WATER RESEARCH 42 (2008) 697–706

Available at www.sciencedirect.com

ScienceDirect

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

Autopsy of high-pressure membranes to compare effectiveness of MF and UF pretreatment in water reclamation

Jeonghwan Kim[a,*], *Francis A. DiGiano*[b], *Roderick D. Reardon*[c]

[a]*Department of Civil and Environmental Engineering, Michigan State University, East Lansing, MI 48823, USA*
[b]*Department of Environmental Sciences and Engineering, University of North Carolina at Chapel Hill, CB 7431, Chapel Hill, NC 27599-7431, USA*
[c]*Carollo Engineers, 1800 Pembrook Drive, Suite 300 Orlando, FL 32810, USA*

ARTICLE INFO

Article history:
Received 31 December 2006
Received in revised form
14 August 2007
Accepted 22 August 2007
Available online 21 September 2007

Keywords:
Secondary effluent
Pretreatment
Membrane autopsy
Specific flux
Polysaccharides
Microfiltration
Ultrafiltration
Reverse osmosis

ABSTRACT

A pilot-plant study was designed to compare the effectiveness of microfiltration (MF) and ultrafiltration (UF) as pretreatment for high-pressure membranes in reclamation of biologically treated wastewater effluent. Granular media, filtered secondary effluent from a full-scale wastewater treatment plant, was fed to MF and UF units that operated in parallel. Each of these filtrates served as the feedwater to two reverse osmosis (RO) units and one nanofiltration (NF) unit that operated in parallel. The decline in specific flux was substantially lower for high-pressure membranes receiving UF than MF pretreatment over the course of each of four pilot plant runs that lasted from 1 to 7 weeks. The removal of organic matter as measured by dissolved organic carbon (DOC) was somewhat higher by UF than MF pretreatment (about 15% by UF compared with 11% by MF). Addition of ferric chloride ahead of the UF unit, but not ahead of the MF unit, may account for this additional removal of organic matter. However, the additional DOC removal appeared insufficient to explain the differential in foulant accumulation between high-pressure membranes receiving UF and MF pretreatment. Extensive autopsy analyses of these high-pressure membranes showed from 35% to 56% less organic carbon on those receiving UF rather than MF pretreatment. A more specific indicator of a differential in organic fouling was the accumulation of polysaccharides and this showed from 27% to 38% less on UF- than on MF-pretreated membranes. Yet another possible source of foulants is inorganic material given that the inorganic and organic weight percentages were nearly equal (56% vs. 44%) on the membrane surface. One specific source was aluminum added for phosphorus removal. Less fouling of high-pressure membranes pretreated by UF than MF could be due to the following: (1) a small, but very important, colloidal fouling fraction may have passed through MF but was rejected by UF pretreatment; (2) organic fouling was not related to organics in either the MF or UF filtrates but rather to organics that are generated *in situ* by microbial activity on the membrane surface; and/or (3) less passage of colloidal Al–P that carried over from secondary wastewater treatment.

---

[*] *Corresponding author.* Tel.: +1 517 355 5155; fax: +1 517 355 0250.
E-mail address: kimjeo21@msu.edu (J. Kim).

0043-1354/$ - see front matter © 2007 Elsevier Ltd. All rights reserved.
doi:10.1016/j.watres.2007.08.042

1. Introduction

While membrane technology offers the potential to increase the value of wastewater reclamation, the occurrence of membrane fouling continues to limit the membrane operation efficiency (Jarusutthirak and Amy, 2002; Decarolis et al., 2001). High-pressure membranes such as nanofiltration (NF) and reverse osmosis (RO) membranes can be fouled easily by colloidal materials and organic matters present at high levels in secondary wastewater effluents (Her et al., 2003; Jarusutthirak et al., 2002; Parameshwaran et al., 2001). Foulants in wastewaters may fall into broad categories of particles, colloids, macromolecules, inorganics and even low molecular weight dissolved organics (Neis and Tiehm, 1997; Adin and Elimelech, 1989; Jarusutthirak and Amy, 2002; Van der Bruggen and Vandecasteele, 2001). The accumulation or adsorption of foulants on the surface or into the membrane matrix results in the loss of membrane performance over time, which results in increasing both capital and operational costs (Jarusutthirak et al., 2002).

The extent of fouling of high-pressure membranes in water reclamation may depend upon the pretreatment processes as well as the chemical formulation of the high-pressure membranes. In the past decade, pretreatment has shifted away from chemical coagulation–sedimentation–granular media filtration and towards low-pressure membrane filtration (Ghayeni et al., 1996; Reardon et al., 2005). The issue that remains unresolved is the choice between microfiltration (MF) or ultrafiltration (UF). Biomass flocs, individual bacterial cells and other particles carried over from the secondary clarifier should be removed by MF. Colloids, high molecular weight soluble microbial products and extracellular polymeric substances (EPS) generated by microbial activity (Jarusutthirak et al., 2002; Fonseca et al., 2003; Uhl et al., 2003) can be removed by UF, but the extent depends upon the molecular weight cut-off (MWCO) of the membrane.

A direct comparison was made of the effectiveness of MF and UF as pretreatment for high-pressure membranes in a recent pilot-plant study of water reclamation (Reardon et al., 2007). The feedwater to the pilot plant was granular media, filtered secondary effluent from the North Buffalo Wastewater Reclamation Facility in Greensboro, NC. The decline in specific flux (flux/pressure) of three commercially available, high-pressure membranes that followed each pretreatment was the indicator of fouling. Statistical analysis showed that the decline in specific flux was lower for each of the three high-pressure membranes that received UF than MF pretreatment. For either type of pretreatment, the differences in rejection and fouling rate among the three high-pressure membranes were relatively small.

The objective in this study was to use data collected from membrane autopsies at the end of the pilot-plant study at the North Buffalo Wastewater Reclamation Facility to explain less fouling of high-pressure membranes that received UF pretreatment. Autopsy measurements included microbial population, specific inorganic ions and several surrogates used to measure organic foulants (organic carbon, polysaccharides and molecular weight fraction).

2. Methods

2.1. Membrane pilot plant for wastewater reclamation

The process flow diagram for the membrane pilot plant is shown in Fig. 1. Granular media, filtered secondary effluent from the North Buffalo Wastewater Reclamation Facility (Greensboro, NC, USA), was pumped at a rate of about 100 gallon per minute (gpm) into a common tank. Feedwater was pumped from this tank to both MF and UF units. Filtrate from the MF (CMF 6M10C, US Filter MEMCOR) and UF (HYDRACap 60, Hydranautics) modules passed into the break tank from which the filtrate was pumped into high-pressure membranes as feedwater. Each type of high-pressure membrane was housed in two pressure vessels that operated in series with each pressure vessel containing three membrane elements. The total water recovery from these elements was 50%. Trains of low- and high-pressure membrane units were operated as MF–RO and UF–RO, although one of three high-pressure membranes was classified as NF. Results from membrane pilot plant operation between April and August in 2005 are discussed in this paper.

General characteristics of high-pressure membranes used in this pilot-plant study are shown in Table 1. All three high-pressure membranes are spiral wound elements of thin film composite materials. The Hydranautics ESPA2 and TriSep X20 are RO membranes whereas the Dow/Film Tech NF90 is an NF membrane. The MWCO ranged from 50 to 500 Da, with the highest for the NF membrane. All three membranes are formed from polyamide that imparts a negative surface charge. However, the residual amino group in the polyamide-urea structure of the Trisep X-20 membrane, in contrast to the carboxylic acid group in the polyamide structure of the Dow/Film Tec NF-90 and the ESPA2 membranes, may suppress the negative charge.

Differences in rate of decline in specific flux among the three high-pressure membranes during the pilot plant study, when normalized by the initial specific flux (lower for Trisep X20 membrane than either the ESPA2 or NF90 membranes), were relatively small and difficult to quantify (Reardon et al. 2007). Differences in foulant accumulation among the three high-pressure membranes were also difficult to discern from the autopsy results. For this reason, the discussion will be restricted to differences in foulant accumulation resulting from the pretreatment selection (MF vs. UF) for each high-pressure membrane.

2.2. Design and operation of low-pressure modules

Important characteristics of the MF and UF modules are given in Table 2. As stated by the manufacturers, the nominal pore size of the MF hollow fibers was 0.2 μm and the MWCO of the UF hollow fibers was 150 kDa. Using an available estimating method, the MWCO of the UF membrane corresponds to a nominal pore size of about 0.01 μm (Howe and Clark., 2002). Both the MF and UF modules operated in the dead-end mode. However, the flow pattern for MF was outside-in while that for the UF module inside-out. Another important difference is that 4 mg/L of $FeCl_3$ is added to the feedwater of the UF