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# Comparisons of the film peeling from the composite oxides of quartz sand filters using ozone, hydrogen peroxide and chlorine dioxide

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## ABSTRACT

To solve the problem of shortened backwashing intervals in groundwater plants, several disinfectants including ozone ( $O_3$ ), hydrogen peroxide ( $H_2O_2$ ) and chlorine dioxide ( $ClO_2$ ) were examined to peel off the film from the quartz sand surface in four pilot-scale columns. An optimized oxidant dosage and oxidation time were determined by batch tests. Subsequently, the optimized conditions were tested in the four pilot-scale columns. The results demonstrated that the backwashing intervals increased from 35.17 to 54.33 ( $H_2O_2$ ) and to 53.67 hr ( $ClO_2$ ) after the oxidation treatments, and the increase of backwashing interval after treatment by  $O_3$  was much less than for the other two treatments. Interestingly, the treatment efficiency of filters was not affected by  $O_3$  or  $H_2O_2$  oxidation; but after oxidation by  $ClO_2$ , the treatment efficiency was deteriorated, especially the ammonia removal (from 96.96% to 24.95%). The filter sands before and after the oxidation were characterized by scanning electron microscopy and X-ray photoelectron spectroscopy. Compared with the oxidation by  $O_3$  and  $H_2O_2$ , the structures on the surface of filter sands were seriously damaged after oxidation by  $ClO_2$ . The chemical states of manganese on the surfaces of those treated sands were only changed by  $ClO_2$ . The damage of the structures and the change of the chemical states of manganese might have a negative effect on the ammonia removal. In summary,  $H_2O_2$  is a suitable agent for film peeling.

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## Introduction

As some water plants apply quartz sands as filter materials (Al-Gabr et al., 2014), a layer of black film tends to gradually be formed on the surface of the materials, and its components are manganese, silicon, magnesium, aluminum, iron and other elements (Guo et al., 2003). The particle size of the quartz sands increases and the surface of the film becomes smooth. The film tends to be overgrown after one to two years' continuous operation, and the resistance of the filter bed gradually increases under the same hydraulic loading, which shortens

the backwashing interval and decreases the water production period.

Ozone ( $O_3$ ), hydrogen peroxide ( $H_2O_2$ ), chlorine dioxide ( $ClO_2$ ) and chlorine are some of the types of disinfectants commonly used in water treatment (Li et al., 2013; Chhetri et al., 2014; Tachikawa and Yamanaka, 2014; Zhang et al., 2014). The hydroxyl radical generated by  $O_3$  oxidation has strong oxidation capability, and it can deal with many water pollution problems and will not pollute the environment (El-Araby et al., 2009).  $H_2O_2$  is used as an efficient oxidant, especially in the removal of organic materials (Pottage et al., 2010), and at high

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concentration it can generate excessive hydroxyl radicals, which can directly oxidize the organic pollutants in water.  $\text{ClO}_2$  has been reported to be effective in pathogenic organisms' inactivation, including the inactivation of *Cryptosporidium parvum*. The oxidation capacity of  $\text{ClO}_2$  is mainly determined by reaction time and pH of the water (Zhu et al., 1998), and it shows almost no reactivity with humic substances to form trihalomethanes (Korn et al., 2002). All the disinfectants have strong oxidation capability, so that it was reasonable to use them to oxidize and peel off the overgrown filter film from filter sand surfaces.

The aim of the present work was to select a suitable disinfectant for oxidation treatment of the overgrown filter film on the surface of the filter sands, which could not only increase the bed voidage and recover the backwashing interval of the filter, but also ensure that the water quality of effluent should be within the permitted limits. A series of batch tests were operated to determine the optimized dosage of oxidants and the oxidation time. Four pilot-scale columns were used to test the recovery of backwashing interval and the influence of the three kinds of disinfectants on the water quality of effluent.

## 1. Materials and methods

### 1.1. Pollution loading and pilot-scale columns

The raw water was pumped from the depth of 40 m by submersible pump in the northwest area of Xi'an City, China. As summarized in Table 1, the main pollutants in raw water were ammonia, iron and manganese.

The pilot-scale column was constructed of Plexiglas tubes with internal diameter of 0.1 m and height of 3.8 m, and the filter bed in the tube was filled to a height of 1.1 m. At the bottom of each tube was a support layer of 0.3 m depth. The filtration velocity of all pilot-scale columns was 7 m/hr. All pipes for water transport were made of random polypropylene, an important class of plastic resins. All pilot-scale columns were backwashed when the water reached a preset

overflow location, about 1.6 m above the bed layer. When the water quality of the effluent was deteriorated, the filter was backwashed as well. Backwashing was conducted via the following steps: air flow intensity = 50.8–72  $\text{m}^3/(\text{m}^2\cdot\text{hr})$ , water flow intensity = 15.1–36  $\text{m}^3/(\text{m}^2\cdot\text{hr})$ : (1) air scouring for 3 min at the bottom of column, (2) simultaneous air and water scouring for 4 min, and finally (3) water scouring for 3 min.

A daily analysis of water samples taken from certain positions of the pilot-scale columns was carried out to determine the concentrations of each pollutant in the filter. A schematic of the pilot-scale filter system is presented in Fig. 1.

### 1.2. Batch tests and the voidage

To determine the optimized oxidant dosage and oxidation time, a series of batch tests were performed as follows: The weighed filter sand (650 g, diameter of about 1–1.5 mm, and the mass of film was about 650 g  $\times$  6.3% = 40.95 g) was introduced into a basket made of stainless steel wire mesh with the pore size of 1 mm (Fig. 2), then the baskets were settled in polyethylene bottles (1 L). The oxidant solution (0.4 L) was added into the bottles. After a certain reaction time, the liquid sample was separated from solid sample in the bottom of the bottles, and the oxidized filter was washed by clean water until the wash water was clear. All the wash water was filtered through a 0.45  $\mu\text{m}$  membrane, and the solid sample was collected in a sand core funnel and the amount of peeled film was weighed; the solids were dried (120°C, 60 min) before being weighed. The batch experiments were performed for different reaction times of 5, 10, 20, 40, 80 min.

The oxidants of  $\text{O}_3$ ,  $\text{H}_2\text{O}_2$  and  $\text{ClO}_2$  were used in batch tests.  $\text{O}_3$  was generated from an ozone generator (CF-G-3-10G, Qingdao Guo Lin Industry Co., Ltd., China), and its production was in the range of 1.17–5.27 g/hr.  $\text{H}_2\text{O}_2$  was made from 30% analytical grade solution (Tianjin Tian Li Chemical Reagent Co., China), and  $\text{ClO}_2$  was made from chlorine dioxide effervescent tablets (Beijing Xinli World Environment Technology Co., Ltd., China). The initial concentration of  $\text{O}_3$  was controlled by adjusting the current intensity of an ozonizer ( $I = 0.2, 0.3, 0.6, 0.9$ ) and the initial concentrations of the other two oxidants ( $\text{H}_2\text{O}_2$  and  $\text{ClO}_2$ ) in the solution were 0.1, 0.2, 0.4, 0.6, 1.2 g/L, respectively.

The procedure of determining the voidage was as follows: as the backwashing finished, all the water of the column was emptied. Clean water was introduced into the filter column, and the water level was controlled to submerge the media exactly. After that, the clean water introduced into the column was drained and collected, the volume of collected water was measured and defined as  $V_1$ . The packing volume of the filter bed was calculated from the diameter and the height of the filter bed and was defined as  $V$ . So the voidage was  $V_1/V$ .

### 1.3. Analytical methods

Concentrations of ammonia, iron and manganese were determined by conventional spectrophotometric methods (China E. P. B. O., 2002) (HACH, DR5000, USA). Dissolved oxygen and pH were determined by a portable instrument (HACH, HQ30d, USA). The concentration of  $\text{O}_3$  was determined by indigo spectrophotometry (Bader and Hoigné, 1981). The concentration of  $\text{H}_2\text{O}_2$

**Table 1 – Water quality of raw water used in pilot-scale filter system.**

Parameters	Value	Quality standard for groundwater of China (GB/T 14848-9)
pH	7.2 $\pm$ 0.3	6.5–8.5
Temperature (°C)	15.0 $\pm$ 0.5	NG
Dissolved oxygen (mg/L)	1.45 $\pm$ 0.56	NG
Ammonium (mg/L)	1.30 $\pm$ 0.25	0.50
Ferrous Fe (mg/L)	0.85 $\pm$ 0.05	NG
Total Fe (mg/L)	1.25 $\pm$ 0.25	0.30
Manganese (mg/L)	1.15 $\pm$ 0.25	0.10
Total phosphorus (mg/L)	0.06 $\pm$ 0.01	NG
Total nitrogen (mg/L)	3.95 $\pm$ 0.85	NG
Turbidity (NTU)	1.45 $\pm$ 0.55	3.00
NG—not given.		

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