



Phosphate dosing to sustain the ammonium removal activity of an iron-manganese co-oxide filter film at pilot scale: Effects on chemical catalytic oxidation



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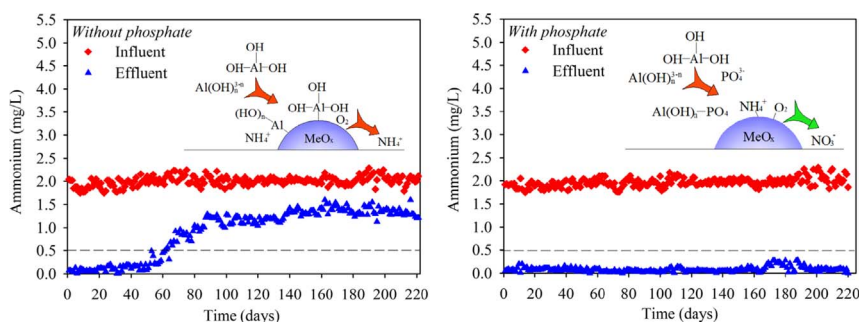
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GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:

Iron-manganese co-oxide filter film
Ammonium removal
Chemical catalytic oxidation
Phosphate
Surface water treatment

ABSTRACT

An iron-manganese co-oxide filter film (MeO_x) coated on filter sand has been shown to be a good catalyst for the oxidation of ammonium in the treatment of groundwater. However, its catalytic activity decreases gradually when it is used to remove ammonium from surface water. In this study, phosphate was added to sustain the ammonium removal activity of MeO_x in a pilot-scale filter system for surface water treatment. The results showed that the ammonium removal efficiency of the MeO_x filter without phosphate dosing decreased gradually, and just 27.69% removal efficiency was obtained at 6.1 °C. After dosing with 30 $\mu\text{g/L}$ phosphate, the ammonium removal efficiency remained high after continuous operation for 176 days and reached 85.74%, even at 6.1 °C. It was found that both the biological nitrification activity and the chemical catalytic activity of MeO_x were markedly enhanced after phosphate dosing (compared with that without phosphate dosing). Through inactivation by ozone, the biological nitrification and chemical catalytic activity were distinguished. It was found that chemical catalytic activity accounted for around 67.59% of the total ammonium removal. Aluminum was found to accumulate on the surface of MeO_x and combine competitively with its active sites. This may be the primary reason for the decrease in the catalytic activity of MeO_x . With phosphate dosing, the accumulation of aluminum decreased by 60.53% and the chemical combination of aluminum and MeO_x was suppressed, thus sustaining the catalytic activity of MeO_x .

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1. Introduction

Ammonium is a major pollutant in surface water, and a strict standard of 0.5 mg/L $\text{NH}_4^+\text{-N}$ in drinking water has been set by the Chinese government [1]. Excessive ammonium in drinking water is undesirable and may cause many problems, including unpleasant odors, the promotion of microbial growth in water distribution systems, a reduction in the effectiveness of chlorine disinfection, and increased chlorine demand [2–4]. The general method to remove ammonium during drinking water treatment is a biological process [5–7]; however, incomplete nitrification may occur in biological filters, leading to nitrite accumulation in the treated water at low temperatures [8–10].

Recently, a new method, the chemical catalytic oxidation of ammonium using an iron–manganese co-oxide filter film (MeO_x), has been reported for groundwater treatment and ammonium removal, and its reaction mechanism has been investigated systematically [11–13]. It was reported that, after the rapid start-up period, the ammonium concentration in the effluent of the pilot-scale MeO_x filter system remained lower than ca. 0.2 mg/L, even at a 6.6 °C, for groundwater treatment when the concentration in the influent ranged from 0.8 to 2.4 mg/L [12]. Further studies showed that the active ingredients in this catalyst were manganese oxides (MnO_x), specifically hexagonal birnessite [13]. In addition, $\text{MeO}_x/\text{MnO}_x$ can be prepared easily and continuously on virgin filter media using potassium permanganate to oxidize the manganese and ferrous ions derived from raw groundwater. It was found that a MnO_x -based filter could be successfully started up within 6 days, increasing the ammonium removal efficiency from 1% to 96% [13]. Two mechanisms have been proposed for this catalytic oxidation reaction. One possible mechanism is that reactive oxygen species are formed that oxidize NH_4^+ to NO_3^- when oxygen is adsorbed on the MnO_x surface [12]. Another possible mechanism is that NH_4^+ is oxidized directly by Mn(IV) or Mn(III) present in the filter film, resulting in the formation of NO_3^- . In this process, Mn(II) would be produced, followed by oxidation via self-catalyzed oxidation by MnO_x [13].

For the traditional catalytic oxidation of ammonium in water, such as photocatalytic oxidation, the catalysts are generally expensive noble metals, and the reactions require harsh conditions [14–16]. Thus, it is challenging to apply these methods to water treatment. Fortunately, the MeO_x filter system has great potential for application in actual water treatment plants because it is inexpensive and easy to use. This new technology has been used for a reconstruction and expansion project of the Fourth Plant in Xi'an (Shaanxi Province, China). Groundwater is the water source for this plant. The full-scale treatment train in this new plant consists of aerators, followed by a filtration treatment using the MeO_x filter. After this, the water is treated using O_3 -BAC biological filters, and then the effluent water is disinfected to produce clean drinking water. The new plant will produce 120,000 m^3/d of drinking water when this project is completed.

However, the application of MeO_x for ammonium removal from surface water is quite unknown. In 2014, surface water provided 80.8% of the total water supply in China, and ammonium is a major pollutant [17]. Compared with groundwater, it has some significantly different characteristics (including low water temperatures in winter, seasonal fluctuations in water quality, low alkalinity, low pH, and requires complex pre-treatment processes), making the removal of ammonium more difficult [8,18,19]. Therefore, it is meaningful to test MeO_x for ammonium removal from surface water.

Preliminary experimental results have shown that, although the MeO_x filter can remove ammonium from surface water effectively over time, its ammonium removal activity gradually decreased. For biological nitrification, the ammonium removal efficiency could be sustained by dosing the water with phosphate before filtration to supply sufficient nutrients for the growth of microorganisms [20,21]. Therefore, the question arose whether the phosphate dosing can enhance the ammonium removal by MeO_x . Because it has been reported that ammonium is removed by catalytic oxidation in a MeO_x filter [12], our research

concentrates on determining the effects of phosphate dosing on the chemical catalytic oxidation. The ammonium removal efficiency of the MeO_x filter with or without phosphate dosing and the changes of the catalytic activity under both operating conditions were investigated systematically. Moreover, the changes of MeO_x characterization were determined to investigate how the addition of phosphate sustains the catalytic activity of MeO_x .

2. Materials and methods

2.1. Pilot-scale filter system

The pilot-scale filters used in this study were made of two identical plexiglass cylinders, each with an inner diameter of 10 cm and denoted A_1 and A_2 , respectively. The active filter beds fixed in the filter columns were 110 cm high (Fig. 1). Sampling ports were placed on the vertical filter column at 10 or 20 cm intervals. First, NH_4Cl and Na_2HPO_4 stock solutions were dosed into the feed water of the filter system using peristaltic pumps.

Then, the synthetic water was passed through the MeO_x filter. The feed water used in this experiment was the effluent water from the sedimentation tank in a pilot-scale surface water treatment plant in Xi'an. The water fed to this plant is mainly reservoir water. As the source water entered the plant, it was dosed with coagulants (polymeric aluminum ferric chloride, PAFC); then, the water was treated in a grid flocculation tank followed by an inclined-tube sedimentation tank.

2.2. Ammonium removal experiments

The MeO_x coated sands used in this study were prepared using potassium permanganate to oxidize the Mn^{2+} and Fe^{2+} derived from groundwater before filtration through a rapid-sand filter, as described by Guo et al. [12]. Such sand has been used for about four years in a pilot-scale plant for the simultaneous treatment of groundwater containing ammonium, manganese, and iron. The quality of the feed water is shown in Table 1 (determined according to the guidelines set by the Ministry of Environmental Protection of China [22]). Phosphate was added to the feed water of filter column A_1 at 30 $\mu\text{g PO}_4^{3-}\text{-P/L}$. For comparison, no phosphate was added to filter column A_2 . The influent and effluent water samples from the two filter columns were taken daily, and water samples along the filter depth were taken every 1–2 weeks. The filtration velocity was maintained at 7 m/h.

2.3. Methods to distinguish the effects of biological nitrification from those of the chemical catalytic oxidation of ammonium

A lab-scale experiment was conducted to distinguish the effects of biological nitrification from those of the chemical catalytic oxidation of ammonium. The experimental reactor was a 25-cm high transparent plexiglass column with an inner diameter of 10 cm, and a stainless-steel mesh was placed in the reactor to allow the placement of the MeO_x -coated sand (Fig. S1). Previous studies have shown that the structural changes to MeO_x caused by ozone (O_3) treatment are negligible [12,23]. Therefore, ozone was used to inactivate the nitrifying bacteria on MeO_x to distinguish the effects of biological nitrification from those of chemical catalytic oxidation of ammonium. The O_3 was produced by an ozone generator (Guo LinCF-G-3-10 g, Qingdao Guo Lin Industry Co., Ltd., China). Ozone was introduced into the reactor continuously from the bottom for about 2.5 h, and the concentration of O_3 in the water was maintained at 6.5–7.0 mg/L. Before and after inactivation, the specific ammonium oxidation rates (SAOR) of the MeO_x coated sand were determined as described by De Vet et al. [24]. The reaction solution (1.5 L) was poured into the reactor, and 100 g of MeO_x coated sand was placed on the stainless-steel mesh. The reaction solution was prepared using the feed water of the filter system. The initial ammonium concentration was 10 mg $\text{NH}_4^+\text{-N/L}$, and the pH was controlled

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