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DEPARTMENT OF CHEMICAL ENGINEERING

REDUCTION OF MEMBRANE FOULING USING CALCIUM CARBONATE
DIFFUSIOPHORETIC MICROPUMPS

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ABSTRACT

Membrane systems are becoming an increasingly important method for filtration, but their utility is often compromised by colloidal fouling. While there are a number of studied causes of fouling, one probable source of membrane fouling that has not been studied is fouling due to diffusiophoresis. Diffusiophoresis is a particle transport phenomenon that occurs from the combination of electrophoretic and chemiphoretic forces due to the presence of a transient salt gradient. In this thesis, we establish that diffusiophoresis is a mechanism that affects the colloidal fouling of microdialysis membranes. By recognizing this mechanism, we also see that diffusiophoresis can mitigate or reverse this fouling through the use of calcium carbonate micropumps. The first part of this hypothesis was explored by modelling the motion of particles near the membrane due to diffusiophoresis and comparing these values to the experimental velocities in transient salt gradients within membrane modules. We found that the motion of the particles closely matched the predicted values under diffusiophoretic motion, demonstrating its presence. The second component was supported by placing calcium carbonate micropumps in the setups that had previously shown extensive fouling due to the salt gradient and contrasting the systems with and without the micropumps. The system with the calcium carbonate micropumps showed no accumulation of particles on the membrane wall and an exclusion region, with no particles around the membrane. By recognizing the importance of diffusiophoresis in membrane fouling, and then using this mechanism to mitigate the problem, we aim to improve membrane performance.
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Chapter 1
Introduction

Membranes are used for filtration in various industries, such as water purification\(^1\) and biopharmaceuticals\(^2\), but these membranes are prone to fouling, which causes a reduction in the flux across the membrane. One of the primary causes of membrane fouling is colloidal particle deposition\(^3\). Fouling is a direct result of filtration. Particles within the liquid are carried by the flow of the liquid to the membrane wall. A fraction of these particles are carried into the pores of the membrane, where they may become lodged, decreasing the flow through that pore. This, combined with the constant accumulation of particles on the membrane wall, results in a decrease in flux over time. This decrease in flux is an important problem because, over time, the flux may fall below a critical threshold below which the filtration process is no longer cost effective\(^4\). Two major, established causes of this flux are the electrostatic\(^5\) and hydrophobic\(^6\) interactions between the particles and membrane. Here, we will determine the effect of a third process, diffusiophoresis, on the flux by analyzing its effect on the motion of particles around the membrane.

This thesis seeks to test the hypothesis that diffusiophoresis as an active mechanism for the migration of particles within a membrane system that can be manipulated to affect the fouling rate of membranes. This will be accomplished by sequentially exploring two questions:

1. Do the transient salt gradients in the membrane systems of interest induce diffusiophoresis, and is this diffusiophoresis an explanation for the motion of tracer particles in these transient salt gradients?
2. Can calcium carbonate micropumps be used to create diffusiophoretic motion away from the membrane and thereby prevent fouling?
The presence of diffusiophoresis due to transient salt gradients and of its effect on the motion of tracer particles is supported by Figure 1, which shows the aggregation of particles on the outside walls of the hollow fiber membrane. The effect of calcium carbonate micropumps on the system is shown by Figure 2. It shows the predicted and experimental results of the same experiment as in Figure 1 but in the presence of the calcium carbonate micropumps. The micropumps prevent the aggregation and consequent fouling of the membrane and even create an exclusion region around the membrane where no particles are present.

Figure 1. Particles aggregate on the walls of the membrane due to the diffusiophoretic effect moving the particles towards the membrane. This shows the ability of diffusiophoresis to affect particle motion and simulates the effect of membrane fouling during use.
Figure 2. Calcium carbonate micropumps are used to prevent the aggregation of particles on the membrane wall and consequent fouling of the membrane. In comparison to Figure 1, there is not only a lack of particle aggregation, but also an exclusion region around the membrane.
Chapter 2

Background

The hypothesis will be evaluated by three sets of experiments. The first part will be tested by quantifying the rate of diffusion of the chloride ions within the membrane system, both theoretically and experimentally, and measuring the zeta potentials of the tracer particles. Quantifying the electric field and zeta potential allows for the numerical prediction of the effect of diffusiophoresis on particle motion. Matching the predicted values to the experimental particle velocities in a transient salt gradient will show that diffusiophoresis is a valid explanation for the motion of the particles, including the accumulation on the membrane wall and, thus, the rate of fouling. The second component is supported by numerically and experimentally showing that the presence of calcium carbonate micropumps in the system can be used to prevent fouling and create a colloid-free exclusion region in membrane system setups that had previously undergone fouling due to the transient salt gradient.

The particles of interest in this study are colloidal particles. Colloidal particles are particles of a size between 1 nanometer and 1 micrometer that are suspended in a solid or fluid, in this case water. The defining characteristic of colloids is that the surface area between the particle and fluid is very high in relation to the mass of the dispersed particles.

Diffusiophoresis is a phenomenon that occurs when a colloidal particle with a surface charge interacts with a chemical gradient. This occurs in both ionic and nonionic gradients. In this experiment, salt, or ionic, gradients were studied. The diffusiophoretic effect is a result of two electrokinetic phenomena: electrophoresis and chemiphoresis. Electrophoresis is the movement of a particle with a charged electric double layer within an electric field. The electric field is generated by the different rates of diffusion for the two ions of the salt. An electric double layer is “the interface between an electrolyte solution and a solid surface”. This layer is comprised of two levels of charge. The first occurs on the surface of the solid. Most solids will develop a surface charge when introduced to a polar fluid, in this case water. This charge causes ions of the opposite charge within the solution to be
attracted to the surface. For example, in a solution of NaCl, a particle with a positive surface charge will attract the negative chlorine ions to its surface. It will have an inner positively charged layer and an outer, negatively charged layer; hence a double layer as shown by the particle in Figure 3. The external charge of the double layer induces motion when in the presence of an electric field. In this case, the electric field is created by the different rate of diffusion for the anions and cations. As shown in Figure 3, the anions diffuse faster, creating an electric field away from the high electrolyte concentration area. In this situation, the particle will therefore move to the left due to electrophoresis. Chemiphoresis is the movement of a particle to the lower end of a concentration gradient. In Figure 3, this is the same direction of motion induced by the electrophoretic mechanism. Since the salt gradient contains both an electric field and a concentration gradient, a combination of the two processes occurs, under the combined effect termed diffusiophoresis.

![Figure 3](image_url)

**Figure 3.** The salt concentration gradient combined with the different diffusion rates of the salt ions create an electric field through which the particle moves due to the presence of its charged double layer.

Particle speeds are the primary variable of interest in these experiments. The velocity of a particle due to electrophoresis is a function of the magnitude of the electric field, the zeta potential of the
particle, the viscosity of the liquid, and the size of the particle. The zeta potential can be defined as “the potential that can be measured at the surface of shear that forms if the solid was to be moved relative to the surrounding ionic medium.” The zeta potential value is related to the strength of the charge on the particle surface. Thus, larger zeta potential values will yield higher absolute velocities in an electric field. The sign of the zeta potential will determine the direction of the velocity of the particle within this field, assuming no other forces are acting on it. With everything but the electric field strength being temporally constant, the velocity effectively varies with the change in electric field over time. Thus, the change in particle speeds over time can be used to demonstrate the effect of diffusiophoresis and its decay over time.

The two types of microscope used in this experiment are an inverted light microscope and a confocal microscope. An inverted light microscope is similar to a standard light microscope except that the light source is above the stage and the objective is below; both parts still point to the stage but from opposite directions to a normal microscope. The laser scanning confocal microscope is a completely different microscope. It is a type of scanning optical microscope, meaning that it does not light the entire field of view but instead scans the field with a concentrated beam. Because the light source is a scanning laser, the wavelength can be carefully controlled, allowing for fluorescence microscopy. Additionally, in a confocal microscope, only the rays that are in focus are allowed into the confocal aperture and into the photomultiplier. This gives confocal microscopy the advantage of a larger depth of field at the cost of lateral resolution.
The membranes of interest in these experiments use pores in the membrane wall to create a semi-permeable barrier between two sides of the membrane\textsuperscript{18}. This barrier allows particles below a certain size threshold, as well as most liquids, to pass through the pores and onto the other side of the membrane\textsuperscript{18}. All particles larger than the threshold are rejected by the membrane, filtering the system. Fouling of a membrane occurs when the particles above the size threshold are carried to the wall of the membrane, usually by the motion of the fluid across the membrane, where some particles enter and clog the pores, and others form a cake layer on wall\textsuperscript{19,20}. This reduces the flux of the solution across the membrane and slows down the filtration process, reducing the life of the membrane and costing time and money\textsuperscript{21-23}. A

**Figure 4.** Exploded drawing of the optics of a confocal microscope with a diagram of the path of light through the system. As shown by the dotted line, the light remains in a single beam that scans the specimen. It is Figure 2 from reference 17.
variety of methods have been tried to reduce fouling. One such method is to periodically reverse the flow to reduce buildup and fouling of the membrane\textsuperscript{24,25}. Modifications to the membrane surface and water affinity of the membrane, as well as electric fields have all been experimented with to reduce fouling\textsuperscript{26-29}. However, using diffusiophoresis, in this case via calcium carbonate micropumps, has yet to be explored as a means to reduce or prevent fouling.

Membrane fouling is an important concern because of the widespread use of membrane filtration and separation. For example, membrane technology is used in the food industry, such as in situations where heat treatment compromises the product or is unable to remove all hazards\textsuperscript{30}. One such example is in the processing of milk. Heat treatment kills all of the bacteria but leaves some threats, such as bacterial spores, unaffected. Membrane filtration is one of the options used to remove these spores. In this case, fouling is specifically mentioned as a concern\textsuperscript{30}. Internal fouling, particles collecting in and clogging the pores, and external fouling, rejected particles collecting on the surface, were both described as problems. Another industry that frequently uses membrane filtration is waste water disposal. For example, in the case of the petroleum industry, membrane filtration is one of the methods used to separate oil droplets from water\textsuperscript{31}. The need to remove this oil is twofold. First, there is a growing demand for clean water and for the protection of the environment, and because of the vast quantities of water used in the petroleum industry, simply dumping the contaminated water has become a far less viable option. Second, failing to filter out the oil wastes a valuable source of fuel. Thus, the oil industry has investigated various physical and chemical methods of separation, and membrane filtration has proven to be one of the most promising options\textsuperscript{31}. However, like in the food industry, the largest problem associated with membrane technology is fouling and the subsequent decrease in flux that it creates. The potential of membrane filtration and the obstacle posed by membrane fouling are issues that also concern municipal waste water treatment\textsuperscript{32,33}. Here, colloidal particles, similar to the ones used in testing this experiment, are the primary cause of membrane fouling. In addition to waste water, membrane technology is used in the production of drinking water\textsuperscript{34-36}. Membrane technology is still a new method of filtration in this field,
having been around for about 25 years. Membranes can be used in water filtration to remove all of the bacteria and viruses in drinking water, but again, fouling is a cause for concern. Fouling decreases the flux of the membrane, increases the frequency of backwashing needed, and shortens the life of the membranes\textsuperscript{34}. As shown in Figure 5, colloids can simulate viruses or bacteria, depending on the size. In our case, the colloids used are better simulations of bacteria than viruses because they are 3 micrometers in diameter. The colloids and membranes used are thus an applicable size for comparing with one of the size thresholds used in drinking water purification. Membrane filtration is used in a wider range of applications than one might expect. One example of this is in the electronics industry, in which silicon is widely used as an important semiconductor\textsuperscript{37}. The silicon used must be very pure and membrane filtration is one of the best options available for purifying it. It requires simple equipment and relatively little energy, meaning that it is a very cost effective option\textsuperscript{25}.

![Figure 5](image)

**Figure 5.** Size chart showing the types of particle filtration and the particles found in water. This shows the actual and relative sizes as well as the types of filtration used to remove different types of particle.

One of the most common uses of hollow fiber membranes in particular is for kidney dialysis. Here, membrane filtration is used to help carry out the functions of damaged kidneys\textsuperscript{38}. In patients with
renal failure, hollow fiber membrane filtration is often used to remove the toxins, which tend to have smaller molecular weights, but retain the needed proteins, which are larger. Additionally the blood cannot sustain too much mechanical disturbance, such as the high pressures found in many membrane filtration systems. Large pressure or significant shearing forces can damage the red blood cells\textsuperscript{38}. Hollow fiber membranes are especially effective because they do not require significant added pressure to operate.
Chapter 3

Materials and Methods

The experiments were run with a hollow fiber membrane and the transport through the membrane was followed using polystyrene latex tracer particles. For the experimental setup of the membranes, the following materials were used: membrane, salt, water, calcium carbonate micropumps, dyes, and colloidal particles. The membrane used was a hollow fiber membrane. Membranes are classified by the maximum size of particles that they allow to pass through. The membrane used in this experiment had a threshold of 13 kDa. One Da, or Dalton, is equal to one atomic mass unit. The membranes had an outside diameter of 280 micrometers and a wall thickness of 40 micrometers. They were procured from Spectrum Laboratories (Rancho Dominguez, CA). Three salts, lithium chloride (LiCl), potassium chloride (KCl), and sodium chloride (NaCl), were used in the experiments. They were all obtained from Sigma-Aldrich. The deionized water came from a Millipore Corporation Milli-Q system. The system produced water with a specific resistance of 1 MΩ cm. The specific resistivity is a measure of the concentration of ions in water. The ions in water allow it conduct electricity. Thus, a high resistance, such as in the water from our system, indicates a low concentration of ions in the water. Much of the conductivity, and thus ion concentration, in the deionized water is due to the carbon dioxide in the air, which reacts with water and makes reaching complete deionization difficult. The calcium carbonate (CaCO$_3$) for the micropumps was made by combining calcium chloride (CaCl$_2$) and sodium carbonate (Na$_2$CO$_3$). These salts were also obtained from Sigma-Aldrich. The dye used to visualize the diffusion rate of chloride ions was Lucigenin, obtained from Invitrogen Molecular Probes (Eugene, OR). The colloidal particles used were sulfated polystyrene latex. The standard particles had diameter 3.0 μm ± 2.1% and a concentration of 8% and the red fluorescent particles had diameter 4.0 μm ± 2.0% and a concentration of 2%, with an excitation wavelength of 580 nm and an emission wavelength of 605 nm. They were obtained from
Interfacial Dynamics Corporation (Portland, OR) and were used to observe the motion due to the salt gradient. The capillaries used were 0.9 millimeter borosilicate glass square capillaries from Vitrocom (Mountain Lakes, NJ). For filling and washing the membranes, 21G precision needles and 1 mL syringes, purchased from BD Sciences, were used. The experiments were set up on 25 by 75 millimeter VWR glass microslides. Paraffin wax was used as a sealant in the experiments.

The observation tools used were an inverted light microscope and a confocal microscope. The inverted light microscope was a Nikon Eclipse TE2000-U, shown in Figure 6. It has a CCD camera and an optical light source. This was used for most of the experiments. The confocal microscope used was a Leica TCS SP5 laser scanning confocal microscope obtained from LSCM, Leica Microsystems. The confocal microscope was used for the experiments mapping chloride diffusion (with fluorescent dye) and the experiments using fluorescent particles. The light microscope experiments were analyzed with Nikon NIS Elements Imaging Software V. 4 and the confocal experiments with Image J software from the National Institutes of Health.

Figure 6. The light microscope used in most of the experiments.
In order to predict the particle motion, the electric field strength and the zeta potential of the particles must be known. The zeta potential was measured with a Zetasizer Nano ZS90, model number ZEN3690, obtained from Malvern, MA and shown in Figure 7 above. This machine measures the zeta potential of a particle through the use of electrophoretic light scattering\textsuperscript{41}. The system uses laser light scattering spectroscopy to measure the particle motion in an oscillating electric field\textsuperscript{42}. The zeta potentials were measured at room temperature (298K) and in salt concentrations varying from 0.1 mM to 100 mM. The pH used in the tests was 5.8.

Most experiments were carried out with the same general setup. The salt and deionized water were put into separate small, sealable vials. The capillary was filled with deionized water by holding one end in the vial. Capillary action then slowly filled the capillary with liquid. With one end of the capillary still in the vial, a small amount of melted wax was applied to the other end, sealing it. Because the
capillary is very narrow, sealing one end effectively keeps all of the water inside. After the wax solidified, the capillary was removed from the vial and placed on a glass slide. The sealed end was then fixed to the slide with another droplet of wax. Next, the hollow fiber membrane was cut with a razor to the approximate length of the capillary and dipped in ethanol. It was then inserted into a 21G precision needle which was attached to a 1 mL syringe filled with ethanol. The hollow fiber membrane was attached to the needle with wax, which also served to form an airtight seal between the hollow fiber membrane and the needle. The 1 mL of ethanol was then slowly pumped through the hollow fiber membrane using the syringe. The ethanol wash serves to remove the preserving fluid from the interior of the membrane and clean it for use in the experiment. Next, the hollow fiber membrane was removed from the needle and attached to another needle, this time attached to a 1 mL syringe containing a 10 mM concentration of NaCl and the sPSL particles. The salt solution was pumped through the hollow fiber membrane, filling it. The exposed end of the membrane was then sealed with a very small amount of wax and then carefully inserted into the capillary. The membrane was then severed from the syringe and the entire system was sealed with wax, which also served to attach the remaining end of the system to the glass slide. These experiments were repeated with deionized water in the capillary and membrane, for control experiments, and with the positions of the deionized water and salt solution reversed. The placement of the sPSL particles was also changed to outside the hollow fiber membrane for some experiments.

Once the slide was completely set up, the experiment was immediately taken to the light microscope for observation. After the lens was focused on the center of the hollow fiber membrane in the z-axis, a video was taken of the experiment. Most observations were done at a 10x magnification.

Particle velocities were obtained from the recorded videos. Using Imaging Elements Software, particle velocities for various times of the videos were obtained. A particle was selected and its position was marked. A short time interval later, usually 10 seconds, the new position was noted and the velocity was determined by the distance moved the time span. Only the velocity perpendicular to the hollow fiber
membrane was of interest because diffusiophoresis only generates motion along that axis. The motion parallel to the hollow fiber membrane would be mostly Browning motion. Thus, the standard automatic tracking software was mostly unsuitable for this experiment because it tracks two-dimensional velocity. Additionally the tracking software’s discreet time intervals were very small, meaning that, even along the correct axis, it would mostly be recording velocities due to the small but relatively quick back and forth movements due to Browning motion rather than the slow overarching motion towards the membrane caused by diffusiophoresis. Thus, though manual tracking was a slower option, it provided a much more accurate representation of the effects of diffusiophoresis.

The measurement of the chloride concentration profile was performed by graduate students Abhishek Kar and Rajarshi Guha. The chloride concentration profile was measured via the quenching of the luminescent dye, Lucigenin. Lucigenin is a compound that exhibits chemiluminescence that is quenched by chloride ions. A system using Lucigenin for measurement can determine the concentration of chloride ions by the measuring the intensity of the luminescence produced. The fluorescent intensity output by the Lucigenin dye is related to the chloride concentration by the Stern-Volmer equation, given below.

\[ \frac{F_0}{F_{Cl^-}} = 1 + K_{Cl^-}[Cl] \]

The fluorescent intensity with a zero concentration of chloride ions is represented by the value $F_0$. $F_{Cl^-}$ represents the intensity at a given concentration of chloride. The Stern-Volmer constant is represented by $K_{Cl^-}$ and, finally, the chloride concentration is $[Cl^-]$. In control studies, the intensity values at various, known chloride concentrations, including zero, were mapped. The correlation between these values yielded a value for the Stern-Volmer constant of 365 mM$^{-1}$ for Lucigenin and chloride. This value was then used in the experiments to calculate the chloride concentrations within the experiment and map these values against distance from the hollow fiber membrane wall. This allowed us to map the rate at which the chloride ions diffuse in the system, comparing this to the predicted diffusion rate. The corroboration of the simulated model supports the hypothesis that a diffusiophoretic effect ought to be
present. Consequently, if the particle velocities behave as predicted, the motion can be ascribed to
diffusiophoresis with reasonable confidence.
Chapter 4

Results and Discussion

This chapter provides the key results of this research. Figure 10 shows a schematic of what we observed with our membranes during colloidal fouling. Figures 1 and 2 in the Introduction show the same setup without and with calcium carbonate micropumps, respectively. The exclusion region and lack of fouling in Figure 2 in comparison to Figure 1 shows the effectiveness of the micropumps and potential to use diffusiophoresis to mitigate membrane fouling.

Experiments were initially conducted to establish the effect of diffusiophoresis on the deposition of colloidal particles on a hollow fiber membrane. The deposition of particles over time and the velocities of the particles with respect to their proximity to the membrane wall were the variables of interest needed to show the effect of diffusiophoresis. Time lapse images are used to show that, in a concentration gradient, the negatively charged sPSL (sulfated polystyrene latex) tracer particles outside the membrane will move towards the membrane filled with NaCl and accumulate on the its walls. This is shown by the deposition of particles over time in Figure 8. When the sPSL is placed inside the membrane, it will form a band near the center of the membrane, as shown in Figure 9. Figures 8 and 9 show that diffusiophoresis can be used to exacerbate or mitigate fouling, respectively.
Figure 8. The diffusiophoretic effect causes particles to move towards the hollow fiber membrane. Over time, this causes an aggregation of particles on the membrane wall.

Figure 9. The particles are placed inside the membrane with the same salt and deionized water setup. The particles are repelled from the walls and migrate to the center of the membrane, forming a band of particles.
The deposition matches the predicted model of colloidal particle deposition on the hollow fiber membrane shown in Figure 10. The similarities in these figures show that there is particle motion affecting fouling due to diffusiophoresis. In the case of sPSL particles, this motion is away from the deionized water and towards the salt.

![Figure 10](image)

**Figure 10.** Model of the deposition of particles on the membrane over time, without the effect of diffusiophoresis, left, and with the effect, right.

The variation in this motion can be understood by mapping the velocities of the particles. Figure 11 is a plot of particle velocity versus distance from the hollow fiber membrane wall. It combines various times of the experiment. The decay shown by the trendline indicates that the motion due to diffusiophoresis is fastest near the hollow fiber membrane and slows as distance from the membrane increases.
The strength of the electric field is proportional to the velocity, as discussed in the background. Thus, the velocity profile should correlate with the chloride ion concentration profile. Figure 12 shows the chloride concentration in a hollow fiber membrane setup. Figure 13 quantifies this image and maps the concentration versus distance from the hollow fiber membrane wall and shows that it correlates with the predicted values. The trendlines of velocity and chloride concentration versus distance from the hollow fiber membrane was indicate a similar exponential decay as seen when comparing Figures 11 and
13, supporting the prediction that velocity of the particles is directly related to the chloride concentration gradient.

Figure 12. The Lucigenin dye is quenched by the presence of chloride particles, showing the distribution of ions. The brightness gradient in the Figure shows this distribution.

Figure 13. A graphical representation of the experimental results for the concentration gradient of the chloride ion gradient, compared with the predicted concentration gradient.
Upon showing the effect of diffusiophoresis on the deposition of particles on the membrane, the next step is to show a viable method of using the process to slow or even reverse this accumulation, thereby reducing the rate of fouling. One method is to employ a salt gradient to directly counter the deposition. Diffusiophoresis employed in one direction will rapidly increase the rate of fouling, as in Figure 8. By changing its direction, this same electrokinetic process can be used to counter fouling. As shown earlier in Figure 9, a salt gradient can be used to keep particles away from the membrane wall in the same manner that it can be used to attract particles to the wall. By reversing the position of the salt with respect to the deionized water and leaving the sPSL particles in the same position, the particles will be forced to the center of the membrane, rather than aggregating on the wall.

Another, more practically feasible way to counter membrane fouling is through the use of calcium carbonate micropumps. The calcium carbonate particles settle on the membrane wall and, when placed in the same solution as the NaCl, generate an electric field in the opposite direction of the NaCl salt gradient. This creates an exclusion region as seen in Figure 14.
Figure 14. Model of the effect of Calcium Carbonate micropumps. They attract the particles by creating an electric field in the opposite direction as the diffusiophoretic effect. This counters the particle aggregation and creates an exclusion region around the membrane.

The exclusion region generated can be compared with the same setup without the calcium carbonate micropumps to show their effect of preventing fouling, as seen in the models of Figure 15. Figure 2 in the Introduction shows this same comparison using experimental evidence demonstrating the prevention of fouling.
**Figure 15.** Diagram of the diffusiophoretic effect with and without the presence of Calcium carbonate micropumps. Without the micropumps, there is an aggregation of particles on the membrane, but with the micropumps there is not only no aggregation but also an exclusion region.

A comparison of Figure 2 with the fouling created in the same setup but in the absence of the calcium carbonate, as in Figures 1 and 8, shows that these particles are very effective at preventing fouling. The microparticles essentially generate a stronger electrophoretic effect in the area near the membrane wall where they settle and, thus, prevent the aggregation of particles on the membrane, even after considerable time has passed. As shown in the Figure 2, initially the particles are attracted to the hollow fiber membrane by the effect of the sodium chloride gradient. This is the same effect as shown earlier. However, the effect relaxes after a short period of time because it is a finite setting. Though the sodium ions diffuse more slowly than the chloride ions, both types of ions soon reach the limits of the system, and the gradient decays relatively quickly. The electric field generated by the calcium carbonate does not relax nearly as quickly as that of the sodium chloride. Therefore, at a time of 4 minutes into the
experiment, the exclusion region has formed, as seen in Figure 2. Without the calcium carbonate, the increase in particle aggregation would have continued past the 7 minute mark, as it did in Figure 8. The particles that initially were accumulating close to the membrane early in the experiment have been repulsed by the effect of the calcium carbonate. This exclusion region remains effective for a much longer period than the effect of the sodium chloride, as shown by the image of the experiment after 15 minutes in Figure 2.
Chapter 5

Conclusion and Next Steps

These experiments sought to answer the following questions:

1. Do the transient salt gradients in the membrane systems of interest induce diffusiophoresis, and is this diffusiophoresis an explanation for the motion of tracer particles in these transient salt gradients?

2. Can calcium carbonate micropumps be used to created diffusiophoretic motion away from the membrane and thereby prevent fouling?

The results of these experiments supported the hypothesis that diffusiophoresis can be an active mechanism for the motion of particles near a membrane and, as such, can be used to counter the fouling of a membrane due to these particles. The first set of experiments showed that the chloride concentration gradient and the zeta potential of the tracer particles create the appropriate conditions for diffusiophoresis, establishing the presence of the electrokinetic phenomenon. The next set demonstrated that motion of the particles near the membrane in a transient salt gradient behave as predicted by the numerical model of particle motion due to diffusiophoresis. Combined, these experiments support the first component of the hypothesis, that diffusiophoresis provides an explanation for the motion of particles in a transient salt gradient within a membrane module. Next, experimentation with calcium carbonate micropumps showed that they can be used to prevent fouling, creating an exclusion region around the membrane as predicted by the numerical analysis of diffusiophoretic motion. Thus it was shown that diffusiophoresis is an active mechanism for the motion of colloidal particles in the membrane system and that it can be used to prevent the fouling of a membrane for extended periods of time through the use of calcium carbonate micropumps.

Since the ability of calcium carbonate micropumps to stop fouling due to the diffusiophoretic effects of a salt gradient has been demonstrated experimentally, its potential to stop fouling due to other effects should be investigated. While hollow fiber membrane systems do not utilize significant pressures,
other membrane filtration systems experience significant fouling due to the pressure gradient. Calcium carbonate micropumps, as well as micropumps from other chemical species, could prove to be an effective method of preventing fouling in these systems as well. Such an experiment would also be useful in quantifying the deposition mitigation abilities of the calcium carbonate. Using a controlled concentration of the micropumps, an experiment could test the maximum pressure at which the micropumps are able to create a visible exclusion region. This maximum pressure could be tested for various concentrations and species to see the effect that the quantity and type of the micropumps has on their ability to control fouling. Additionally, for a constant concentration of calcium carbonate, the size of the exclusion region at a constant time in the experiment with varying pressure could be tested. These two experiments would be a means of quantifying the effect of the micropumps as well as testing their viability in preventing membrane fouling due to pressure differential. These studies would be the next step in building on the results of this thesis, which has provided experimental evidence for the effect of diffusiophoresis on particle motion in membrane systems and the potential to use this effect to mitigate membrane fouling. Eventually, this information could be used to increase the effectiveness and consequently utility of membrane systems in a variety of settings.
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Objective: Currently seeking a Mechanical Engineer position, which challenges my critical thinking skills in a team environment, beginning after my May 2015 graduation.

Education:
The Pennsylvania State University, University Park, PA
- Candidate for Bachelor of Science in Mechanical Engineering, expected graduation May 2015.
- Candidate for Honors in Chemical Engineering, Schreyer Honors College.
- Extensive course work using Microsoft Office Programs, Solidworks, MATLAB, and Minitab.
- Participated in Study Abroad Program in France for a semester, as part of pursuing a goal of becoming trilingual. Current language abilities are: Spanish (Intermediate), French (Intermediate)

Work Experience:
Process Development Engineer Intern, Keurig Green Mountain Inc., Waterbury VT May—August 2014
- Assisted Process Development Manager in evaluating and qualifying process technology/processing parameters leading to the ability to quickly produce new beverages.
- Designed experiments, established project timelines, and recorded findings from experiments.
- Solved specialized technical problems relating to powder mechanics and solid/liquid interactions.
- Worked extensively with operations and manufacturing plants to ensure the successful production start-ups of new beverage products and to determine production equipment for new beverages is fully-functional.
- Partnered with the several other R&D functional groups including product development, analytical services, and the sensory test center to develop and test products.
- Conducted appropriate studies to demonstrate the ability of the product/device to function as intended in accordance with Keurig’s CQV policy (i.e. verification and validation process).
- Used statistics based experimentation and principles of process excellence. Frequently generated and reviewed SPC data using Minitab for new and commercialized products.
- Used SolidWorks to create a sampling system to enable in-flow sampling of processed powders.
Undergraduate Research, Velegol Laboratory, University Park PA

September 2012-May 2013

- Worked in the laboratory of Dr. Darrell Velegol, experimenting with colloids and studying their motion in the flows created by salt gradients and the potential application of this principal to slowing the fouling of membranes, particularly in water purification.
- Extensive training in light and confocal microscopy for the purpose of studying particles across multiple layers of a cylindrical membrane, as well as tracking software to quantify and analyze particle motion.
- Assisted in the training of new lab students upon completion of microscopy training.
- Designed experiments for proving the effects of salt gradients on particle motion, experimenting with various salts, concentrations, and experimental setups.

Activities and Interests:

Springfield THON, University Park PA

- Member of one of the organizations in charge of Penn State’s THON, a charity raising funds for research to fight pediatric cancers as well as assist patients and their families.
- Assisting in fundraising efforts, particularly canning, around Pennsylvania.