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Mn oxide coated catalytic membranes for a hybrid ozonation–membrane filtration: Comparison of Ti, Fe and Mn oxide coated membranes for water quality

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ABSTRACT

In this study the performance of catalytic membranes in a hybrid ozonation–ceramic membrane filtration system was investigated. The catalytic membranes were produced by coating commercial ceramic ultrafiltration membranes with manganese or iron oxide nanoparticles using a layer-by-layer self-assembly technique. A commercial membrane with a titanium oxide filtration layer was also evaluated. The performance of the coated and uncoated membranes was evaluated using water from a borderline eutrophic lake. The permeate flux and removal of the organic matter was found to depend on the type of the metal oxide present on the membrane surface. The performance of the manganese oxide coated membrane was superior to that of the other membranes tested, showing the fastest recovery in permeate flux when ozone was applied and the greatest reduction in the total organic carbon (TOC) in the permeate. The removal of trihalomethanes (THMs) and haloacetic acids (HAAs) precursors using the membrane coated 20 times with manganese oxide nanoparticles was significantly better than that for the membranes coated with 30 or 40 times with manganese oxide nanoparticles or 40 times with iron oxide nanoparticles.

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1. Introduction

As a result of the demand for high quality water there is a need for more effective, economical and energy efficient processes for the treatment of surface and contaminated water sources (Shannon et al., 2008). Over the last two decades, the use of membrane filtration for water treatment has expanded rapidly (Leiknes, 2009) as costs have decreased (Laine et al., 2000) and performance has increased (Song et al., 2003; Weber et al., 2003).

Although polymeric membranes are used in most water and wastewater applications, the use of ceramic membranes is becoming more common. For example, METAWATER (formerly NGK) has installed ceramic membranes in more than 70 water and wastewater treatment plants (Myers, 2009). One significant advantage of ceramic membranes is their long service life compared to polymeric membranes. In fact, Krüger Inc. offers a 20-year warranty on their ceramic membranes (Myers, 2009). The chemical resistance of ceramic membranes allows for the use of ozone and other strong oxidants to

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improve contaminant removal and reduce the problems associated with fouling. The use of ozone for pretreatment (Park, 2002; Lee et al., 2004) or in combination with membrane filtration (Schlichter et al., 2003; Karnik et al., 2005a; You et al., 2007; Kim et al., 2008) reduces membrane fouling. The effectiveness of the hybrid ozonation–membrane filtration process depends upon the gaseous ozone concentration; the pH of the feed water (Lee et al., 2004; Karnik et al., 2005a,b; Kim et al., 2008); operational parameters such as transmembrane pressure (TMP) and cross flow velocity (Kim et al., 2008); and the nature of the membrane surface (Karnik et al., 2005c, 2009). With both fluoropolymer and ceramic membranes, the concentration of residual ozone in the reject stream was found to be critical to system performance in terms of permeate flux (e.g., You et al., 2007; Karnik et al., 2005a). With a polyvinylidene fluoride ultrafiltration membrane, You et al. (2007) found that a dissolved ozone concentration of approximately 4.0 mg/L was necessary to maintain a permeate flux of 90% of its initial value. By contrast, with a titania membrane, Karnik et al. (2005a) found that fouling could be significantly reduced at much lower ozone concentrations; at an ozone residual concentration of 0.05 mg/L, in the recirculation feed water, the permeate flux was 95% of the clean water flux.

The surface properties of ceramic membranes allow for the use of coating, doping and grafting techniques to improve membrane performance (Karnik et al., 2005c, 2006, 2007, 2009; Zhang et al., 2009; Wei and Li, 2009). Metal oxides promote the decomposition of ozone and the formation of OH or other radicals (for example, see Sánchez-Polo et al., 2006; Wu et al., 2008; Zhao et al., 2008, 2009). Consequently, the performance of the hybrid ozonation–membrane filtration system can be enhanced by coating the membrane surface with metal oxide catalysts such as iron oxide (Karnik et al., 2005c, 2006, 2007, 2009) and manganese oxide (Davies et al., 2010). Karnik et al. (2005b) reported the enhanced removal of the THM precursors and ozonation byproducts, such as aldehydes, ketones and ketoacids using hybrid ozonation–filtration and confirmed the OH radical reaction mechanism using salicylic acid as a probe (Karnik et al., 2007). The morphological characteristics related to enhanced catalytic efficiency of the iron oxide coated membranes were described by Karnik et al. (2005c, 2009).

In this paper, the performance of ceramic membranes with three different filtration layers (Ti, Fe and Mn oxide), in a hybrid ozonation–membrane filtration system is compared. The permeability of the coated membranes and the removal of TOC, trihalomethanes (THMs) and haloacetic acids (HAAs) precursors by the hybrid ozonation–filtration system were studied to gain a better understanding of how the coating of the membrane affected the performance of the system.

2. Experimental methods

2.1. Feed water

The feed water was obtained from Lake Lansing (Haslett, Michigan), a borderline eutrophic lake. Water samples were collected at the boat ramp at the Lake Lansing Park – South, Haslett, Michigan in five-gallon carboys and stored at 4 °C in a refrigerator. Water samples were pre-filtered through a 0.5-μm ceramic cartridge microfilter (Doulton USA, Southfield, MI). Table 1 shows the water quality data obtained for the water used in this study. Simulated distribution system (SDS) THMs and SDS HAAs were measured after chlorination to simulate the formation of these disinfection byproducts (DBPs) in the distribution system. As shown in Table 1, seasonal variations of UV_{254} and SDS THMs and SDS HAAs were observed. The sample collected in Spring contained more compounds that absorbed UV radiation at 254 nm and had a higher concentration of HAA and THM precursors than that collected in Winter. Other water quality parameters for Lake Lansing have been reported elsewhere (Karnik et al., 2005a,b).

2.2. Hybrid ozonation–filtration setup

Fig. 1 presents a schematic of the hybrid ozonation–membrane filtration apparatus used in these experiments. The apparatus consists of an ozone injection system, membrane module, recirculation pump, feed tank and data acquisition system. The membrane module housing (TAMI North America, St. Laurent, Québec, Canada) is made of stainless-steel. The system was operated in a total (permeate and retentate) recycling mode in which the permeate was, as shown in Fig. 1, recycled using a pump (Masterflex®, Cole Parmer Inc., Vernon Hills, IL) into a feed tank at 15 min intervals using a timer. The transmembrane pressure (TMP) and cross flow velocity were controlled using a recirculation pump (Gear Pump Drive, Micropump®, Cole Parmer Inc., Vernon Hills, IL) and a back pressure regulator (Swagelok®, Solon, OH). The temperature of the water was 22.5 ± 0.5 °C during all the experiments. Temperature, cross flow rate and pressure were monitored continuously with a multifunctional sensor (L Series, Alicat Scientific, Tuscon, AZ) installed in the recirculation line and recorded using a LabView data acquisition system. The permeate mass was measured by an electronic balance (Adventure Pro Analytical Balance, Ohaus Corp., Pine Brook, NJ) at intervals of 60 s. The operational conditions for hybrid ozonation–membrane filtration are summarized in Table 2.

2.3. Ozone injection system

Ozone was generated using a high-pressure ozone generator (Atlas Series, Absolute Ozone® Generator, Absolute System Inc., Edmonton, Canada) that can operate as pressures of up to

Table 1 – Water quality characteristics of Lake Lansing (LL) after prefiltration.

Water quality parameters	Winter (2009) (Dec. 18)	Spring (2009) (Mar. 17)
pH	8.0	7.9
TOC (mg C/L)	10.4 ± 0.12[a]	12.0 ± 0.5
UV_{254} (1/cm)	0.186	0.368
SUVA (L/mg cm)	1.78	3.07
SDS HAA (μg/L)	347 ± 5	455 ± 13
SDS THM (μg/L)	137 ± 3	436 ± 1
Conductivity (μS/cm)	270	268

a Standard deviation for analysis.

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