Variations in backwash efficiency during colloidal filtration of hollow-fiber microfiltration membranes

Seungkwan Hong\textsuperscript{a*}, Praveen Krishna\textsuperscript{b}, Colin Hobbs\textsuperscript{b}, Dohee Kim\textsuperscript{c}, Jaeweon Cho\textsuperscript{c}

\textsuperscript{a}Civil and Environmental Engineering Department, Korea University, 1, 5-ka, Anam-dong, Songbuk-ku, Seoul, 136-701, Korea
\textsuperscript{b}Department of Civil and Environmental Engineering, University of Central Florida, PO Box 162450, Orlando, Florida 32816-2450, USA
\textsuperscript{c}Department of Environmental Science and Engineering, Kwangju Institute of Science and Technology, 1 Oryong-dong, Puk-gu, Gwangju, 500-712, South Korea

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Abstract

A series of filtration experiments was performed systematically to investigate physical and chemical factors affecting the efficiency of backwashing during microfiltration of colloidal suspensions. In this study, all experiments were conducted in dead-end filtration mode utilizing an outside-in, hollow-fiber module with a nominal pore size of 0.1 µm. Silica particles (mean diameter = 0.14 µm) were used as model colloids. Using a flux decline model based on the Happel’s cell for the hydraulic resistance of the particle layer, the cake structure was determined from experimental fouling data and then correlated to backwash efficiency. Modeling of experimental data revealed no noticeable changes in cake layer structure when feed particle concentration and operating pressure increased. Specifically, the packing density of the cake layer (1-cake porosity) in the cake layer ranged from 0.66 to 0.67, which corresponds well to random packing density. However, the particle packing density increased drastically with ionic strength. The results of backwashing experiments demonstrated that the efficiency of backwashing decreased significantly with increasing solution ionic strength, while backwash efficiency did not vary when particle concentration and operating pressure increased. This finding suggests that backwash efficiency is closely related to the structure of the cake layer formed during particle filtration. More densely packed cake layers were formed under high ionic strength, and consequently less flux was recovered per given backwash volume during backwashing.

Keywords: Colloidal fouling; Microfiltration; Backwashing efficiency; Cake layer structure; Water treatment

*Corresponding author.

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1. Introduction

The use of size-exclusion membrane technologies such as microfiltration (MF) and ultrafiltration (UF) has increased dramatically over recent years [1], and MF/UF membranes are now commonplace in numerous industrial processes including wastewater treatment [2–4] and drinking water treatment [5,6]. They present a physical barrier to the suspended particles in the feed stream, whereby all particles larger than the pore are retained on the feed side of the membrane. The retained particles, however, accumulate on the surface of the membrane and increase the resistance to water flow across the membrane. As a result, MF/UF membranes must be periodically backwashed by reversing the direction of flow through the membrane to remove the deposited particles. However, backwashing typically recovers only a portion of productivity lost through operation, which results in membrane fouling (i.e., irreversible productivity loss) as shown in Fig. 1. The productivity loss due to fouling can be restored only by aggressive chemical cleaning, which significantly increases operating costs.

The effective control of membrane fouling in MF/UF processes is largely dependent on the mode and efficiency of backwashing. Several pilot-scale studies demonstrated that an increase in backwash frequency (i.e., shorter operation times between backwash cycles) and duration significantly reduced membrane fouling [3,4,7–9]. Other studies investigated variations on the methods of backwashing, such as air sparging [10] and backpulsing [11]. However, there are no systematic studies in the literature which investigated the relationship between various chemical and physical operating conditions and backwashing efficiency.

In this study it is hypothesized that backwashing efficiency can be affected by the structure of particle cake layer formed on the membrane surface. Considering the relative sizes of the membrane pore \(d_{\text{pore}}\) and the colloidal particle \(d_{\text{particle}}\), three primary modes of colloidal fouling exist in MF/UF processes (Fig. 2): adsorption [12–14], pore blocking [15–18], and deposition of a cake layer [1,19,20]. Among these mechanisms, cake formation is considered to be the most dominant mode of colloidal fouling, since in a normal membrane filtration, the mean diameter of membrane pores is selected in such a way that a majority of the particles to be separated are larger than the pore size. For dead-end filtration processes, the cake layer grows infinitely [21], while during cross flow filtration, the growth of the cake layer is limited by the tangential fluid flow in the module [22,23].

There has been a variety of studies pertaining to elucidation of the structure of the cake layer under the influence of hydrodynamic and colloidal interactions. The hydrodynamic interactions among the particles retained in the cake layer have been evaluated by the Happel’s cell model, which incorporates the influence of the neighboring particles on the hydrodynamic drag force [24,25]. This approach has often been suggested as a substitute for the Kozeny–Carman equation for evaluating the specific resistance of cake layers. Aside from hydrodynamic interactions, several recent studies have attempted directly to incorporate colloidal interactions in predicting the
structure and permeability of the cake layer deposits [26–29]. In such studies, the influence of particle charge, background electrolyte concentration, and other physicochemical conditions was experimentally assessed and/or theoretically quantified to determine the cake layer structure.

The purpose of this study was to examine the effect of feed water quality and operational parameters on the efficiency of backwashing. In this study, a series of fouling experiments was first performed under various operating conditions. The primary operating parameters varied throughout the experiments were particle concentration, operating pressure and solution ionic strength. A theoretical model for flux decline due to cake formation was evaluated and utilized to determine the structure of the cake layer formed during fouling experiments. Following each fouling experiment, the membrane was backwashed in order to relate the backwash efficiency to both physical and chemical parameters. Finally, a theoretical value for the particle packing density of the cake layer calculated from the model was correlated to the backwash efficiency in order to elucidate the effect of cake structure on the efficiencies of backwashing.

2. Experimental

2.1. Colloidal particles

Silica (SiO$_2$) particles from Nissan Chemical Industries (Houston, TX) were used as model colloids for all of the fouling and backwashing experiments. The particles were received as a stable concentrated (40.7% by weight) aqueous suspension at an alkaline pH. The manufacturer’s certificate reported a mean particle diameter of 0.10±0.03 µm (as determined by the centrifugal method) and a specific gravity of 1.301 at 20°C. The size and shape of these model colloids were further verified by a scanning electron microscope (JEOL Model 400, JEOL, Peabody, MA) and by dynamic light scattering (Nicomp Model 380, Particle Sizing Systems, Santa Barbara, CA). The SEM images and DLS analyses revealed that the model silica particles are monodispersed with a mean particle size of 140 nm. Lastly, the zeta potential of the colloidal silica was determined from electrophoretic mobility measurements (Zeta PALS, Brookhaven Instruments, NY). The results showed the zeta potentials of silica particles to be in the range of ~27 to ~30 mV at pH 8 and 10$^{-2}$ M NaCl, which were the solution
environments employed in the majority of fouling and backwash experiments. More detailed properties of these silica particles are well documented in a recent paper by Vrijenhoek et al. [30].

2.2. Microfiltration membranes

All experiments conducted during this study utilized a bench-scale, outside-in, hollow-fine fiber, MF module (SK Chemicals, Seoul, South Korea). Manufacturer’s specifications revealed the following physical characteristics of the membrane: nominal pore size of 0.1 µm, inside fiber diameter of 0.7 mm (2.3×10⁻³ ft), outside fiber diameter of 1.0 mm (3.3×10⁻³ ft), and a fiber length of 520 mm (1.7 ft). Containing a total of 150 hollow-fine fibers, the MF module provided approximately 0.25 m² (2.7 ft²) of membrane area. The average specific flux of this membrane was estimated at 3.55 ± 0.20 lmh/kPa (14.43 ± 0.83 gfd/psi) under given operating conditions.

Prior to all experiments, the operational integrity of the membrane module was verified. This was accomplished by the filtration of a high concentration (0.05% v/v) colloidal silica suspension. During filtration, feed and permeate samples were collected and analyzed for turbidity and total suspended solids (TSS). Results from these tests were then compared to results obtained through the filtration of a DI (blank) water sample to identify any defects in the membrane module.

2.3. Standards and reagents

All solutions were prepared with ACS-grade NaHCO₃ and NaCl (Fisher Scientific, Pittsburgh, PA). These salts were dissolved in DI water (LD5A and MegaPure, Barnstead/Thermolyne, Dubuque, IO). Adjustments in pH, for both zeta potential measurements and fouling studies, were made with ACS-grade HCl. Lastly, all cleaning solutions were made with USP-grade sodium hydroxide and citric acid dissolved in DI water.

2.4. Bench-scale membrane filtration unit

The colloidal suspensions were prepared and stored in a magnetically stirred high-density polyethylene 20 L (5.3 gal) feed reservoir. The temperature of this suspension was maintained at 20°C (68°F) by a Neslab CFT-33 (Portsmouth, NH) digital refrigerated recirculator. The feed suspension was delivered to the MF module by a 6.83 lpm (1.8 gpm) constant flow diaphragm pump (Hydracell, Wanner Engineering, Minneapolis, MN) with a maximum pressure of 3,447 kPa (500 psi). Initial operating conditions (e.g., filtrate flux and cross flow velocity) were set and maintained through the careful manipulation of feed, concentrate, and bypass needle valves (Swagelok, Solon, OH). Feed pressure was monitored by an analog pressure gauge (Dresser Industries, Stratford, CT). Concentrate and filtrate flows were measured both by an in-line flowmeter (Blue-White Industries, Westminster, CA) and by the timed collection of filtrate in a graduated cylinder.

2.5. Sequence of fouling and backwash experiments

Prior to each fouling experiment, the bench-scale MF unit was thoroughly cleaned by recirculating sodium hydroxide (pH 11) and citric acid solutions (pH 3) for a minimum of 1 h. In addition, the module was backwashed with these solutions at a pressure of 68.9 kPa (10 psi). After chemical cleaning was completed, the system was rinsed and flushed with DI water.

An initial clean water test was performed to determine membrane productivity prior to each fouling experiment. Each clean water test was conducted in a dead-end mode of operation at a feed pressure of 41.4 kPa (6 psi) with a background electrolyte solution identical to that which would be used for the ensuing fouling study (e.g., 10⁻³ M NaHCO₃ and 10⁻² M NaCl). A total of 5 L of filtrate was collected, and a stopwatch was
used to measure the collection times associated with 1, 2, 3, 4, and 5 L of filtrate accumulated.

Following the initial clean water test, feed, concentrate, and bypass valves were manipulated to achieve the desired initial operating conditions for the fouling study. Once stable operation was attained, the predetermined volume of concentrated silica particles was added to the feed solution to achieve the desired particle concentration. Immediately following the addition of silica particles, 5 L of filtrate was collected, and collection times were measured and recorded for 1, 2, 3, 4, and 5 L.

Once the fouling study was completed, 1 L of DI water was backwashed through the MF module at a pressure of 68.9 kPa (10 psi), and a final clean water test was conducted. Similar to the initial clean water test, the final clean water test was conducted in a dead-end mode of operation at a feed pressure of 41.4 kPa (6 psi) with a background electrolyte solution identical to that which was used for the previous fouling study. Again, a total of 5 L of filtrate was collected, and a stopwatch was used to measure the collection times associated with 1, 2, 3, 4, and 5 L.

Feed and filtrate samples for each fouling study were collected and analyzed for conductivity, pH, and turbidity. Conductivity and pH measurements were made with an Accumet AR-50 conductivity and pH meter (Fischer Scientific, Pittsburgh, PA), and turbidity was determined using a Hach Ratio Turbidimeter (Loveland, CO).

2.6. Evaluation of flux decline and backwash efficiency

In order to compare multiple data sets obtained under various experimental conditions, it was necessary to analyze all experiments on a dimensionless basis. Two parameters of particular interest throughout this study were the normalized flux \( J_n \) and the backwash efficiency \( \eta \). The normalized flux was calculated from

\[
J_n = \frac{J_w}{J_0}
\]

where \( J_w \) is the flux after the collection of 5 L of filtrate and \( J_0 \) the initial filtrate flux. Similarly, the backwash efficiency \( \eta \) was estimated by

\[
\eta = \frac{t_i}{t_f}
\]

where \( t_i \) is the time required to collect 5 L of filtrate during the initial clean water test and \( t_f \) the time required to collect the same 5 L during the final clean water test.

3. Results and discussion

3.1. Cake layer structure

In pressure-driven membrane filtration of colloidal suspensions, particles are transported to the membrane surface by the filtrate flow, which results in the formation of a cake layer on the membrane surface. Particle accumulation in the cake layer provides an additional resistance to filtrate flow and, hence, reduces flux. Resulting pressure drops in the membrane system can be expressed as

\[
\Delta P = \Delta P_m + \Delta P_c
\]

As shown in Eq. (3), the applied (transmembrane) pressure drop \( \Delta P \) is equal to the sum of the pressure drops across the membrane \( \Delta P_m \) and the cake layer \( \Delta P_c \).

The pressure drop across the membrane is simply the product of membrane resistance \( R_m \) and filtrate flux \( J_w \):

\[
\Delta P_m = J_w R_m
\]

The pressure drop in the cake layer is associated with the frictional drag resulting from the flow of
filtrate through the dense layer of accumulated particles:

$$\Delta P_e = \frac{kT}{D} A_s(\theta) J_w M_c$$  \hspace{1cm} (5)

Here, $kT/D (=6\pi\mu a_p)$ is the frictional drag coefficient, $k$ is the Boltzmann constant, $T$ is absolute temperature, $D$ is the particle diffusion coefficient, $\mu$ is the solvent viscosity, $a_p$ is the particle radius, and $M_c$ is the total number of particles (per unit area) accumulated in the cake layer. The $A_s(\theta)$ is a correction function accounting for the effect of neighboring retained particles and can be evaluated from Happel’s cell model [24]:

$$A_s = \frac{1 + \frac{2}{3} \theta^5}{1 - \frac{2}{3} \theta + \frac{1}{2} \theta^5 - \theta^6}$$  \hspace{1cm} (6)

where $\theta = (1 - \epsilon)^{1/3}$ is a porosity-dependent variable, with $\epsilon$ being the porosity of the cake layer of accumulated particles. As shown in Eqs. (4) and (5), the pressure drop across the cake layer is primarily influenced by the structure of the cake layer, as well as particle size and concentration.

The flux decline observed during the membrane filtration of colloidal suspensions can be estimated based on Happel’s cell model for the hydraulic resistance of the particle cake layer formed:

$$J_w = \frac{J_0}{2 \pi a_p D R_m J_w A} \left[1 + \frac{3 kT A_s(\theta) \Delta P C_0}{2 \pi a_p^3 D R_m J_w A} V \right]^{1/2}$$  \hspace{1cm} (7)

Here $J_0$ is the initial filtrate flux, $C_0$ is the bulk (feed) particle concentration, $A$ is the membrane area, and $V$ is the filtrate volume. A detailed theoretical development is well presented in a paper by Hong et al. [25]. By utilizing Eq. (6), structural characteristics of the cake layer formed under various operating conditions can be determined from filtration experiments. Specifically, the correction function, $A_s(\theta)$, is estimated first by fitting flux decline experimental data and then the particle packing density (i.e., 1-cake porosity) is calculated based on the Happel cell model.

3.2. Membrane integrity and particle removal

The results of the membrane integrity tests clearly demonstrated that the membrane fibers were intact and undamaged. Measurements of feed samples (0.05% v/v) revealed that the feed suspension had an average turbidity of 119 NTU. The turbidity was completely removed by filtration as the average turbidity of the permeate samples was 0.02 NTU. These results were further supported by data obtained through TSS measurements. The average TSS of the silica feed suspension was determined to be 0.613 g/L, with a standard deviation of 0.038 g/L. Once again, the silica particles were completely rejected by the membrane as silica permeate samples had insignificant TSS concentrations when compared to both DI feed and DI permeate samples. The TSS values of these three samples were statistically indistinguishable based on hypothesis testing at a 5% level.

Similar to the results of integrity testing, filtrate turbidity was always below the detection limit (0.02 NTU) regardless of experimental conditions employed in this study. This is due to the fact that the particles used were larger than the membrane pores (approximately 0.14 µm and 0.10 µm, respectively). Thus, all particles were retained on the feed side of the membrane and formed a particle cake layer on the membrane surface as shown in Fig. 2.

3.3. Particle loading

As expected from Eq. (7), the extent of flux decline in dead-end filtration of colloidal suspensions was directly related to cumulative particle loading to the membrane system ($C_0 \times V$). In this
study particle volume concentrations were varied from 0.005% to 0.045% under identical physical and chemical operating conditions. Results of these tests are presented in Fig. 3. As shown, the extent of membrane fouling increased with the concentration of colloidal particles. Specifically, after the filtration of 5 L, the averaged normalized flux values were 0.87, 0.79, 0.62, and 0.56 for feed particle concentrations of 0.005%, 0.015%, 0.030%, and 0.045%, respectively. This observation was explained by the concept of mass loading. As more particles were transported to the membrane surface, the resulting cake layer grew and provided greater resistance to filtrate flow and ultimately a more significant decline in flux.

Utilizing the procedures previously described, the structure of each cake layer formed during filtration experiments was determined. These results are presented in Fig. 4. As shown, the structure of the cake layer for each particle concentration was relatively consistent, with particle packing density factors ranging from 0.66 to 0.67, which correspond well to random packing density factors [24,25]. This observation suggested that the structure of the cake layer was independent of particle concentration, which is not surprising as all of the operating parameters remained constant except particle concentration, which only affected the thickness of the cake layer, not its structure.

In addition to fouling experiments, backwashing studies were also conducted to determine the effectiveness of the backwashing under various feed particle concentrations. The results of these experiments are summarized also in Fig. 4. The average efficiency of all backwashing procedures ranged from 0.969 to 0.985; however, no clear correlation was observed between particle concentration and backwash efficiency considering variations, although backwash efficiency decreased slightly at high particle concentrations. The apparent lack of dependence of backwash efficiency on particle concentration was in accordance with no changes in particle packing density with feed particle concentrations. Thus, under the given range of particle loading to the
membrane systems, it was hypothesized that the backwash efficiency was more closely related to cake structure than particle mass accumulated on the membrane surface.

3.4. Operating pressure

Filtration experiments were also conducted to determine the effect of operating pressure (or initial flux) on particle fouling. It should be noted that, unlike the previous set of experiments, particle loading per unit membrane surface area was held constant throughout this series of experiments. Thus, the number of particles transported to the surface of the membrane at any given volume of filtrate would be the same, regardless of the initial value of operating pressure. Results are presented for five different values of operating pressures as shown in Fig. 5. After the collection of 5 L of filtrate, operating pressure of 20.7, 34.5, 41.4, 65.6 and 41.4 kPa resulted in average normalized flux values of 0.920, 0.916, 0.867, 0.915, and 0.912, respectively, indicating that particle fouling did not vary significantly with increasing operating pressure, with an exception of 41.4 kPa. It may be expected that, as the operating pressure is increased, the force that transports suspended particles to the membrane surface is also increased, which would cause the formation of a more densely packed and hydraulically resistant cake layer. However, the effect of filtrate drag was not significant under the operating pressure range investigated in this study as shown in the Fig. 5.

Cake layer structures were again determined for each set of experimental conditions. Fig. 6 presents these results. As shown, the structure of the cake layer for each operating pressure was relatively consistent, with particle packing density factors ranging from 0.66 to 0.67. Once again, these values are consistent with accepted values of random packing density factors for rigid spherical particles. Since all data points in this figure are within one standard deviation, it may be concluded that the density of the cake structure did not significantly change with increasing operating pressure. While many studies have...
shown a direct relationship between operating pressure and cake layer density, it is believed that the low pressures used throughout these experiments did not allow for the clear observation of this phenomenon from a statistical standpoint. However, it should be noted that a direct relationship between operating pressure and cake layer density was observed for average data points.

Upon completion of each fouling run, a backwashing experiment was performed to evaluate the reversibility of fouling experienced during each filtration study. As shown in Fig. 6, the average efficiency of the backwashing procedure for each operating pressure value was relatively consistent, with efficiencies ranging from 0.98 to 0.99. This finding was attributed to no significant variation in cake layer structure under given pressure range investigated, again suggesting that backwashing efficiency may be closely related to the structure of the cake layer formed during filtration.

3.5. Ionic strength

The effect of solution chemistry, specifically ionic strength, on the fouling of a hollow-fiber MF module was also investigated. A total of three ionic strengths, spanning two orders of magnitude, were tested: 0.001 M, 0.01 M, and 0.1 M NaCl. Once again, the mass loading of particles on the surface of the membrane was held constant throughout all experiments conducted in this series, as the particle concentration was fixed at 0.005%. Furthermore, the operating pressure was set at 41.4 kPa for all experimental runs. The results of these experiments are shown in Fig. 7 and clearly showed that membrane fouling became more severe as the ionic strength of the solution was increased. The average normalized flux values after 5 L of filtrate was collected were 0.97, 0.87, and 0.81, for ionic strengths of 0.001 M, 0.01 M, and 0.1 M NaCl, respectively.

According to the classic theory of colloidal interactions, the magnitude of the electrical double layer (EDL) repulsion is inversely proportional to the ionic strength of the solution. The reduction in repulsive forces resulted in the formation of a more densely packed cake layer on the surface of the membrane, which presented a greater resistance to filtrate flow. This finding was further validated through the determination of cake density at various ionic strengths based on the model previously described in Section 3.1. The modeling results are summarized in Fig. 8 and clearly demonstrated that the density of the cake layer increased with the ionic strength of the particle suspension. However, it should be noted that the packing density for the 0.1 M NaCl experiment was estimated to be 0.76 which is higher than theoretical values. For hard spherical particles, theoretical particle volume fractions used in the literature for the cake layer ranged...
Fig. 8. Correlation between particle packing density (1-cake porosity) and backwash efficiency of hollow-fiber MF membranes under various solution ionic strength. Experimental conditions identical to those described in Fig. 7.

from 0.64 to 0.72, which correspond to random close packed and hexagonal close packed cake structures, respectively [24,25,31]. The high packing density calculated for high ionic strength is likely attributed to additional fouling mechanisms (i.e., adsorption and pore blockage), which were not accounted for by the model.

Lastly, backwashing studies were performed to investigate if a relationship exists between the ionic strength of the solution and the efficiency of backwashing events. The results indicated that the average efficiency of backwashing events decreased as the ionic strength of the feed solution increased, as presented in Fig. 8. The average backwash efficiency for solution ionic strengths of 0.001 M, 0.01 M, and 0.1 M NaCl were estimated at 0.986, 0.982, and 0.962, respectively. These findings further suggest that the efficiency of backwashing events is a function of the structure of the cake layer formed during the filtration process. Specifically, the efficiency of backwashing decreased with an increase in the density of the cake layer. At high ionic concentrations, a more densely packed cake layer was formed due to repressed electrostatic interactions among particles, and consequently less flux was recovered per given backwash volume. Particle adsorption and/or pore blockage, as evidenced by the higher packing densities calculated, may also contribute to the reduced backwash efficiencies observed for the high ionic solutions.

4. Conclusions

Primary inferences from this research are summarized as follows:

1. An increase in particle concentration resulted in a reduced normalized flux under identical operating conditions, which was attributed to the formation of a thicker cake layer caused by an increase in particle loading. Despite varying degrees of cake thickness, the structure of the cake layer, as determined by the Happel’s cell model, did not vary with particle concentration. In addition, the efficiency of the backwashing procedure remained relatively constant throughout all particle concentration experiments, suggesting a close relationship between backwash efficiency and cake structure.

2. Increasing operating pressure under identical particle loading did not cause severe flux decline. The particle packing density remained constant at a random packing density (~0.66–0.67), and thus the compression of the cake layer was not clearly observed for the range of operating pressures investigated in this study. Accordingly, the backwash efficiency was not varied significantly with operating pressures primarily due to similar cake structure.

3. The normalized flux was significantly reduced as the ionic strength of the feed solution increased even when particle loading to the membrane system was kept constant. This was explained by the formation of a more compact cake layer at higher salt concentrations. Modeling data clearly demonstrated a direct relationship between the density of the cake layer and ionic strength, as predicted by the colloidal interactions among the particles accumulated in the cake.
layer. Furthermore, an inverse relationship was observed between backwash efficiency and the ionic strength of the feed solution, which indicates that the efficiency of backwashing deteriorated as the packing density of the cake layer increased. In addition, particle adsorption and/or pore blockage might contribute to the reduced backwash efficiencies observed for the high ionic solutions.

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