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Human adenovirus removal by hollow fiber membranes: Effect of membrane fouling by suspended and dissolved matter

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ABSTRACT

In this study, we evaluated removal of human adenovirus 40 by hollow fiber ultrafiltration (UF, $d_{pore}=0.04$ μm) and microfiltration (MF1, $d_{pore}=0.22$ μm; MF2, $d_{pore}=0.45$ μm) membranes in the presence of suspended and dissolved foulants: SiO_2 microspheres and Aldrich humic acid (HA). Average removal of adenovirus from DI water by UF, MF1 and MF2 membranes was 2.3 log, 0.7 log and 0.7 log, respectively. The observed decrease in adenovirus removal due to SiO_2 fouling (δLRV of −1.2 and −0.2 for UF and MF1 respectively) was attributed to the cake-enhanced accumulation of viruses at the membrane surface. In contrast, fouling by HA led to higher virus removals (δLRV of 0.8 and 1.2 for UF and MF1, respectively), which was attributed to pore blockage by HA. In experiments with MF2 membrane, neither HA nor SiO_2 had significant effects on adenovirus removal. The results indicate that the extent of fouling is not a reliable predictor of adenovirus removal. Instead, feed water composition and membrane pore size together govern virus removal with fouling mechanisms playing a mediating role: pore blockage improves removal while cake formation can either increase or decrease removal depending on cake properties.

1. Introduction

More than 150 types of enteric viruses have been found in contaminated waters [1–4]. Because of their low infectious dose and long survival in the environment viruses pose a considerable threat to human health. Human adenovirus (HAdV) is one of enteric viruses on the U.S. EPA's contaminant candidate list. Various species of HAdV can cause a range of diseases [5,6]; for example, HAdV-F is the known etiological agent of gastroenteritis while HAdV-B and HAdV-E may lead to acute respiratory diseases. A double-strand DNA virus, HAdV is one of largest virions ranging from 70 to 140 nm in size [7]. What makes HAdV particularly problematic is its resistance to UV disinfection [8–10]; for example, UV dosages as high as 217.1 mW/cm^2 are required for 99.99% deactivation of HAdV 40 [11]. The large size of HAdV and its resistance to UV light point to the promise of membrane filters as a treatment process for removing this virus from water.

Although some pathogen removal occurs during wastewater treatment, even advanced technologies may not provide an absolute barrier for viruses. Indeed, recent studies report the presence of human enteric viruses in the effluents of state-of-the art treatment wastewater facilities such as membrane bioreactors (MBR) plants [12,13] and drinking water treatment plants [14,15]. MBRs can achieve high and stable removal efficiency for chemical oxygen demand [16,17], biochemical oxygen demand [18], nitrogen [18,19], phosphorus [20], and coliform bacteria [21]. Virus removal, however, has not been a criterion in the design and operation of MBR plants. In fact, some MBRs employ membranes with the nominal pore size larger than the size of a typical virus (20–200 nm), in which case membrane fouling and cleaning may control virus removal.

Multiple studies evaluated virus removal as a function of membrane and feed properties; some of this work has employed bacteriophages as human virus surrogates. Langlet et al. showed an increase in MS2 phage removal with a decrease in the membrane pore size [22]. Lu et al. found a strong linear correlation between MS2 log removal and permeability of ultrafilters in the presence of foulants in the feed: on average, fouling increased MS2 removal by 1.23 logs [23]. Working with the same type of phage, Jacangelo et al. reported that membrane fouling contributed up to 2.6 logs removal of MS-2, which was much more significant in comparison with physical sieving/adsorption (0.3 log) and cake layer formation (0.1–0.5 log) [24]. Wu et al. [25] reported that gel layer contributed to the removal of somatic coliphage removal, more so at a higher permeate flux. High removals of T4 coliphage have been reported and partly attributed to the formation of a cake layer formed on membrane surface [26–29]. Shirasaki et al. carried out filtration experiments in a coagulation–MF system and concluded that irreversible fouling played a more important role than reversible fouling in enhancing virus removal [30]. Farahbakhsh and Smith investigated coliphage

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removal from secondary effluent of wastewater treatment plant by microfiltration membrane [31] and reported that fouled membranes rejected viruses more effectively. Composition of the feed water (pH, ionic strength, the presence of divalent actions and organic matter) and pretreatment were suggested as key factors governing virus removal [32–37].

To our knowledge, there have been only six studies on adenovirus removal by membranes with all this work performed in the context of MBR treatment. Sedmak et al. reported the presence of HAdV in Milwaukee's Jones Island wastewater treatment plant (WWTP) effluent although in a much smaller fraction of samples and much lower titers than in the influent [14]. Albinana-Gimenez et al. reported sporadic qPCR-positive but PFU-negative results in the effluent from drinking water treatment plants [15]; in contrast, culturable HAdV in MBR effluent was measured in effluents of each of 10 conventional WWTPs sampled by Hewitt et al. [38].) Kuo et al. showed that HAdV species A, C, and F were removed only partially in the Traverse City MBR WWTP and showed that with the average HAdV removal of 5.0 ± 0.6 logs over the 8 month long study, the effluent contained on average $\sim 10^3$ HAdV particles/L [12]. In their study of enteric virus removal in conventional WWTPs and microfiltration MBR WWTPs (equipped with Kubota membranes), Francy et al. showed that HAdV was detected by q-PCR in a subset of MBR effluent samples both before and after UV disinfection [39]. In a survey of virus removal in nine MBR WWTPs employing different kinds of membranes (tubular, hollow fiber and flat sheet; MF and UF), Hirani et al. reported that adenoviruses were detected in effluents of all MBR facilities [40]; this result was consistent with the findings by Kuo et al. [12] and was particularly striking because enteroviruses, rotaviruses and hepatitis A viruses were absent in all samples. The authors tentatively attributed this finding to the fact that HAdV concentration in the influent is typically very high and concluded that "additional research and risk assessment on the presence of adenovirus in MBR effluents is warranted." In a follow-up study with four different membrane systems, these authors showed that adenoviruses were always detected in MBR filtrate samples by PCR regardless of whether the membrane was breached (effluent turbidity > 0.5 NTU) or cleaned (0.2% NaClO) [41].

The objective of the present work was to elucidate mechanisms of HAdV removal by membranes in the presence of foulants in the feed. To facilitate mechanistic insights, we employed two well characterized model foulants (humic acid and silica particles), three commercially available hollow fiber membranes (with pore sizes typical for membranes used in MBRs), and filtration conditions that matched, to the extent possible, the protocol used at full-scale MBR facilities (i.e. constant flux regime and aeration).

2. Materials and methods

2.1. Cell culture experiment and virus incubation

A549 cell line has been suggested as an efficient cell line for HAdV [42,43], and it was selected in this study. A549 cells (ATCC, cell passage) were incubated at 37 °C with growth medium (minimum essential medium with 10% fetal bovine serum, L-glutamine, Earle's salts) until confluence of cell layer reached 90%. Used media was discarded from the flask, and HAdV 40 was added and incubated at 37 °C with growth media (2% fetal bovine serum) until cytopathic effect became apparent. In order to isolate viruses from cell debris, virus suspension was centrifuged at $400 \times g$ for 4 min, and then filtered through 0.22 μm syringe filter (Millipore). Filtered virus stock suspension had HAdV concentration of approximately 10^{10} copies/mL and was stored at −80 °C before use.

Table 1
Characteristics of hollow fiber membranes.

	Membrane type		
Notation	UF	MF1	MF2
Manufacturer	General Electric	Shenzhen Youber Technology.	
Material	polyvinylidene fluoride		
Nominal pore size, μm	0.04	0.22	0.45
Outer diameter, mm	2.0	1.3	

2.2. Membrane preparation

Three types of hollow fiber membranes were used in this study. The characteristics of the membranes are shown in Table 1. The hollow fibers were cut into 80 cm long segments and assembled by looping and potting them in a short (~10 cm) piece of 1/2″ ID PTFE tubing using an adhesive (Loctite). Each membrane bundle (12 loops of 0.45 μm and 0.22 μm membranes and 8 loops of 0.04 μm membranes) had a total membrane surface area of ~ 300 cm^2. After the adhesive dried, membranes were soaked in deionized (DI) water for at least 24 h before use.

2.3. Foulant preparation and particle size

Silica microspheres and humic acid (HA) were selected as model foulants. According to the manufacturer, the average particle size of SiO_2 spheres (99.998% purity, Nanostructured & Amorphous Materials) was in the 1–3.5 μm range. To prepare a feed suspension with silica, SiO_2 particles were added to 0.5 L of DI water, mixed for 1 h and then added to the feed tank. To prepare a feed suspension with HA, 12 g of HA (Aldrich) were added into 4 L of DI water in an amber jar and the pH was adjusted to 8. The solution was mixed using a magnetic stir bar for 72 h, and then filtered through 0.45 μm membrane filters (Millipore). Filtered HA solution was stored at 4 °C until use. Total organic carbon (TOC) content of the feed water was measured using TOC analyzer (OI Analytical). Particle size distribution in the stock was measured using Mastersizer 2000 (Malvern).

2.4. Membrane filtration experiment

The schematic of the experimental unit is shown in Fig. 1. The total volume of the reactor was 25 L. Diffusers were placed at the bottom of the feed tank to supply air and mix the feed water. Peristaltic digital pump (model 07523-80, MasterFlex L/S) was used to apply transmembrane pressure. Permeate flow rate and transmembrane pressure were measured using a digital flow meter (model 106-4-C-T4-C10, McMillan) and digital pressure sensor (Cole-Parmer, 68075-00), respectively. A LabView code was developed to record readings from the flowrate and pressure sensors and to control the flow rate of the pump.

Three experiments with feeds of different compositions were carried out with each type of hollow fiber membranes. Each experiment included 4 stages:

1. Stage 1 (duration = 1 h). The feed tank, with air diffusers on the bottom, was filled with 18 L of DI water and 10 mL of HAdV 40 stock suspension was added to the DI water in the tank. Averaged over all experiments, the initial feed concentration of HAdV 40 in the tank was 7.03 ± 0.32 (see Supplementary material, Fig. S2, for feed concentration of HAdV 40 as a function of time). The pH of the feed was adjusted to 7.
2. Stage 2 (duration = 6 h). The transmembrane pressure was applied and filtration was carried out in a constant flux regime ($Q = 50$ mL/min; $j = 2.78 \times 10^{-5}$ m/s). Samples of feed and