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Analysis of energy costs for catalytic ozone membrane filtration

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ABSTRACT

Membrane fouling can be reduced through shear stress generated by cross-flow at the membrane surface. Previous work has shown that presence of ozone can reduce membrane fouling. In this work, the effect of ozonation and cross-flow on both membrane fouling on ceramic membranes and the energy cost for the process was studied. The effect of ozone dosage on membrane fouling was studied in both cross-flow and dead-end configurations. The performance of a manganese oxide coated membrane was compared with that of uncoated titanium oxide membrane. Membrane fouling decreased with increasing ozone dosages in the manganese oxide coated catalytic membrane, although increasing the dosage beyond 10–15 μg/s yielded limited improvement. The most energy-efficient mode of operation was found to be dead-end filtration using a manganese oxide coated membrane and a 10 μg/s ozone injection rate.

1. Introduction

Ultrafiltration (UF) membrane filtration can effectively remove suspended particles, colloids, bacteria, viruses and significant fraction of natural organic matter (NOM) from feed water [1]. There are a number of advantages of UF membrane treatment over conventional water treatment technologies, including reduced sludge production [2], smaller footprint [3], and the physical exclusion of microorganisms from the product water. However, membrane fouling continues to be one of the main factors that limit the application and increase the operational costs of membrane technologies [4]. Fouling results in a decrease in the production rate of purified water and a concomitant increase in the requisite transmembrane pressure [5].

In water treatment, NOM is the predominant cause of UF membrane fouling [6–9]. The use of ozone in combination with membrane filtration has been reported to reduce membrane fouling [10–14]. Ozone, a powerful oxidant, is reactive with NOM [15]. Pre-ozonation can also reduce formation of trihalomethanes (THMs) during subsequent chlorination [16].

Ceramic membranes are chemically stable, have longer lifetimes than polymeric membranes, and they can be used in conjunction with ozonation without degradation of the membrane [17,18]. Moreover, the surface properties of ceramic membranes may be altered to improve membrane performance. Metal oxides, such as titania [10,11], iron oxide [11], and manganese oxide [11,19,20], can act as catalysts for the degradation of ozone and the formation of hydroxyl radicals. These catalytic reactions are assumed to occur at the metal oxide surface. Kim et al. [12] studied the performance of nanoparticle-enhanced membranes and concluded fouling was dependent upon the physicochemical aspects of nanoparticles, such as particle size, hydrophilicity, and surface charge. Byun [11] reported that when used with ozone the performance of the manganese oxide coated membrane was superior to that of Ti and Fe oxide membranes in terms of fouling mitigation and the reduction of TOC in the permeate. In addition, the coating of the membranes with manganese oxide nanoparticles significantly reduced the concentrations of THM and haloacetic acids (HAA) precursors found in the permeate [19]. Szymanska et al. [13] and Zouboulis et al. [21] showed that with hybrid ozonation ceramic membrane microfiltration, mitigation of membrane fouling was well controlled. Wei et al. [22,23] demonstrated that pre-ozonation mitigated fouling on a UF membrane that was caused by soluble extracellular organic matters that had been released from *Microcystis aeruginosa.* They found that ozone reacted with the cake and gel layers and prevented membrane pore blocking.

The objective of this study was to analyze the energy costs for membrane filtration, in the presence of ozone, using either a manganese oxide coated ceramic membrane or titania oxide coated membrane (virgin membrane). Studies were conducted to determine the optimum ozone dosage and hydrodynamic conditions for this hybrid membrane system. The energy costs for both ozone generation and pumping were evaluated to determine the operating conditions for which the energy costs were minimized.

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2. Experimental methods

2.1. Feed water

The source water was obtained from Lake Lansing (Haslett, Michigan), a borderline eutrophic lake. The water was stored in the dark at 4 °C until use. Water samples were pre-filtered through a 0.5-μm ceramic cartridge micro-filter (Doulton USA, Southfield, MI) to remove larger particles. After filtration, the average TOC concentration in the test water was 10 ± 1 mg C/L. Before commencing each experiment, the temperature of the feed water was adjusted to room temperature (20 ± 3 °C).

2.2. Membrane module

The nominal molecular weight cut-off of the virgin membrane (Inside CeRAM, TAMI North America, Saint - Laurent, Quebec, Canada) was 5 KDa. The seven-channel membrane had a total filtering surface area of 131.9 cm^2, an active length of 25 cm, and an external diameter of 10 mm. The grain size within the support layer and the filtration layer of the virgin membrane varied between 132 nm and 296 nm and between 1.05 and 6.64 nm, respectively [11]. The clean water permeability of the coated membrane was 80 ± 2 L/m^2 h.

The virgin membrane was coated with manganese oxide according to the procedure described by Corneal et al. [20]. In this study, the performance of a membrane coated twenty times manganese oxide nanoparticles was examined, as higher permeate fluxes were obtained with membrane coated twenty times coated than with those coated thirty or forty times. Byun et al. [11] found the Mn oxide coating to be crystalline Mn_2O_3, and that for the membrane that was coated 20 times, the thickness of coating ranged from 14 to 54 nm. With the manganese oxide coated membranes, the lower operating pressure was used because the permeability of the manganese oxide coated membrane was higher than that of the uncoated membrane, as sintering at 500 °C leads to coarsening of the grains within the filtration layer [19].

2.3. Ozone contactor

A schematic for the ozone contactor is shown in Fig. 1. The water in the contactor was maintained at a constant level using a conductivity water level sensor connected to a programmable logic relay (SG2 PLR, B&B Electronics, IL), which opened or closed a solenoid valve (6013, Bürkert, Germany) to regulate water flow from the sample reservoir to the contactor. Ozone was generated from pure, dry oxygen (99.999%) using a corona discharge ozone generator (Absolute Ozone®, Absolute System Inc., Edmonton, AB, CA). An ozone monitor (Model 450H, Teledyne Technologies Inc., San Diego, CA) was used to measure ozone concentration. The flow rate of ozone gas was controlled at 10 mL/min by a rotameter (Cole-Parmer Inc., Vernon Hills, IL) installed between the ozone monitor and the membrane module. The ozone transfer efficiency in the contactor was determined before each experiment, by measuring the difference in the influent and effluent gaseous ozone concentrations and multiplying that difference by the gas flow rate.

2.4. Hybrid ozonation – filtration setup

The schematic of the ozone injection system is shown in Fig. 1. Nitrogen gas (99%, Airgas) was used for pressurizing feed tanks. The membrane module housing (TAMI North America, St. Laurent, Québec, Canada) was made of stainless steel. Both dead-end filtration and cross-flow filtration were performed at volumetric flow rate through the membrane of 18 ± 1.5 mL/min. This corresponds to a flux of 81.2 ± 2.8 L/m^2 h, which is typical of that used in full-scale water treatment. The flux was maintained using a peristaltic pump (Eldex®, Eldex Laboratories, Inc., Napa, CA). A recirculation pump (Materflex®, Cole Parmer Inc., Vernon Hills, IL) used when operating the system in cross-flow mode to obtain the constant cross-flow velocity of 0.5 ± 0.1 m/s. The transmembrane pressure (TMP) was recorded by a multifunctional sensor (L Series, Alicat Scientific, Tuscon, AZ) every 60 s. Temperature and atmosphere pressure were also monitored every 60 s by the multifunctional sensor. The permeate flux was measured using an electronic balance (Adventure Pro Analytical Balance, Ohaus Corp., Pine Brook, NJ) every 60 s. The transmembrane pressure (TMP, bar) and permeate flux (J, L/m^2 h) were recorded by Flow Vision SC (Alicat Scientific, Tucson, AZ) data acquisition software.

Experiments were conducted at ozone dosage injection rates of 5, 10, 15, and 20 μg/s. Control experiments were conducted to measure membrane flux without gas injection. The range of ozone dosages was selected based on prior work, which indicated that ozone dosages greater than 20 μg/s resulted in little enhancement

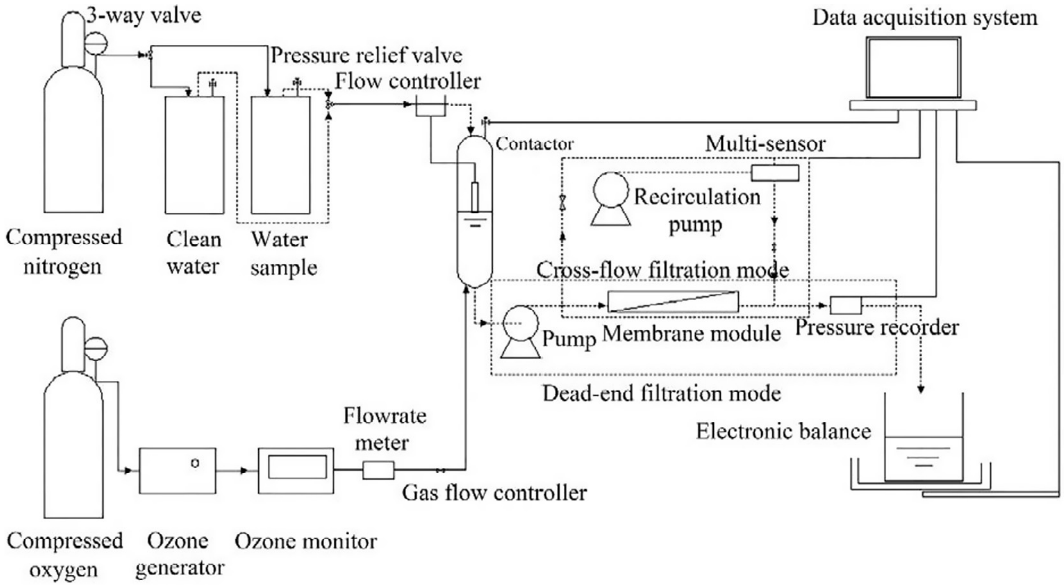

Fig. 1. Schematic of the ozonation-membrane filtration system.