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Performance of an integrated process combining ozonation with ceramic membrane ultra-filtration for advanced treatment of drinking water

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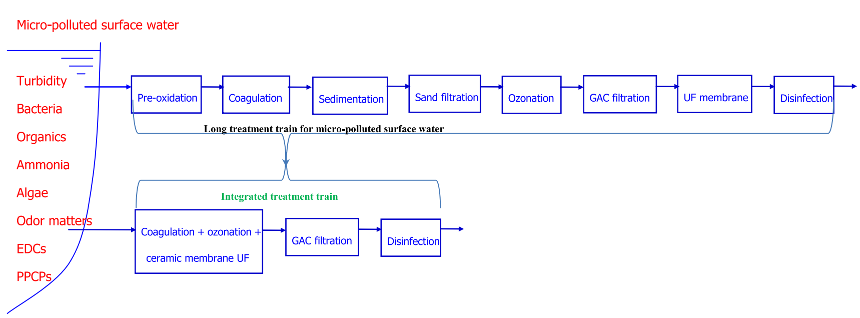
HIGHLIGHTS

- Integrated process includes coagulation, ozonation, ceramic UF and GAC filtration.
- Full-performance evaluations of the pilot-scale integrated process of 120 m^3/d
- Turbidity, DOC, ammonia, geosmin, 2-MIB, EDCs and PPCPs are efficiently removed.
- UF membrane fouling can be controlled in situ by ozonation.

GRAPHICAL ABSTRACT

The integration of UF with ozonation (i.e. UF/ozonation) in this paper merged 5 units (including pre-oxidation, sedimentation, filtration, main oxidation and UF) into one unit. Such a hybrid treatment train is much shorter and suitable for the upgrade of conventional water treatment plants in dealing with micro-polluted water sources for better supply of drinking water.



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ABSTRACT

An integrated process including coagulation, ozonation, ceramic membrane ultra-filtration and activated carbon filtration was investigated for the treatment of drinking water from the micro-polluted surface water in the southern China. A pilot-scale plant with the capacity of 120 m^3/d was set up and operated. Submerged flat-sheet ceramic membranes were selected with the average pore diameter of 60 nm and the filtration area of 50 m^2. Quite a number of water quality parameters were evaluated including turbidity, particle counts, coliform bacteria, *Cryptosporidium* and *Giardia* cysts, dissolved organic carbon (DOC), ammonia, geosmin, 2-methylisoborneol (2-MIB), 4 trihalomethanes, 6 haloacetic acids, 8 endocrine disrupting compounds (EDCs) and 14 pharmaceutical and personal care products (PPCPs). The experimental results showed that the removal efficiencies of all the evaluated parameters ranged from 64% to 100%. The ozonation in membrane tank is very helpful both in enhancing the performance of removing multiple contaminants and in controlling the membrane fouling with ozone dosage of 2–5 mg/L at the permeate flux of 100 L/m^2·h. The innovation of the integrated process was discussed based on the concept of nano-reactor to probe into the reaction activities inside the membrane pores. However, the detailed mechanisms need to be explored in the future.

1. Introduction

In some fast-developing and densely-populated areas in the south of China, many drinking water sources have been polluted with a variety of contaminants [1,2]. Natural organic matters (NOM) and ammonia are the main ones that cause many water quality problems. It was also

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found that geosmin and 2-mehtylisoborneol (2-MIB) are typical compounds that cause taste and odor problems [3]. With the help of advanced analytical instruments, many endocrine disrupting compounds (EDCs) and pharmaceuticals and personal care products (PPCPs) have also been detected at the level of ng/L in drinking water [4,5]. In a previous research, fifteen PPCPs were identified with the concentration of 0–36 ng/L in the source water and twelve PPCPs in the product water in a southern city of China [5]. The influence of both EDCs and PPCPs on water safety has attracted wide attention in China. Thus, the removal of PPCPs and EDCs along with regular contaminants should be taken into account in the treatment of drinking water.

The current technologies available for drinking water include some conventional methods, for example, coagulation, sedimentation, sand filtration and chlorination; and some advanced processes of ozonation and granular activated carbon. In addition, per-oxidation with oxidants, like ozone, chlorine or permanganate, and pretreatment with powdered activated carbon (PAC) were frequently used to control algal growth, color, taste or odor. In recent years, the whole treatment train will be too long and too costly for the complete removal of all the pollutants in the micro-polluted surface water. On the contrary, low pressure membrane ultra-filtration (UF) might provide a promising way to make the treatment train shorter by integrating multiple treatment units, for example, only one membrane unit can replace sedimentation and sand filtration operations. Membrane filtration is effective in removing particles, microorganisms and some organic matters with relatively smaller footprint and lower cost [6,7]. Therefore, the application of membrane filtration to drinking water treatment has developed quite fast in recent years [7]. However, membrane fouling is still one of the major challenges in membrane application, for it leads to the increase of operational cost with the decrease of specific permeate flux. Furthermore, chemical oxidation is usually used to alleviate membrane fouling [8–10]. But it may shorten the life of polymeric membranes, for it is weak in oxidation resistance.

Ceramic membranes are resistant to chemical oxidation so they have longer life time than polymeric membrane [11]. In this paper, ceramic membranes with ozonation process are set up in one tank to replace regular sedimentation, sand filtration, pre-ozonation and major ozonation. In this way, the whole treatment train would be much shorter. It is reported that the hybrid process combining ozonation with ceramic membranes is helpful to control membrane fouling [12–15]. When the ceramic membrane surface was coated with manganese, iron or titanium oxides, membrane fouling was alleviated more effectively [16,17]. Such a hybrid process has been demonstrated to be viable for dispersive supply of drinking water under tropic conditions [18]. However, most of the previous studies were focused on the alleviation of membrane fouling and the removal of regular organic matters, and very less attention was paid to the removal of contaminants like ammonia, EDCs and PPCPs. The evaluation of the whole treatment train for drinking water treatment has not been found in any literatures.

The objective of this study is to develop a highly integrated process including coagulation, ozonation, ceramic UF and GAC filtration to deal with micro-polluted source water. A pilot scale plant with the capacity of 120 m^3/d was set up and was operated like a normal water treatment plant in Dongguan City in the southern China. Full removal performances were evaluated, such as turbidity particles, bacteria, organic matters, DBPFPs, ammonia, geosmin, 2-MIB, EDCs, and PPCPs.

2. Materials and methods

2.1. Raw water

Raw water was collected from the Dongjiang River, an important drinking water source in the southern China, and a city canal in Guangdong province of South China, which is highly polluted by domestic sewage and industrial effluents. During rainy seasons, some water in the city canal overflows into the Dongjiang River. The raw water was taken directly by two submerged pumps to simulate the seasonal pollution; one was put in the Dongjiang River, while the other in the city canal; then mixed in a tank for the experiments. Two fixed sieves with a mesh width of 2 mm were used before the submerged pumps respectively to prevent small impurities from entering the pipes of the treatment equipment. Table 1 shows the composition of the raw water used in the test.

2.2. Membrane modules

Flat-sheet ceramic membranes with multi-channels (Meidensha, Japan) were used in this study. They are made of Al_2O_3 with a mean pore size of 60 nm. A single membrane sheet is 1046 mm (length) × 260 mm (width) × 6 mm (thickness). One membrane module contains 50 membrane sheets with the total filtration area of 25 m^2. Two membrane modules are selected for this study.

2.3. Experimental setup and system operation

Fig. 1 shows the schematic of the pilot scale treatment process in this study. The process contains coagulation, ozonation, ceramic UF, GAC filtration as well as disinfection. Coagulation is used as pretreatment for the destabilization of colloidal particles, for it is necessary to make the small particles form larger flocs in order to prevent them from blocking the membrane pores. Then, ceramic UF is used to remove flocs and microorganisms in the water. Ozonation is set up with ceramic UF in one reactor so that ozonation can take place not only on the membrane surface but also inside the membrane pores, to oxidize the organic contaminants into smaller molecules, and to detach the adsorbed foulants. Meanwhile, granular activated carbon filtration can further remove the residual organic matters, ammonia, EDCs, PPCPs, etc.

The water sample from the Dongjiang River and the city canal was mixed in a storage tank of 7.5 m^3 by an agitator, then, pumped into the coagulation tank with the prepared polymeric aluminum chloride as coagulant; the dosage was 2.4 mg/L, and the hydraulic retention time was 12 min. After the coagulated water was fed into the membrane reactor for ultra-filtration with a constant suction-flux of 100 $L/m^2 \cdot h$, ozone was aerated into the membrane reactor via a diffuse disc at the bottom of the membrane module. The permeate entered the following activated carbon filter (0.8 × 0.8 m in cross section, 2 m in depth) with a filtration velocity of 10 m/h. The granular activated carbon was selected with the diameter ranged from 0.6 mm to 2.0 mm; the mean equivalent diameter was 1.19 mm, iodine sorption value was 950 mg/g. Finally, the effluent of GAC filtration was disinfected with sodium hypochlorite. The hydraulic retention time of coagulation, ozonation, membrane and GAC filtration is 48 min, while 120 min for disinfection with an available chlorine dosage of 1.5 mg/L.

During the experiments, the membranes were back flushed automatically for 3 min at intervals of 240 min with a flush flux of 15 m^3/h, in order to remove the cake layer on the membrane surface. The sludge water was discharged periodically after back flushing.

Pure oxygen gas was vaporized from a liquid cylinder, and then fed into the ozone generator (OZONIA CFS-3 2G). The ozone concentration was modified by varying the voltage applied to the ozone generator. The excess ozone was vented through an ozone destruction unit (OZONIA ODT-003).

The electricity requirement for the pilot plant was 0.51 kWh/m^3 including raw water supplying, ultra-filtration, stirring, dosing, ozone generation and destruction of the residual ozone. The cost on chemicals was less than 0.16 RMB/m^3 including coagulants, disinfectants and pure oxygen.

2.4. Analytical methods

The flow rate of raw water and permeate as well as the cross-membrane pressure were monitored continuously with electronic