Journal of Membrane Science 371 (2011) 99–107

Contents lists available at ScienceDirect

Journal of Membrane Science

journal homepage: www.elsevier.com/locate/memsci

Membrane fouling due to dynamic particle size changes in the aerated hybrid PAC–MF system

Mohiuddin Md. Taimur Khan[a,*], Satoshi Takizawa[b], Zbigniew Lewandowski[c], Warren L. Jones[c], Anne K. Camper[c], Hiroyuki Katayama[b], Futoshi Kurisu[b], Shinichiro Ohgaki[d]

[a] Center for Molecular Discovery, MSC08 4640, University of New Mexico, Albuquerque, NM 87131, USA
[b] Department of Urban Engineering, University of Tokyo, Tokyo 113-8656, Japan
[c] Center for Biofilm Engineering and Department of Civil Engineering, Montana State University, Bozeman, MT 59717, USA
[d] National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba-City, Ibaraki 305-8506, Japan

ARTICLE INFO

Article history:
Received 9 September 2010
Received in revised form 6 January 2011
Accepted 14 January 2011
Available online 22 January 2011

Keywords:
PAC–MF hybrid system
Particle size change
Suspended solids degradation
Membrane resistance
Biofiltration

ABSTRACT

To quantify the effect of dynamic particle size changes and degradation and accumulation of suspended solids (SS) in influents to reactors on membrane fouling frequency in hybrid powder-activated carbon (PAC)–microfiltration (MF) reactors, we operated a PAC–MF system (hollow-fiber module) for more than five months to purify river water before and after pretreatment by a biofilter. The transmembrane pressure, backwashing pressure, resistance to filtration, and SS accumulation and degradation during these dynamic changes were evaluated. The initial dose of PAC was 40 g/L of the reactor and no additional PAC was added during this continuous operational period. The presence of PAC reduced the membrane resistance to filtration even at the end of filtration period when the number of particles in the smallest range (>1.0–3.6 μm) was the highest measured by the flow cytometer and microscopy image analysis. This resistance was reduced further when the river water was biofiltered prior to membrane filtration. This real-time study demonstrates that over time PAC and other particles coming into the reactors through the influents degrade and/or become smaller because of the turbulence caused by continuous aeration below the MF membrane fibers. The number of particles in the reactors with diameters less than 10 μm increased with time, increasing the fouling frequency; however, the presence of PAC further reduced the particle enhanced fouling. The presence of PAC also increased SS degradation by up to 10%. The increased number of bacteria inside the PAC–MF systems did not contribute to the number of membrane fouling. Even though the particle sizes inside the reactors became smaller with time, the gradual increase in net accumulation of SS was also an important factor controlling the performance of the PAC–MF system.

Published by Elsevier B.V.

1. Introduction

During the past decades, the microfiltration (MF) membranes emerged as one of the most reliable, cost-effective and sustainable units for surface water treatment separating macromolecules, bacteria and discrete particles [1–6]. When surface waters are treated using MF membranes, suspended solids (SS) carrying organic materials and different types and sizes of particles cause a rapid flux decline [7–10]. It has been demonstrated that particle sizes change during the filtration process: for high cross-flow velocity and in the presence of aeration, large particles break down into smaller ones [11]. These small particles then cause pore blocking of MF membranes during filtration [12–14] and increase transmembrane pressure (TMP).

If the natural organic matter and microorganism load can be reduced at upstream of the membranes, the extended membrane filtration period can be achieved [15]. This may be accomplished by optimizing the adsorption of organics and microorganisms by powdered activated carbon (PAC) and/or by the use of biological filtration. There have been several discussions about whether PAC itself fouls membranes attenuating membrane permeability/productivity reduction [16,17]. Several reports indicated that the addition of PAC in the membrane process decreased the rate of flux decline [2,18,19]. The addition of PAC sometimes fouls the membrane, depending on the PAC–membrane interaction and the characteristics of the PAC cake layer [3,20,21]. Moreover, it is not

* Corresponding author at: Center for Molecular Discovery and Translational Technology, MSC08 4640, University of New Mexico, Albuquerque, NM 87131, USA. Tel.: +1 505 925 7762; fax: +1 505 272 6995.
E-mail address: taimur2000@hotmail.com (M.Md.T. Khan).

0376-7388/$ – see front matter. Published by Elsevier B.V.
doi:10.1016/j.memsci.2011.01.017

entirely clear how the dynamic particle size changes in a PAC–MF hybrid system affect the rate of membrane fouling or TMP during surface water treatment.

To the best of our knowledge there have been no studies or published reports on the dynamic effects of particle size changes in the presence of PAC on increase in TMP or fouling of MF membranes during long-term treatment of real surface water with variable water quality parameters. The models demonstrating particle-enhanced fouling that have been developed were tested in controlled laboratory environments using particles of unique shapes or even having the same mean particle sizes [22–24], which did not change their sizes during filtration and using unique influent qualities.

Several studies used PAC in the pretreatment step, prior to membrane filtration [21,25–28], or in combination with membrane filtration in the same tank [29,30] to demonstrate the membrane fouling mitigation mechanisms. PAC was combined with MF membrane forming a hybrid system to remove organic compounds [8,17,30–32], 17β-estradiol (an endocrine disrupting compound) [33], disinfection byproducts [30], atrazine [34], and microorganisms like *Norovirus* (27–40 nm in diameter) [35]. However, the performance of the PAC–MF process depends on several factors.

In addition, such questions as how the size changes of PAC and other particles entering the membrane reactor continuously affect the membrane fouling in real filtration systems using real surface water; whether the presence of PAC affects the degradation of SS in influents and reduce their effects on membrane fouling; and how TMP and resistance to filtration change in the presence or absence of biofiltration of the river water are still unanswered. In our previous studies [5,14] on PAC–MF systems for surface water treatment, in which the PAC dose was varied from 0 to 50 g/L of the reactor, we observed that at 40 g of PAC per liter of the reactor, the membrane fouling frequency was the lowest and the removal rate of dissolved organic carbon (DOC) was more than 80%. For this study, we hypothesized that PAC and other particles entering the reactors would be smaller with time because of the aeration under the membrane modules and the presence of PAC would lessen the effect of particle and bacterial enhanced foulings. Moreover, pretreatment of the river water would provide additional benefits. We also expected that the presence of PAC would affect the degradation of SS.

In this study we used the flow cytometer (FCM) to count and distinguish the particles and bacteria in different samples. Cluster composition in aggregation processes of multiple particle species can be dynamically determined using the novel FCM analysis [36,37].

A better understanding of the interactions among the various factors, such as pretreatment of influent, particle size changes and the role of SS in the influents to the reactors on the dynamics of TMP changes, is needed to optimize the performance of hybrid PAC–MF reactors. To provide insight on these interactions, the objectives of this study were to: (1) quantify the combined effect of PAC and pretreatment of influent on MF membrane fouling and the dynamic particle size changes in the reactors, and (2) estimate the accumulation and degradation of SS in the influents to membrane bioreactors.

2. Materials and methods

2.1. Site location and influents to the reactors

This study was conducted at a water treatment plant operated by the Tokyo Metropolitan Authority. The plant is located in the southwest region of Tokyo, about 15 km from the main city, and treats water from the Tama River. The details of this river water quality are mentioned elsewhere [30]. A bench-scale study of the PAC–MF system was performed in this treatment plant using the river water.

In the treatment process, the river water was pumped from the intake point to a series of sedimentation ponds. The water from the secondary sedimentation pond was used as the raw water and fed into two membrane reactors. The raw water was treated with a biofilter, and the effluent of the biofilter was fed into another membrane reactor. The biofilter consisted of a column packed with polypropylene pellets with a length of 5 mm, an inner diameter of 3 mm and an outer diameter of 4 mm. The filtration velocity and residence time of this biofilter were 320 m/d and ~10 min, respectively. Discharges from the biofilter and raw water were stored separately in two 100-L reservoirs and stirred continuously prior to being fed into the reactors.

2.2. Experimental design and operational conditions of the reactors

We operated three reactors (R1, R2, and R3) continuously for 151 days in this study. Each reactor consisted of a hollow-fiber MF membrane module operated in suction mode to maintain constant flux. The polyethylene hydrophilic membrane was made by Mitsubishi Rayon Co., Ltd., Japan. The nominal pore size was 0.1 μm, and the outer and inner diameters were 0.41 mm and 0.27 mm, respectively. Each membrane module had 320 (16 × 20) 120-mm-long fibers, with a combined surface area of 0.05 m^2. The reactors were made of 5-mm-thick polyvinyl chloride plates, and each had an effective volume of 5 L. The membrane modules were cleaned with Milli-Q water when received and soaked in fresh Milli-Q water prior to use. The experiment was carried out under ambient conditions (17–28 °C). The raw water was fed as an influent into reactors R1 and R2, and the effluent from the biofilter (BF) was fed into reactor R3. Fig. 1 shows the entire system and specifies the operational protocol for each reactor.

PAC (coconut shell origin, JWWA K 113-1985, Shirosagi-C, Takeda Chemical Co., Japan) was used as received and the dose was 40 g of PAC per liter of the reactor added into reactors R2 and R3 at the beginning of the operation. Reactor R1 was operated without PAC as a control. The filtration flux was kept at 20.8 L/m^2/h, the flow rate was 25 L/day, and the residence time was 0.2 day. All membrane modules were equipped with a backwash mechanism, which was controlled by electronic valves and timers, providing 2 min of backwashing after 20 min of filtration. Level sensors were used to ensure a constant reactor volume, while suction through the membrane module maintained a constant flow rate. Fresh filtrates were used for backwashing of the membrane modules. The backwash-water reservoirs were cleaned once a week and refilled with fresh filtrate, and the backwashing flux (41.6 L/m^2/h) was twice as high as the filtration flux. The reactors were aerated continuously at 1000 L/m^3/min (5 L/min), and the air was delivered below the fiber modules. Transmembrane pressures (TMP) and backwashing pressures (BWP) were recorded using a data logger.

Once TMP values reached to 50 kPa (0.5 bar), the fouled membrane modules from the reactors were removed and washed with Milli-Q water and a soft brush to remove the foulants from the membrane's surfaces. The resistance to filtration and pure water flux were measured right after physical cleaning of the membrane modules and prior to starting the next cycle of filtration. At the end of the study (on Day-151), the membrane surfaces were chemically cleaned by soaking in 20 mM EDTA and 4% NaOH solutions for 2 h and finally being rinsed with Milli-Q water. The resistance to filtration and pure water flux were also measured after this chemical cleaning of the membrane modules.