JOURNAL OF ENVIRONMENTAL SCIENCES XX (2018) XXX–XXX

ELSEVIER

Available online at www.sciencedirect.com

ScienceDirect

www.elsevier.com/locate/jes

JES JOURNAL OF ENVIRONMENTAL SCIENCES www.jesc.ac.cn

Structural characteristic and ammonium and manganese catalytic activity of two types of filter media in groundwater treatment

Ya Cheng, Tinglin Huang*, Lijie Cheng, Yuankui Sun, Laisheng Zhu, Ye Li

Key Laboratory of Northwest Resource, Environment and Ecology, MOE, Xi'an University of Architecture and Technology, Xi'an 710055, China
Shaanxi Key Laboratory of Environmental Engineering, Xi'an University of Architecture and Technology, Xi'an 710055, China

ARTICLE INFO

Article history:
Received 3 November 2017
Revised 22 December 2017
Accepted 22 December 2017
Available online xxxx

Keywords:
Natural filter media
Active filter media
Manganese and ammonium removal
Manganese oxides
Characterization
Activity

ABSTRACT

Two types of filter media in groundwater treatment were conducted for a comparative study of surface structure and catalytic performance. Natural filter media was adopted from a conventional aeration–filtration groundwater treatment plant, and active filter media as a novel and promising filter media was also adopted. The physicochemical properties of these two kinds of filter media were characterized using numerous analytical techniques, such as X-Ray diffraction (XRD), scanning electron microscope (SEM), energy dispersive X-ray (EDX), X-ray photoelectron spectroscopy (XPS) and Zeta potential. The catalytic activities of these filter media were evaluated for ammonium and manganese oxidation. XRD data showed that both active filter media and natural filter media belonged to birnessite family. A new manganese dioxide (MnO_2) phase (PDF#72-1982) was found in the structure of natural filter media. The SEM micrograph of natural filter media showed honeycomb structures and the active filter media presented plate structures and consisted of stacked particle. These natural filter media presented lower level of some trace elements such as calcium and magnesium, lower degree of crystallinity, lower Mn(III) content and lattice oxygen content than that of active filter media, which were associated with its poor ammonium and manganese catalytic activities. In addition, some γ-Fe_2O_3 and $MnCO_3$ were found in the coating which may hinder the ammonium and manganese catalytic oxidation. This study provides a thorough and comprehensive understanding about the most commonly used filter media in water treatment, which can provide a theoretical guide to practical applications.

Introduction

Manganese (Mn^{2+}) and ammonium (NH_4^+) are common inorganic pollutants with salient features present in groundwater (Du et al., 2016; Tekerlekopoulou et al., 2013). Presence of manganese in finished water may cause organoleptic and operational problems including discoloration of water, unpleasant metallic taste and odor, increased turbidity and biofouling of pipelines as well as staining of laundry and plumbing fixtures (Cai et al., 2015; Hasan et al., 2013). The presence of ammonium in water systems also leads to oxygen depletion, eutrophication of surface water and toxicity for fish

* Corresponding author. E-mail: huangtinglin@xauat.edu.cn (Tinglin Huang).

https://doi.org/10.1016/j.jes.2017.12.014
1001-0742/© 2018 The Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences. Published by Elsevier B.V.

Please cite this article as: Cheng, Y., et al., Structural characteristic and ammonium and manganese catalytic activity of two types of filter media in groundwater treatment, J. Environ. Sci. (2018), https://doi.org/10.1016/j.jes.2017.12.014

(Cai et al., 2015; Cheng et al., 2017a). The increasing and very often uncontrollable use of fertilizers has led to increased amounts of ammonium and manganese in potable water, which often exceed the upper permitted limit (Tekerlekopoulou and Vayenas, 2012). Most countries have their own maximum concentration limits as a guideline to produce good quality drinking water. The World Health Organization (WHO, 2004) has regulated that standard limits for NH_4^+–N and Mn^{2+} in drinking water should be below 1.5 and 0.1 mg/L, respectively (SEPA, 2002). In China, the maximum concentration limits for NH_4^+–N and Mn^{2+} in treated drinking water are 0.1 and 0.5 mg/L, respectively. Manganese and ammonium removal becomes a problem when using groundwater as a drinking water, especially in country sites, outside big cities (Taffarel and Rubio, 2010). Therefore, controlling the NH_4^+–N and Mn^{2+} content in drinking water has become a great public health issue.

Processes for the removal of ammonium ions from potable water include chlorination, ion exchange, membrane filtration and biological treatment processes (Oehmen et al., 2006; Vaaramaa and Lehto, 2003; Fu et al., 2011). Chlorination is usually limited due to the high volumes of sludge and the cost of the oxidants and ion exchange, and membrane filtration are limited due to their higher capital and operational costs (Tekerlekopoulou et al., 2013; Cai et al., 2015). Biological treatment processes as an additional and effective advanced biological system for the simultaneous removal of NH_4^+-N and Mn^{2+} in the treatment of drinking water attracts many attentions of researchers. However, actually numerous groundwater treatment facilities have opted to reduce their costs by applying origin quartz sand filter media without any pretreatment, which is called "conventional aeration-rapid sand filtration". It is reported that more than 95% of the water treatment plants (WTPs) in China were still using the conventional water treatment process as of 2010 (Feng et al., 2012). Also, in European countries, the removal of manganese from groundwater is commonly achieved by aeration-rapids and filtration (Bruins et al., 2015a). The effect, over time, is the generation of a manganese oxide film on the media surface of the quartz sand (hereafter called the "natural filter media"). However, as general, in-rapid sand filters used for groundwater treatment, conditions for chemical formation of auto-catalytically active manganese oxides on virgin filter media, which can adsorb and subsequently oxidize dissolved manganese, are poor since most groundwater has a low redox potential and pH (Bruins et al., 2015a). Limited removal of manganese and ammonium could be achieved. Bruins et al. (2015a) have paid much excellent effort on how the filter media ripening process and confirmed the origin of the manganese oxide film. Some other researchers proposed the property of the manganese oxide depends on the valence of manganese in the oxide (Anschutz et al., 2005). Despite this, the relationship between the structure and ammonium and manganese catalytic activity of the mature filter media has not been established.

Besides, another novel and promising active manganese oxide filter media (hereafter called the "active filter media") has been developed for simultaneous removal of ammonium and manganese in recent years and these filter media exhibit some advantages, such as short start-up time and good low temperature resistance (Guo et al., 2017; Cheng et al., 2017b, 2017c). This filter media have special ammonium and manganese catalytic oxidation ability (Cheng et al., 2017b). A review of water research publications was conducted in this research effort to seek out any previously performed studies related to the comparative investigation of structure and activity of these two types of filter media. However, up to now, rarely, work has been done on the comparative study of Q5 the above filter media.

In this study, we make a comparative study of the active filter media and natural filter media which have been running for more than one year. Structural (X-ray diffraction (XRD)), morphological (scanning electron microscope (SEM)), compositional (energy dispersive X-ray (EDX) spectroscopy) and elements state (X-ray photoelectron spectroscopy (XPS)) characterization of the filter film have been used to understand differences among these filter media and their effects on catalytic activity. Catalytic activity tests were carried out with contaminant such as ammonium and manganese.

1. Materials and methods

1.1. Source of filter media

The active filter media were adopted from pilot-scale water treatment plant. The initiating process of this type filter was carried out by oxidation–reduction method. The potassium permanganate ($KMnO_4$) solution, the manganese chloride ($MnCl_2$) solution and ferrous chloride ($FeCl_2$) solution were separately pumped into the tube. After mixed by static mixer, the oxidation reaction of manganese and iron with potassium permanganate are listed as follows:

$$3Fe^{2+} + MnO_4^- + 7H_2O \rightarrow 3Fe(OH)_3 + MnO_2 + 5H^+ \quad (1)$$

$$3Mn^{2+} + 2MnO_4^- + 2H_2O \rightarrow 5MnO_2 + 4H^+ \quad (2)$$

During the process, manganese and iron were dosed at the concentration of 2 and 1 mg/L, respectively. The concentration of potassium permanganate was determined by the stoichiometry, and then, iron-manganese oxides were generated. The iron-manganese oxides along with the raw water was carried to the filter and deposited on the surface of the quartz sands. With more and more iron-manganese oxides deposited during the initiating process, an active film finally was formed. The active filter has running continuously for 1 year. The active filter media samples were taken from the top of the filter bed.

The natural filter media were adopted from large water treatment plant. The characteristics of feed water are as follows: Fe 0.05–0.10 mg/L, Mn 0.05–0.10 mg/L, NH_4^+-N 0.01–0.10 mg/L, pH 7.8–8.0, Redox potential 200–280 mv and hardness190–200 mg/L. The feed water is faced with seasonal iron and manganese pollution and ammonium in the feed water is always below the limited concentration (<0.5 mg/L). This plant has been running for more than 1 year. A thick film was formed on the surface of the quartz sand. All filter media samples were taken from the top of the Q6 filter bed.

Please cite this article as: Cheng, Y., et al., Structural characteristic and ammonium and manganese catalytic activity of two types of filter media in groundwater treatment, J. Environ. Sci. (2018), https://doi.org/10.1016/j.jes.2017.12.014