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Effect of Fouling Conditions and Cake Layer Structure on the Ultrasonic Cleaning of Ceramic Membranes

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Abstract: Homogeneous alumina membranes fouled by polystyrene latex particles at different pH values and ionic strengths were subjected to ultrasonic cleaning. Cleaning was more effective at high and low pH than at neutral pH. At low pH values, less repulsive particle-particle interactions resulted in the removal of millimeter-scale aggregates and highly effective cleaning. At near-neutral pH, stronger repulsive particle-particle interactions caused detachment to occur as individual particles from the cake layer rather than as flocs, which was a slightly less effective cleaning mechanism. Ultrasonic cleaning of cake layers formed at high ionic strength (>0.3 M KCl) was less effective than cleaning at lower ionic strength (<0.3 M KCl). High ionic strength caused particles to coagulate in solution and deposit as flocs on the membrane surface forming a highly permeable fouling layer. This fouling layer was resistant to ultrasound at the sub-optimal cleaning conditions used in this study, perhaps due to particle attachment occurring within a primary energy minimum. Membrane cleaning experiments performed with particles of varying size showed that particle size was less important than the surface potential of the particles. For a given mass, particles that possessed the largest surface potential formed the thickest fouling layer, irrespective of particle size, and showed the greatest improvement in flux with ultrasonic cleaning. These results demonstrate that solution conditions influence ultrasonic cleaning of membranes primarily by

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modifying particle-particle and particle-membrane interactions as well as cake layer structure, rather than by impacting the extent or magnitude of cavitation events.

Keywords: Ultrasound, membrane cleaning, ultrasonic cleaning, particle fouling

INTRODUCTION

The use of ultrasound for membrane cleaning has been investigated by a number of researchers. These studies have shown that ultrasound can effectively clean fouled membranes and increase permeate flux through membranes (1–8). Most of these earlier studies focused on polymeric membranes, fouled with small organic molecules, and subjected the membranes to ultrasound via an ultrasonic bath or other ultrasonic device.

Our previous work investigated the mechanisms controlling the ultrasonic cleaning of ceramic, particle-fouled membranes (9). We determined that cavitation played a significant role in detaching particles from membranes and disrupting aggregates, while hydrodynamic turbulence accompanying ultrasound transported particles away from the membrane surface. Of particular importance were microstreamers, or millimeter-scale phenomena with bubbles that were formed by cavitation and migrated along the membrane surface towards pressure antinodes, thereby effectively scouring particles from the surface.

Aspects of the ultrasonic cleaning process that have not been addressed are the effect of solution conditions during fouling and cleaning, as well as the role of cake layer structure. We suspect that changes in both solution conditions (e.g., pH and ionic strength) and cake layer structure affect ultrasonic membrane cleaning. Changes in pH and ionic strength alter particle-particle and particle-membrane interactions and also influence the viscosity of the solution (17). Decreasing pH, for example, can render particles and membranes less negatively- or more positively-charged, and also lowers viscosity, causing an increase in the permeate flux (10). Higher ionic strength decreases the range of repulsive forces among particles and between particles and the membrane surface (10), and also increases the surface tension of the solution (11). Changes in cake layer structure alter the strength of the cake as well as the hydrodynamics at the cake-water interface, both of which may lead to changes in cleaning effectiveness (12).

In addition to changes in surface-chemical interactions and cake layer structure, solution conditions will also influence the ultrasonic process. Increased surface tension at high ionic strength results in fewer cavitation bubbles formed, but increased violence of collapse (13). It has also been shown that bubble coalescence decreases with increasing ionic strength (14), which may alter ultrasonic mechanisms leading to membrane cleaning such as cavitation and microstreamers (9). Understanding these different factors will aid in the future design of ultrasonic membrane cleaning systems. To this end, we performed ultrasonic cleaning experiments with membranes fouled by polystyrene latex particles in a dead-end filtration cell over a variety of pH and ionic strength conditions. These measurements are combined with zeta potential data and scanning electron microscope images to elucidate the mechanisms by which fouling conditions influence the ultrasonic cleaning process.

EXPERIMENTAL

Surfactant-free polystyrene latex particles with sulfate functional groups from Interfacial Dynamics Corporation (Portland, OR) were chosen to foul membranes. The particles are hydrophobic, spherical, and monodisperse. Although the particles contain sulfate functional groups, they maintain hydrophobic properties due to the polystyrene backbone. Three particle sizes were used for the experiments: 1.0 μm, 0.53 μm, and 0.30 μm. The membranes used in these experiments were hydrophilic Anodisc™ γ-alumina (γ-Al_2O_3) ceramic membranes (25 mm diameter) manufactured by Whatman (NJ). This 60 μm thick homogeneous membrane has a 0.2 μm uniform pore size throughout the membrane. Water used for all experiments was from a Milli-Q (Millipore Corportation, Bedford, MA) water system.

Fouling Experiments

A detailed description of the system used for fouling of Anodisc™ membranes is given in Lamminen et al. (9). Briefly, all membranes were fouled in a 10 mL stirred dead-end Amicon Ultra filtration cell (Bedford, MA). Ionic strength-adjusted and pH-adjusted solution was fed from a Nalgene polypropylene 4 L mixing vessel into the filtration cell. The system was pressurized using ultra high purity nitrogen gas at a constant pressure of 0.7 atm (10 psig).

After initial flux measurements, the membrane was removed, inspected visually for defects, and placed back into the filtration cell. The latex particle solution was then passed through the membrane. The solution consisted of 10 mg L^{-1} sulfate polystyrene latex particles suspended in Milli-Q water at a designated pH and ionic strength. The membrane was fouled until 550 mL of the solution had passed through the membrane. Thus, each fouled surface had the same mass of particles attached. The pH did not change over the course of the experiments.

Ultrasonic Cleaning Experiments

A detailed description of the ultrasonic cleaning system is given in Lamminen et al. (9). The water jacketed cleaning vessel, ultrasonic generator and transducer were manufactured by L3 Communications Elac Nautik (Kiel, Germany). After fouling, the membrane was removed from the Amicon cell and clamped to a stainless-steel stand which was then placed into the glass cleaning vessel that was open to the atmosphere. The cleaning vessel was then filled with 500 mL of Milli-Q water adjusted to the same pH and ionic strength conditions used for fouling. The stainless-steel stand holding the membrane was suspended 10.5 cm away from the transducer. Ultrasound was set at a frequency of 620 kHz, a power intensity of 0.21 W cm^{-2} and duration of 5 seconds for all experiments performed. Power intensity input into the solution was measured using calorimetry (15). Due to the rapid nature of ultrasonic cleaning and near complete particle removal for all treatments at optimal cleaning conditions, runs were performed under sub-optimal cleaning conditions.

Analysis of Cleaning

After the membrane had been treated with ultrasound, the flux was measured using the filtration cell with pH-adjusted and ionic strength-adjusted solution without latex particles. The flux of the cleaned membrane was then compared to the initial flux measured without latex particles. The "cleaned flux ratio" was calculated as J/J_o where J is the flux of the particle-free solution through the membrane following ultrasonic treatment and J_o is the flux of the particle-free solution through the membrane prior to fouling with particles. Because the cleaned flux ratio is not able to distinguish between an experiment with little flux decline but poor ultrasonic cleaning, and an experiment with significant flux decline and effective ultrasonic cleaning (i.e. both give cleaned flux ratios near 1.0), the results are also presented as the cleaned flux/fouled flux ratio. The cleaned flux/fouled flux ratio was calculated as J/J_F where J_F is the flux at the end of the fouling run. This ratio is equal to one if no cleaning occurred and increases with increased cleaning to a potential maximum of the initial clean water flux divided by the fouled flux (if complete flux recovery occurred). Following ultrasonic cleaning, membranes were air dried for two days and weighed. Using the mass difference before and after treatment, the amount of particles removed was estimated.

Membrane samples were prepared for scanning electron microscopy (SEM) by coating the membrane surface with gold to a thickness of several hundred angstroms. A XL-30 FEG scanning electron microscope (Philips Electron Optics, Netherlands) was used for SEM analysis at voltages of 1–5 V. The entire membrane was visible under the lowest magnification using SEM. Cross-sections of several membranes were also examined by breaking fouled membranes in half and placing the membrane in an orientation so that it could be viewed by SEM. The entire thickness and diameter of the membrane could be viewed under SEM. The thickness of the membrane was given as 60 μm and used as a guide to calculate cake layer thicknesses.

The zeta potentials of the latex particles at different pH values and ionic strengths were determined using a ZetaPlus (Brookhaven Instruments Corp, Holtsville, NY). An electro-kinetic analyzer (EKA) (Brookhaven Instruments Corp, Holtsville, NY) combined with a clamping cell was used to determine the zeta potential of the membrane at different pH values and ionic strengths, following a previously developed method (16).

RESULTS

pH Effect on Cleaning

Figure 1 shows the ultrasonic cleaning of membranes fouled using 0.53 μm particles and 1 mM KCl at different pH values. The ultrasonic cleaning of the latex particles was most effective at either high or low pH values. For example, at pH 3.5 J/J_0 was 0.94 indicating that the water flux after cleaning was nearly the same as an unfouled surface. J/J_F at pH 3.5 was greater than 5.0 indicating that the water flux after cleaning increased more than 5 times compared to the fouled flux before cleaning. At neutral pH, ultrasonic cleaning was not as effective with $J/J_0 = 0.5$. At high pH both J/J_0 and J/J_F increased indicating that the improvement in membrane performance was due to cleaning and not less fouling or changes in specific resistance of the membrane.

The effectiveness of ultrasonic cleaning at different pH values was visualized with SEM images of the surfaces after cleaning as shown in Fig. 2.

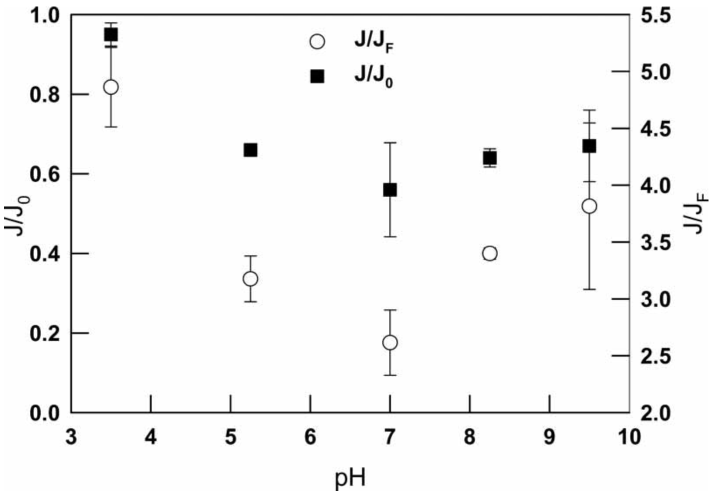

Figure 1. Cleaned flux ratio (J/J_0) and cleaned flux/fouled flux (J/J_F) of ultrasonically cleaned membranes (620 kHz, 0.21 W cm^{-2}, 5 sec treatment) at different pH values.

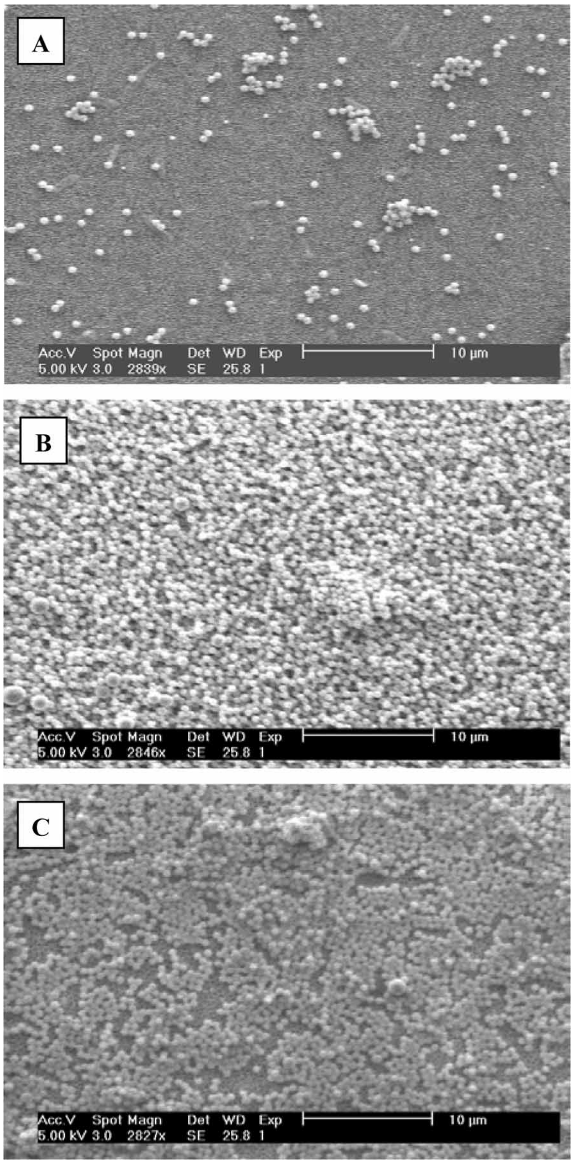

Figure 2. SEM images of ultrasonically cleaned membranes at different pHs; (A) pH = 3.5; (B) pH = 7.0; (C) pH = 9.5.

The lowest pH (panel A) resulted in a nearly particle-free surface; in contrast, neutral pH (panel B) had no exposed membrane surface after cleaning. At the highest pH (panel C) there were several areas of exposed membrane, but the surface was covered with more particles after cleaning than at the lowest pH. These images are consistent with cleaning ratio trends observed in Fig. 1.

To elucidate the role of pH in affecting the ultrasonic cleaning process, the zeta potential values of the particles and membranes were determined. The zeta potentials of the membrane and particles play a significant role in controlling membrane fouling (17) and cake layer structure, and therefore, affect the ultrasonic cleaning process. Zeta potentials of the membrane and latex particles measured as a function of pH at two ionic strengths are shown in Fig. 3. The lines in Fig. 3 represent fits to the data obtained by performing a least squares analysis, and are shown to illustrate trends. The membrane (open and filled circles) is positively charged below pH = 8.1 and negatively charged at higher pH values for both ionic strengths examined. These data are consistent with the range of values for the point of zero charge of γ-alumina reported previously (18). The latex particles had negative zeta potentials for all pH values measured, decreasing in magnitude with decreasing pH. Increasing ionic strength decreased the magnitude of the zeta potentials for both the membrane and particles.

Zeta potential data provide mechanistic insight into the role of solution conditions in ultrasonic cleaning. At the lowest pH investigated (3.5), the particles had the smallest magnitude zeta potential (least negative) while the membrane was highly positively charged. Therefore, particle-particle repulsive forces were lower than at higher pH values. The lower particle-particle repulsive forces also likely resulted in a more compact cake structure. As a result, greater force was required to remove individual particles from the cake layer compared to higher pH values. Visual observations made during ultrasonic cleaning at pH 3.5 showed large, millimeter-scale particle aggregates detaching from the membrane surface. Once in

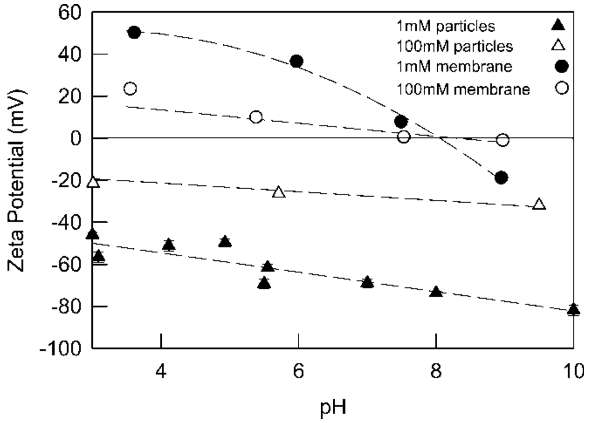

Figure 3. Zeta potentials of membrane and sulfate latex particles at 1 mM KCl and 100 mM KCl.

solution, the flocs slowly disaggregated due to the ultrasonically generated turbulence. The stronger particle-particle interactions and more dense cake layer structure at this low pH apparently facilitated patch removal of aggregates from the surface. This removal mechanism resulted in very effective membrane cleaning with a cleaned flux ratio of 0.94 and a cleaned flux which was more than 5 times greater than the fouled flux.

Ultrasonic cleaning was poorest at intermediate pH values. At pH values up to 8.1, the particles were increasingly negatively charged, while the membrane was less positively charged than at lower pH. As a result of increased particle charge, more repulsive interparticle forces resulted in weaker particle-particle binding, and a less dense cake layer, than at lower pH values. At these intermediate pH values, visual observations showed that individual particles or very small aggregates were released during the cleaning process rather than large flocs. At pH 7.0, the cleaned flux ratio was only 0.60, and the cleaned flux was only about 3 times greater than the flux observed for the fouled membrane. Thus, the more repulsive particle-particle interactions at intermediate pH facilitated the removal of individual particles by ultrasound, rather than large aggregates. Subsequently, the overall effectiveness of ultrasonic cleaning was lower at the short cleaning times used in this study. Altmann et al. (19) and Rubin (20) demonstrated that the lift force on a particle attached to a surface increases with particle size. Thus, the less effective removal of individual particles, compared to aggregates, is perhaps due to lower lift forces generated by the ultrasonic velocity field for smaller sized aggregates or particles compared to the lift forces generated for larger aggregates. We have previously shown that cake layers composed of smaller particles are more difficult to clean by ultrasound compared to cakes formed by larger particles (21).

At the highest pH (9.5), the particles had a very strong negative charge and the membrane was also negatively charged. In this case, particle-particle and particle-membrane interactions were very repulsive. Subsequently, particles were easily detached from the cake layer in smaller clusters and the membrane was cleaned effectively. With a cleaned flux ratio of 0.72 and a cleaned flux 3.6 times greater than the fouled flux, the cleaning was not as complete as with the lowest pH. At the short cleaning times used in this study, large aggregate removal was more effective at cleaning than removal of individual particles. Increasing the time of cleaning from 5 seconds to 5 minutes completely restored the initial flux for all of the pH values tested.

Ionic Strength Effect on Ultrasonic Cleaning

The effect of ionic strength, at constant pH (7.0), on ultrasonic cleaning was examined from 1 mM to 1 M potassium chloride. At all ionic strengths, membranes were fouled with 5.5 mg of latex particles. We expected that increasing the ionic strength would result in less repulsive particle-particle and particle-membrane interactions, resulting in less effective cleaning by ultrasound. Figure 4 shows that there was indeed a decrease in J/J_0 with increasing ionic strength, but this was followed by a sharp increase at ionic strengths greater than 0.1 M. The ratio J/J_F generally decreased with increasing ionic strength over the range of ionic strengths tested (see Fig. 5). At ionic strengths greater than 0.2 M, the cleaned flux/fouled flux ratio (J/J_F) was close to 1.0 indicating no improvement in flux upon cleaning. Thus, at high ionic strength the increase in cleaned flux ratio was not a result of increased ultrasonic cleaning. Instead, little flux decline was observed as a result of membrane fouling at the higher salt concentrations, and therefore, both J/J_0 and J/J_F remained close to 1.0. The improved cleaned flux ratio at low ionic strength, however, can be attributed to more effective cleaning as the ratio J/J_F was greater than 1.

Figure 5 demonstrates that little particle release from the membrane surface occurred at high ionic strength, as determined from the fraction of particles remaining on the membrane surface. Figure 6 shows the SEM images of membranes fouled with latex particles at 1 mM KCl (panel A), 0.1 M KCl (panel B), and 1 M KCl (panel C). The 1 mM and 100 mM ionic strength images show a well packed, ordered surface while the high ionic strength (1 M KCl) image shows dark areas which suggest a more porous fouling layer. In fact, the cake layer at higher ionic strength (1 M KCl) was approximately twice as thick and less dense (Fig. 6, panel E) than at the lower ionic strength of 1 mM KCl (Figure 6, panel D), as determined by SEM cross sections. Direct visualization of cake layer cross-sections by Wiesner and colleagues has demonstrated that the outer portions of cake

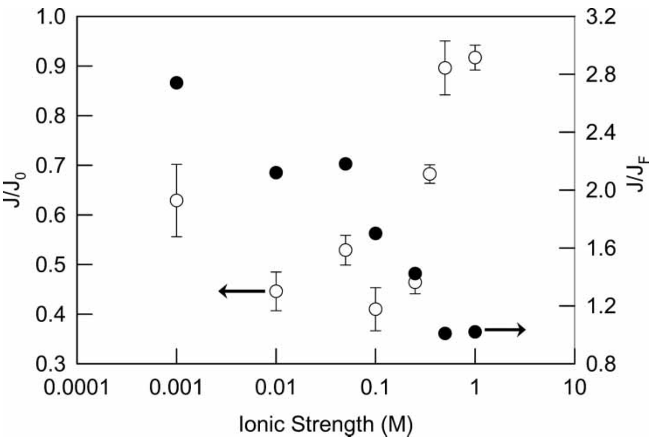

Figure 4. Cleaned flux ratio (J/J_0) and cleaned flux/fouled flux (J/J_F) of ultrasonically cleaned (620 kHz, 0.21 W cm^{-2}, 5 sec treatment) membranes at different ionic strengths.

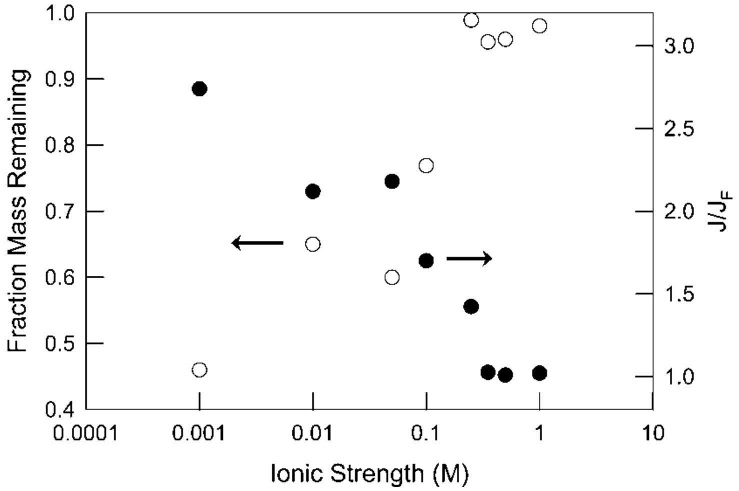

Figure 5. Ratio of ultrasonically cleaned flux/fouled flux (J/J_F) and the fraction of mass remaining on the surface following ultrasonic cleaning at different ionic strengths.

layers are more porous when formed at higher ionic strength due to higher collision efficiencies (22). The higher ionic strengths in our experiments were also above the salt concentration in which the particles are destabilized (0.3 M as stated by manufacturer). Therefore, at least some particles agglomerated in solution above the membrane with subsequent floc deposition onto the surface. Hwang et al. also found that coagulation of particles due to compression of the electric double layer resulted in high cake porosity and low specific filtration resistance of the cake layer (23).

These findings indicate that the initial decrease in cleaning with increasing ionic strength was a result of less repulsion among the particles, and therefore, stronger particle-particle interactions. Elzo et al. also observed that at higher ionic strength, more hydrodynamic turbulence was required to remove particles from surfaces than at low ionic strengths (23). This was attributed to compression of the double layer and less particle-particle repulsion at the higher ionic strengths. However, unlike at low ionic strength and low pH, flake removal was not observed at higher ionic strengths. This result can be explained by considering the relative importance of primary- versus secondary-minimum particle-particle interactions at different ionic strengths. At high ionic strength, the particles aggregate and deposit onto the membrane surface in a primary energy minimum. As a result, the fouling layer was very difficult to remove and resisted ultrasonic cleaning. At low pH and low ionic strength, on the other hand, a significant energy barrier remained between the particles, and therefore, particles were aggregated within the cake layer in a secondary energy minimum.

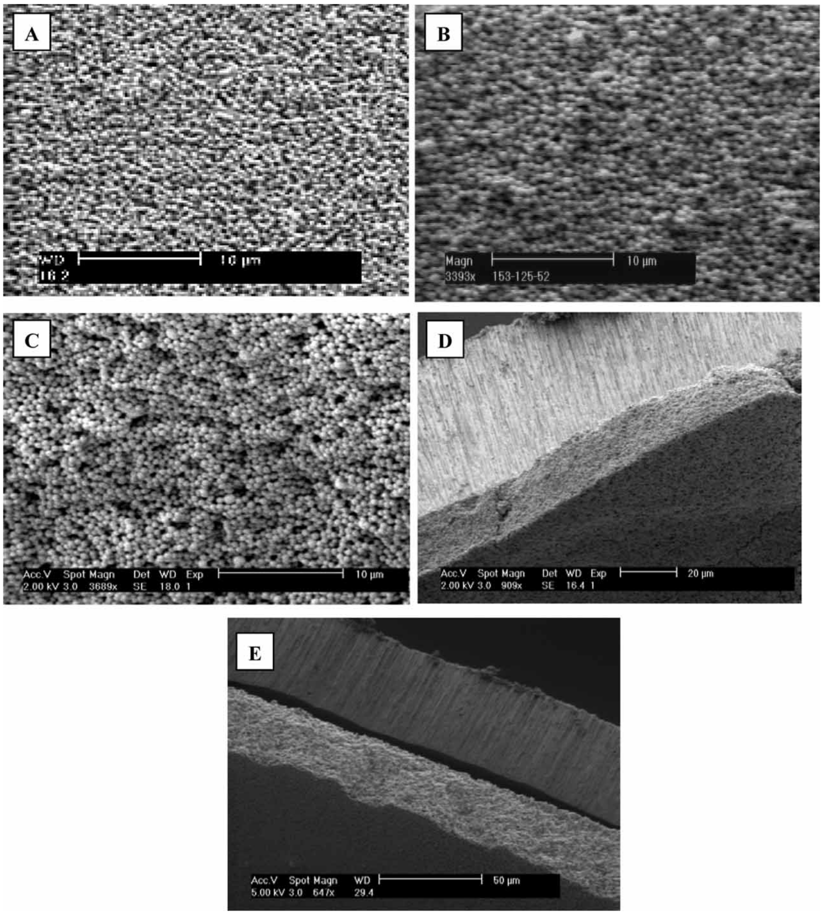

Figure 6. SEM images of fouling layers on membranes fouled with particles; (A) 1 mM KCl fouling solution with 10 mg L^{-1} 0.53 mm latex particles; (B) 0.1 M KCl fouling solution with 10 mg L^{-1} 0.53 mm latex particles; (C) 1 M KCl fouling solution with 10 mg L^{-1} 0.53 mm latex particles; (D) SEM cross section of the fouling layer at 1 mM KCl; (E) SEM cross section of the fouling layer at 1 M KCl.

Thus, less force was needed to dislodge sections of the cake layer from the membrane surface.

The reduction in ultrasonic cleaning with increasing ionic strength may also be influenced by changes in cavitational activity. With increasing ionic strength there is an increase in the surface tension of solution. In our experiments an increase of 1.8 dynes/cm from pure water to 1 M KCl is expected (24). This increase in surface tension results in more energy needed to form cavitation bubbles. However, the resulting bubbles produce more erosive turbulence (25). It has been demonstrated that these cavitation bubbles are largely responsible for the ultrasonic cleaning of the membranes (9). Cavitation bubbles that are formed under higher ionic strength collapse more violently, and are expected to clean more effectively than bubbles formed at lower ionic strengths. It has also been shown that cavitation bubbles do not coalesce as readily as ionic strength increases (26). This decrease in coalescence reduces one of the significant contributors to ultrasonic cleaning, microstreamers. Microstreamers are bubbles that travel in solution and coalesce with other bubbles. They have the ability to scour the membrane surface and detach fouling materials. This mechanism was shown to contribute greatly to the cleaning of membranes (9).

In order to determine if cleaning was dependent on changes in cavitational activity, experiments were performed with membranes fouled with high ionic strength solution (1 M KCl), but ultrasonically cleaned in Milli-Q water. To ensure that there were no structural changes in the cake layer as a result of the Milli-Q water, membranes which had been fouled with particles at 1 M KCl, were removed from the fouling cell, soaked in Milli-Q water for 10 minutes and placed back into the cell to test fluxes with particle-free 1 M KCl solution. There was no change in the flux of the solution suggesting little alteration of the cake layer structure due to changing the solution to Milli-Q water. The fouled membrane which was cleaned in Milli-Q water was found to be resistant to ultrasonic cleaning. Cleaning in both high ionic strength and Milli-Q water solutions resulted in $J/J_F \approx 1.0$, indicating no significant cleaning. This result suggests that the reduction in cleaning was not a result of changes in cavitation with increasing ionic strength. Instead, ionic strength influenced ultrasonic cleaning of membranes primarily by modifying particle-particle and particle-membrane interactions as well as cake layer structure.

Particle Size Effect on the Ultrasonic Cleaning of Membranes

The effect of particle size on ultrasonic cleaning was also examined. Particles of different size create cake layers with different compositional structures (10), which in turn, may influence ultrasonic cleaning due to the different packing densities, permeabilities and particle-particle interactions. Figure 7 shows the initial clean solution flux (first 10 minutes) followed by the flux during fouling at 1 mM KCl, pH = 7.0 using different sized particles at concentrations of 10 mg L^{-1}. From Fig. 7, the normalized flux was highest for the largest particle size (1.0 μm) compared to the permeate flux through cake layers formed from smaller particles (0.3 and 0.53 μm). Fouling by the smallest particles resulted in the greatest flux decline and took the longest time to filter 550 mL of solution. With respect to ultrasonic cleaning, the largest recovery in flux was seen with medium sized particles (0.53 μm). Both the largest and smallest particles experienced less improvement in flux as a result of ultrasonic treatment.

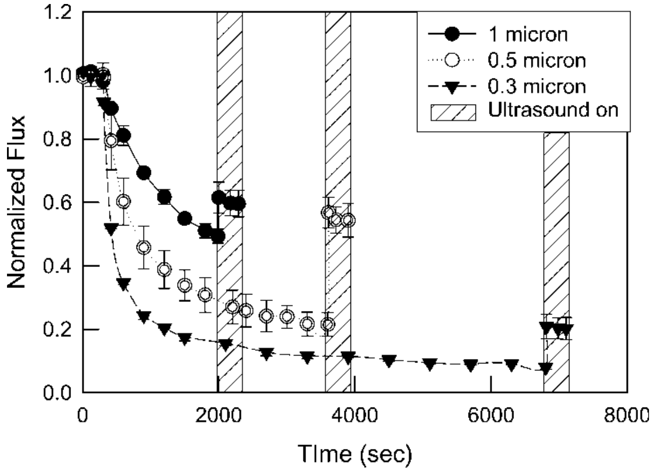

Figure 7. Normalized permeate flux of the membranes during initial particle free flux test followed by fouling with particles of sizes, 1.0 μm, 0.53 μm, and 0.3 μm, followed by ultrasonic cleaning at 620 kHz, 0.21 W cm^{-2}, and a duration of 5 seconds and then a particle free permeate flux test.

Figure 8 shows cake layer cross-sections for different sized particles prior to ultrasonic cleaning. The cake layer is the block of material on the left hand side of each image. Using the membrane as a reference thickness of 60 μm, the thickness of each of the fouling layers was calculated from several different points along the fracture interface. The thickness was largest for the 0.53 μm particles (45 μm), followed by the 1.0 μm particles (38 μm) and then the smallest particles, 0.30 μm, (29 μm). All of the membranes had approximately the same mass of particles on the surface, which was verified gravimetrically. Thus, intermediate size particles (0.53 μm) had the most porous cake layer structure, with larger (1.0 μm) and smaller (0.3 μm) particles having more dense cake layers. Although the medium-sized particles (0.53 μm) in our study were not expected to have the thickest fouling layer, it may be due to these particles having the most negative zeta potential (−68 mV at pH = 5.5). The largest particles (1.0 μm) and the smallest particles (0.30 μm) both possessed zeta potentials that were similar (−36 mV for pH = 5.6 and −41 mV for pH = 5.5 respectively). Higher zeta potentials create a thicker, more porous fouling layer due to the increased particle-particle repulsion and larger inter-particle distances (10, 27). Also, with highly charged particles, ultrasonic cleaning was faster because the particles readily detached from the cake layer. The cake layers that were formed by these particles were similar in thickness and showed little improvement with ultrasonic cleaning. This suggests that the zeta potentials of the particles were more important in determining the structure of the cake layer and the effectiveness of membrane cleaning than particle size.

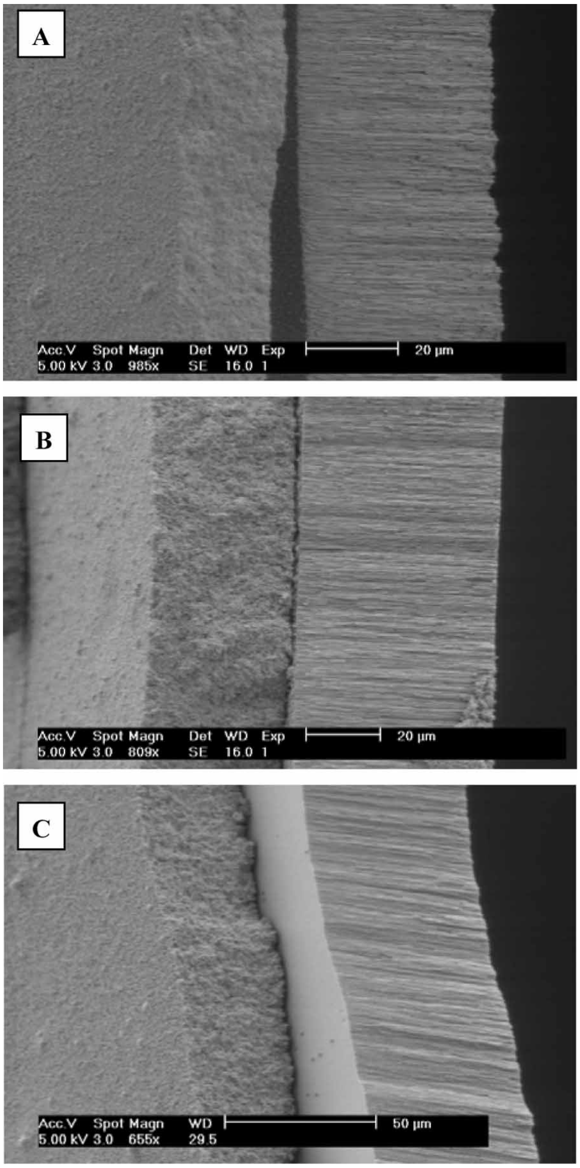

Figure 8. SEM cross sections of the fouling layers (no cleaning) created by the different sized particles. (A) 0.3 μm, (B) 0.53 μm and (C) 1.0 μm.

CONCLUSIONS

Ionic strength, pH, and particle interactions have significant impacts on the ultrasonic cleaning of membranes. Ultrasonic cleaning was greatly affected by the pH of the solution, which is attributed to changes in particle-particle and particle-membrane interactions, and subsequent changes to the cake layer structure. Cleaning was most effective at low and high pH. Ultrasonic cleaning at higher ionic strength was not as effective as cleaning fouling layers formed at lower ionic strengths. High ionic strength caused particles to coagulate in solution and deposit as flocs on the membrane surface. These particles may have been attached to each other within a primary energy minimum and thus were resistant to ultrasonic cleaning as a result of their strong interactions. Particle size experiments revealed that the zeta potentials of the particles were a significant factor for increased ultrasonic cleaning. The repulsive particle-particle interactions of the highly charged particles made for better removal than with lesser charged particles regardless of size.

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REFERENCES

1. Kobayashi, T., Kobayashi, T., Hosaka, Y., and Fujii, N. (2003) Ultrasound-enhanced membrane cleaning processes applied water treatments: influence of sonic frequency on filtration treatments. *Ultrasonics*, 41: 185.
2. Kobayashi, T., Chai, X., and Fujii, N. (1999) Ultrasound enhanced cross-flow membrane filtration. *Separation and Purification Technology*, 17: 31.
3. Chai, X., Kobayashi, T., and Fujii, N. (1998) Ultrasound effect on cross-flow filtration of polyacrylonitrile ultrafiltration membranes. *Journal of Membrane Science*, 148: 129.
4. Chai, X., Kobayashi, T., and Fujii, N. (1999) Ultrasound-associated cleaning of polymeric membranes for water treatment. *Separation and Purification Technology*, 15: 139.
5. Kokugan, T., Kaseno, Fujiwara, S., and Shimizu, M. (1995) Ultrasonic effect on ultrafiltration properties of ceramic membrane. *Membrane*, 20: 213.
6. Zhu, C. and Liu, G. (2000) Modeling of ultrasonic enhancement on membrane distillation. *Journal of Membrane Science*, 176: 31.
7. Li, J., Sanderson, R.D., and Jacobs, E.P. (2002) Ultrasonic cleaning of nylon microfiltration membranes fouled by Kraft paper mill effluent. *Journal of Membrane Science*, 205: 247.
8. Muthukumaran, S., Yang, K., Seuren, A., Kentish, S., Ashokkumar, M., Stevens, G.W., and Grieser, F. (2004) The use of ultrasonic cleaning for ultrafiltration membranes in the dairy industry. *Separation and Purification Technology*, 39: 99.
9. Lamminen, M.O., Walker, H.W., and Weavers, L.K. (2004) Mechanisms and factors influencing the ultrasonic cleaning of particle-fouled ceramic membranes. *Journal of Membrane Science*, 237: 213.

10. Fabish, R.S., Elimelech, M., and Cohen, Y. (1998) Effect of interparticle electrostatic double layer interactions on permeate flux decline in crossflow membrane filtration of colloidal suspension: an experimental investigation. *Journal of Colloid and Interface Science*, 204: 77.
11. Yu, Y.-X., Gao, G.-H., and Li, Y.-G. (2000) Surface tension for aqueous electrolyte solutions by the modified mean spherical approximation. *Fluid Phas Equilibria*, 173: 23.
12. Elimelech, M. and O'Melia, C.R. (1990) Kinetics of deposition of colloidal particles in porous media. *Environmental Science and Technology*, 24: 1528.
13. Pee, G.Y., Rathman, J.F., and Weavers, L.K. (2004) Effects of surface active properties on the cavitational degradation of surfactant contaminants. *Industrial Engineering Chemical Research*, 43: 5049.
14. Weissenborn, P.K. and Pugh, R.J. (1996) Surface tension of aqueous solutions of electrolytes: relationship with ion hydration, oxygen solubility, and bubble coalescence. *Journal of Colloid and Interface Science*, 185: 550.
15. Raso, J., Manas, P., Pagan, R., and Sala, F. (1999) Influence of different factors on the output power transferred into medium by ultrasound. *Ultrasonics Sonochemistry*, 5: 157.
16. Walker, S.L., Bhattacharjee, S., Hoek, E.M.V., and Elimelech, M. (2002) A novel asymmetric clamping cell for measuring streaming potential of flat surfaces. *Langmuir*, 18: 2193.
17. Childress, A.E. and Elimelech, M. (2000) Relating nanofiltration membrane performance to membrane charge (electrokinetic) characteristics. *Environmental Science and Technology*, 34: 3710.
18. Kasprzyk-Hodern, B. (2004) Chemistry of alumina, reactions in aqueous solution and its application in water treatment. *Advances in Colloid and Interface Science*, 110: 19.
19. Altmann, J. and Ripperger, S. (1997) Particle deposition and layer formation at the crossflow microfiltration. *Journal of Membrane Science*, 124: 119–128.
20. Rubin, G. (1977) Widerstands- und Auftriebsbeiwerte von ruhenden kugelfoermigen Partikeln in stationaeren, wandnahen laminaren Grenzschichten, Dissertation. TH Karlsruhe.
21. Chen, D., Weavers, L.K., and Walker, H.W. (2006) Ultrasonic control of ceramic membrane fouling: effect of particle characteristics. *Water Research*, 40: 840.
22. Volodymyr, T.V., Koyuncu, I., and Wiesner, M.R. (2004) Effect of hydrodynamics and solution ionic strength on permeate flux in cross-flow filtration: direct experimental observation of filter cake cross-sections. *Journal of Membrane Science*, 241: 65.
23. Hwang, K.J. and Liu, H.C. (2002) Cross-flow microfiltration of aggregated submicron particles. *Journal of Membrane Science*, 201: 137.
24. Weissenborn, P.K. and Pugh, R.J. (1996) Surface tension of aqueous solutions of electrolytes: relationship with ion hydration, oxygen solubility, and bubble coalescence. *Journal of Colloid and Interface Science*, 185: 550.
25. Iwai, Y. and Li, S. (2003) Cavitation erosion in waters having different surface tensions. *Wear*, 254: 1.
26. Wall, M., Ashokkumar, M., Tronson, R., and Grieser, F. (1999) Multibubble sonoluminescence in aqueous salt solutions. *Ultrasonics Sonochemistry*, 6: 7.
27. Huisman, I.H., Tragardh, G., and Tragardh, C. (1999) Particle transport in crossflow microfiltration – II. Effects of particle-particle interactions. *Chemical Engineering Science*, 54: 281.