water below 0.2 mg/L, radiation time required was above 3 hr.

As shown in Fig. 5b ozonation had positive effects on Al control. However, its dosage when used alone as pretreatment was much higher. When ozone dosage was
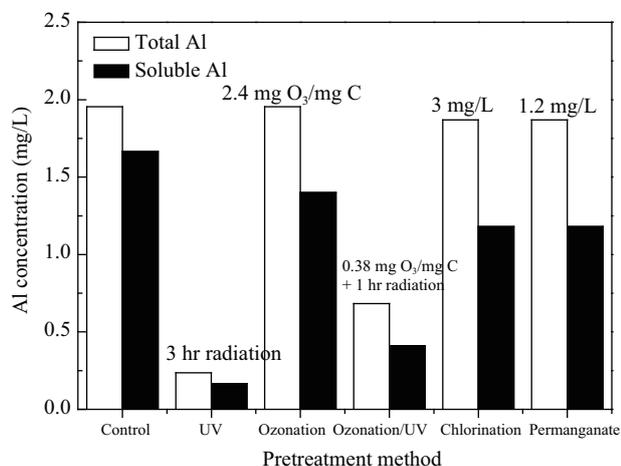

Fig. 4 Effects of oxidation on residual Al concentration in the treated water, with 15 mg/L humic acid in the raw water and PACl dosage of 12 mg/L.

less than 2.4 mg O_3/mg C, total-Al in the treated water varied little (1.9 mg/L); when dosage was increased to 9.0 mg O_3/mg C, 30 times that of the engineering dosage, total-Al concentration decreased to 0.74 mg/L, which was still higher than 0.2 mg/L (Fig. 5b).

2.3 Residual Al control applying O_3 and UV combined pretreatment

The combination of UV radiation with ozonation also decreased residual Al concentration. With 1 hr UV radiation and 0.38 mg O_3/mg C ozonation, soluble-Al and total-Al in the treated water were 0.41 and 0.68 mg/L (Fig. 6a), respectively, much lower than separate pretreatment with 1 hr radiation or 0.38 mg O_3/mg C ozonation (Fig. 5).

When radiation time was increased from 1 to 3 hr, soluble-Al and total-Al concentrations were 0.06 and 0.07 mg/L, much lower than the values in the raw water, which indicated that both the Al salts added in coagulation and the Al in the raw water were removed.

Accordingly, the combination of ozonation and UV radiation was recommended for high humic acid natural water pretreatment.

When radiation time was less than 1 hr, residual Al concentration decreased notably; when radiation time was above 1.0 hr, the decreasing ratio of residual Al concentration became much slowed. Fixing UV radiation time at 1 hr, the effects of ozone dosage on residual Al concentration were investigated (Fig. 6b). When the ozone dosage was 0.5 mg O_3/mg C, soluble-Al and total-Al concentrations in the treated water were 0.40 and 0.55 mg/L, respectively. When the ozone dosage was increased to 2.4 mg O_3/mg C, total-Al removal ratio increased from 72% to 85%, indicating that when ozone concentration was above 0.5 mg O_3/mg C, it had little effect on residual Al control.

Based on single factor experimental results, the optimal conditions for ozonation/UV method were ozone dosage 0.5 mg O_3/mg C and 3 hr radiation for raw water with 15 mg/L humic acid, however, with humic acid variation, the optimal dosages may differ. The differences in humic acid characteristics and water quality in the raw water also effected optimal oxidation conditions. Accordingly, the optimal ozone dosage and radiation time should be determined by pilot tests.

3 Discussion

By providing an excess of metal hydroxide precipitate surfaces, most dissolved organic matter can be removed through enhanced coagulation. However, coagulant dosage was usually high, especially for raw water with high content of soluble humic acid. With the application of UV radiation, coagulant efficiency was enhanced. Both humic acid and Al in the treated water were controlled at an acceptable coagulant dosage. Considering the mineralization effect of UV radiation on humic acid, samples with the same humic acid concentration were coagulated before and after UV radiation. We found coagulation efficiency was much higher for water after pretreatment.
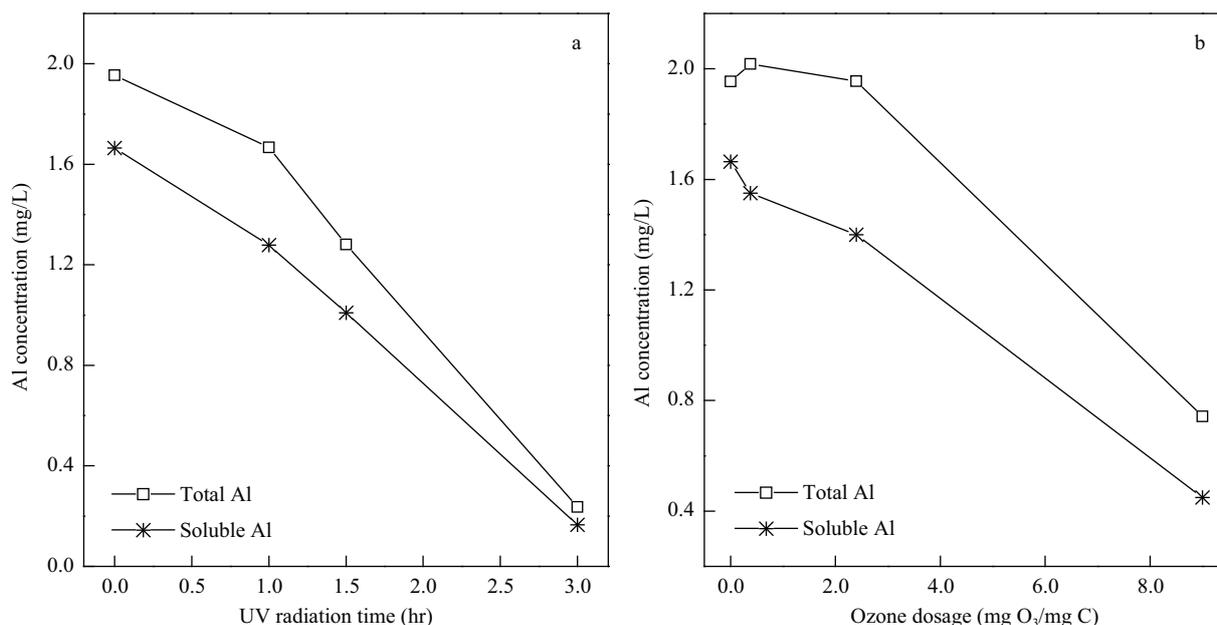
Fig. 5 Residual Al concentration in systems pretreated with UV radiation (a) and ozonation (b) separately, with 15 mg/L humic acid in the raw water and 12 mg/L PACl addition in coagulation.
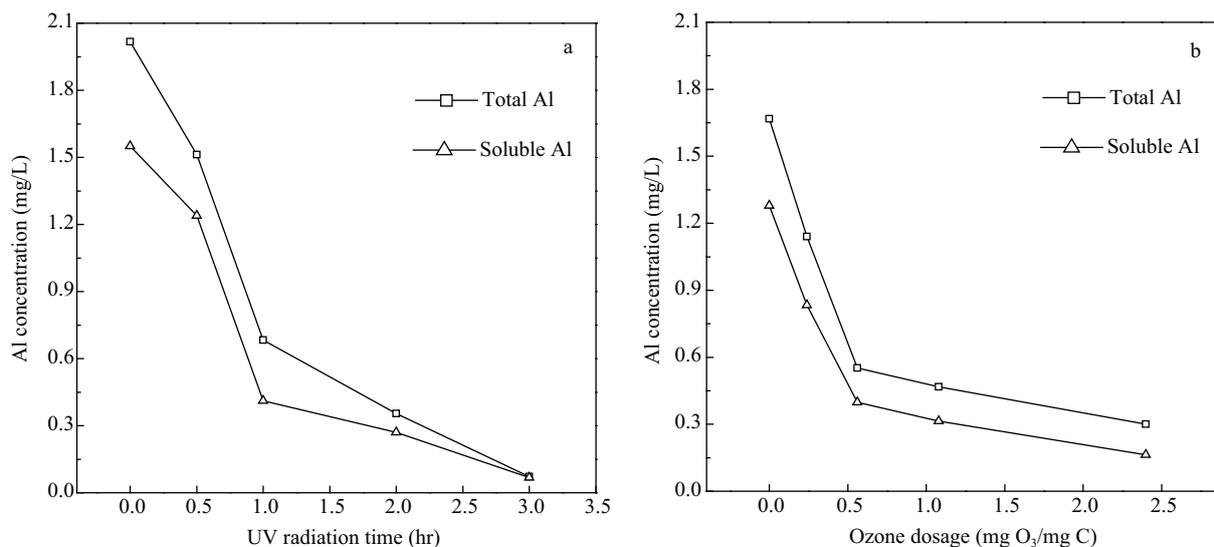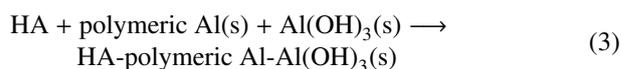

Fig. 6 Effects of ozonation and UV combined pretreatment on residual Al concentration. (a) Aluminum concentration varied with UV radiation time at 0.38 mg O_3/mg C ozone dosage; (b) aluminum concentration varied with ozone dosage at 1 hr UV radiation.

In water treatment plants where raw water is coagulated with PACl, Al concentration in the treated water is positively correlated with coagulation efficiency, and humic acid is removed either by complexation with the coagulant or by precipitation after being adsorbed. Under neutral and basic conditions, the hydrolysis of PACl is enhanced, and thus the chance to form $Al(OH)_3(s)$ increases. Simultaneously, the opportunity for humic acid to contact with the free Al-salt decreased, so that the humic acid is mainly removed by adsorption onto polymeric aluminum and $Al(OH)_3(s)$ (Bose and David, 2007). Conversely, the slow hydrolysis of the pre-hydrolyzed PACl can neutralize the charge of humic acid before the formation of $Al(OH)_3(s)$, and form the humic acid-polymeric-$Al(OH)_3(s)$ which can be removed through adsorption. The reactions can be described by Eqs. (1)–(3).

$$HA + \text{polymeric Al} \longrightarrow \text{HA-polymeric Al(s)} \quad (1)$$

$$HA + Al(OH)_3(s) \longrightarrow HA\text{-}Al(OH)_3(s) \quad (2)$$

$$HA + \text{polymeric Al(s)} + Al(OH)_3(s) \longrightarrow \\ \text{HA-polymeric Al-}Al(OH)_3(s) \quad (3)$$
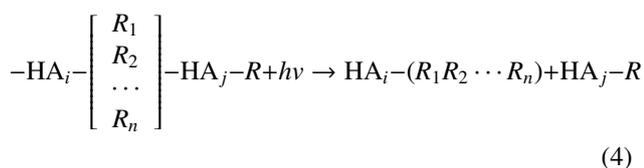
HA: humic acid.

In oxidation, the structure of humic acid would be destroyed, leading to a notable decrease of its average molecular weight. Ozonation is effective for the treatment of waters containing aliphatic and aromatic compounds (Staehelin and Hoignle, 1985; Camel and Bermond, 1998). The hydroxyl radicals attack organic compounds relatively non-selectively, oxidizing them by hydrogen atom

abstraction or by addition to double bonds, leading to a notable increase in hydrophilization, which is not good for humic acid coagulation and residual Al control (Gül, 2002; Sanly et al., 2006). As hypochlorous and potassium permanganate oxidation have little effect on the structure of humic acid in neutral and basic conditions, neither can improve coagulation efficiency.

Different from ozonation, UV radiation improved humic acid solution coagulating efficiency notably. This might be connected with the effects of UV radiation on humic acid structure. The species, number, and position of the functional groups on organic matter all had significant effects on humic acid coagulation efficiency (Becker and O'Melia, 1996). Experimental results showed that both the fluorescence fingerprints and zeta potential of the humic acid varied little in solution with the same TOC value, before and after UV radiation. However, the average particle size of humic acid colloid decreased notably with increasing radiation time. Based on above experimental results, humic acid structure was considered to be changed as shown in Eq. (4).

$$-\mathrm{HA}_i-\begin{bmatrix} R_1 \\ R_2 \\ \cdots \\ R_n \end{bmatrix}-\mathrm{HA}_j-R+h\nu \rightarrow \mathrm{HA}_i-(R_1R_2\cdots R_n)+\mathrm{HA}_j-R \tag{4}$$

Humic acid generally existed in aggregation formed by large amounts of humic acid molecules. These molecules connected with each other by hydrogen bonds, chemical bonds such as –O– and –NH–, and forces existing among humic acids (Sanly et al., 2006). Under the radiation of UV light, reactions mainly occurred on these connecting bonds. As the large molecule aggregation colloids broke into small ones, new adsorption sites were produced, thus enhancing the adsorption of humic acid onto polymeric aluminum and $Al(OH)_3(s)$. With UV radiation time increasing, more reactive sites formed, and the solution destabilized more easily. However, as direct evidence was not found for the selective destruction characteristics of UV radiation and the O_3/UV combined oxidation routes was not clear, the reaction mechanism of humic acid under UV light radiation requires further study.

4 Conclusions

(1) Traditional coagulation, sedimentation, and filtration treatment processes were effective in Al control for the raw water without humic acid; when the concentration of humic acid increases, the concentration of Al in treated water increased notably. For raw water with 15 mg/L of humic acid, soluble-Al and total-Al residue were 1.8 and 2.1 mg/L, respectively, close to the quantity of Al addition. However, both enhanced coagulation and advanced pretreatment processes including ozonation, hypochlorous, and potassium permanganate oxidation had little effect on residual Al control.

(2) UV radiation decreases residual Al concentration notably. After 3 hr radiation, soluble-Al and total-Al in the treated water were 0.16 and 0.24 mg/L, respectively, much lower than those in the treated water without pretreatment. Residual Al concentration was controlled by the combination of ozonation and UV radiation. The optimal dosage was 0.5 mg O_3/mg C and radiation time was 3 hr for the raw water with 15 mg/L humic acid.

(3) In ozonation, the hydroxyl radicals attack organic compounds non-selectively, leading to a notable hydrophilization increase. Different from ozonation, the oxidation of UV light on humic acid was selective. Under UV light radiation, part of the bonds that existed among humic acid molecules were destroyed, and thus the adsorption of humic acid on polymeric Al and $Al(OH)_3(s)$ was enhanced. However, further study is needed to obtain direct evidence on the reaction mechanism of humic acid under UV radiation.

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