

Evaluation of GE Water & Process Technology's UF Membrane at Various Water Quality Parameters

Name: Doo Hyun Chung (dchung@hmc.edu), Harvey Mudd College

Advisors: Dr. Jaehong Kim (jaehong.kim@ce.gatech.edu)

Dr. Pyung Kyu Park (pyung.park@ce.gatech.edu)

Department of Civil and Environmental Engineering, Georgia Institute of Technology

HMC Liaison Advisor: Dr. Nancy Lape (lape@hmc.edu)

Department of Engineering, Harvey Mudd College

The main limitation of the ultrafiltration (UF) process in drinking water treatment is membrane fouling. This constant-flux, pilot-scale research evaluates General Electrics (GE) Water & Process Technology's UF211/UFC211 membrane at various source water conditions such as different humic acid (HA) concentrations, turbidity, pH, alkalinity and calcium ion concentrations. Different backwashing intervals were also tested to see what effects backwashing has on membrane fouling. Membrane performance was evaluated in terms of pilot plant run time and the resulting foulants were characterized. The results show that humic acid fouls membrane by adsorption and pore blocking while bentonite is mainly responsible for forming a cake layer on the surface of the membrane. pH and alkalinity did not affect the membrane performance. High calcium concentrations were found to change HA characteristics dramatically and GE's UF membrane was found to be incapable of filtering out this transformed HA. Shorter Backwashing interval improved backwashing efficiency. However, presence of calcium dramatically reduced backwashing efficiency. For further applications, these results can be used to optimize the best operating configurations for a filtering unit to process source water of different qualities.

I. Introduction

Since 1990's the applications of membrane processes such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) have rapidly increased in full-scale water and wastewater treatment [1-4]. Especially, during the past decade UF has emerged as one of the most reliable, cost-effective, and sustainable unit process for the production of drinking water [5]. Capillary UF is usually used as a final treatment step in advanced treatment plans all over the world to successfully remove viruses, bacteria and the finest particles. Today, such plant treats different types of waters (mainly surface water and ground water, typically of high turbidity) at a rate between 1000 m³/d and 150,000 m³/d [6].

UF technology has been adopted and developed by General Electrics (GE) Water & Process Technology and currently, GE's filtration module with UF membrane is filtering faucet water to provide drinking water of highest quality to regular households. Recently, GE has shown interest in bringing the same membrane technology out of households where the pre-filtered water quality is much worse off. The ultimate goal of this project is to be able to bring a filtering machine to places where fresh water is scarce, such as developing countries or tsunami sites, quickly take samples of local water, analyze the water quality and finally operate the filtering machine at the optimal settings to provide fresh water to people in need. The objective of this preliminary research is to evaluate the performance of GE's UF membrane at various conditions such as different normal organic matter (NOM) loadings, turbidity, pH, alkalinity, calcium ion concentrations, and backwashing interval.

It is generally accepted that NOM represent main foulant for surface waters. Within NOM, humic substances and polysaccharides were shown to be critical to UF due to membrane adsorption and pore blocking [7-12]. Humic substances are refractory anionic macromolecules of low to moderate molecular weight. They contain both aromatic and aliphatic components with primarily carboxylic and phenolic functional groups with carboxylic functional groups accounting for 60%-90% of all functional groups. Therefore, humic substances are generally negatively charged in the pH range of natural waters [8, 13].

Humic substances in water stabilize inorganic particles such as bentonite by readily adsorbing to mineral surfaces [14]. Mallevialle et al. [15] characterized the fouling layer formed by NOM in presence of inorganic particles. They found that the fouling layer was composed mostly of clay and organic matter, which was found to be packed under the inorganic fouling layer. Previous studies also revealed that NOM adsorption is influenced by divalent cations and pH [16-18]. Especially, calcium had very strong influence on destabilizing the particles [19, 20]. Therefore, the combined interactions between humic substances and other substances in aquatic environments are very complicated, even more so in membrane studies as the fouling mechanism not only deals with chemical interactions but also physical ones.

There have been many extensive studies to investigate the exact fouling mechanisms of specific substances. In this particular study, the focus is not so much on understanding the exact fouling mechanisms. Rather, more emphasis will be put on generating big pictures of the effects of particular water quality parameters on membrane fouling. In this paper, direct-flow UF using outside-in hollow fiber membranes with source water of various quality and different operating conditions is analyzed to find the effect of each water quality parameter on the membrane performance in terms of pilot plant run time and its cake characteristics.

II. Calculations

Calculation of Permeate Flux

The permeate flux J ($L T^{-1}$) is calculated by

$$J = \frac{Q}{A_m} \quad \text{--- Equation 1}$$

where Q ($L^3 T^{-1}$) is the volumetric flow rate and A_m (L^2) is the total outside area of the membrane fibers.

Categorization of Fouling Materials

For a fouled membrane unit, the fouling materials were split into three categories: i) deposited particles in form of reversible cakes that can be eliminated by a process of running water and air across the surface of the membrane fibers called flushing, ii) absorbed or gelled organic and inorganic compounds, which can be cleaned by chemical cleaning, and iii) any other compounds that cannot be removed by chemical cleaning. Resistance was correspondingly categorized as cake resistance (R_c , type i), reversible resistance (R_{rf} , type ii) and irreversible resistance (R_{if} , type iii).

Cake Filtration

The permeate flux J ($L T^{-1}$) of a fluid with absolute viscosity μ ($ML^{-1} T^{-1}$) during filtration under a transmembrane pressure ΔP ($ML^{-1} T^{-2}$) is given by Darcy's law as

$$J = \frac{1}{A_m} \frac{dV}{dt} = \frac{\Delta P}{\mu(R_m + R_c + R_{rf} + R_{if})} \quad \text{--- Equation 2}$$

where R_m (L^{-1}) is the membrane resistance. This equation is used to calculate the resistance of each type of foulant which corresponds to how much foulant of each type is present after filtration.

III. Materials and Methods

Materials

Calcium chloride and bentonite were purchased from Fisher Scientific (Pittsburgh, PA). Humic acid sodium salt was obtained from Sigma-Aldrich (St. Louis, MO). 1 N HCl was prepared by diluting 37% HCl (Acros Organic, Morris Plains, NJ). 1 N NaOH was prepared by dissolving NaOH pellet (Fisher Scientific, Pittsburgh, PA) in water purified by the Milli-Q water purification system (Millipore, Billerica, MA). Humic acid stock solution was prepared by adding 2 g of humic acid sodium salt to 1 L of Milli-Q water. Bentonite stock solution was prepared by adding 10 g of bentonite to 1 L of Milli-Q water. UF membrane (Model UF211 / UFC211) was provided by GE Water & Process Technology. Its specifications appear in Table 1.

Table 1: Specifications of UF membrane used in this research

Manufacturer	GE Water & Process Technologies
Model	UF 211 / UFC 211
Size (l x w x h)	63.5" x 16" diameter
Inside Diameter of Fiber	0.016" (0.4mm)
Outside Diameter of Fiber	0.032" (0.8mm)
Flow Direction	outside in
Membrane Construction	Hollow fiber
Membrane Material	Polyvinylidene Fluoride (PVdF)
Membrane Surface Characteristics	Hydrophilic
Membrane Charge	<slightly> negative /positive Non ionic
Nominal Molecular Weight Cutoff	100,000 Daltons
Nominal Membrane Pore Size	0.02 microns

Synthetic Source Water

Source water was prepared by the following procedure: 1) Deionized (DI) water provided by Georgia Institute of Technology's lab facility was poured into a 20-L container up to about 80% of total volume; 2) Sodium Bicarbonate ($NaHCO_3$) was added to control alkalinity (0, 50, 150 mg $CaHCO_3/L$); 3) Calcium Chloride ($CaCl_2$) was added to control calcium concentration (0, 0.5, 5 mM); 4) Humic acid was added for NOM control (0, 2, 10, 50 mg C/L); 5) Bentonite was added to control turbidity (0, 5, 20, 50 NTU); 6) pH was controlled at 5, 7, and 9 with 1 N HCl and 1 N NaOH; 7) DI water was poured into the container to make total volume 20 L. The solution was thoroughly mixed after each step to avoid any possible chemical change due to uneven concentration gradient as humic acid is reported to change for different concentrations of metal ions such as calcium [21]. The temperature for all source water was maintained at 21- 22 °C.

Water Quality Measurements

When the source water was prepared, samples were taken to measure humic acid concentration, turbidity and pH. Humic acid concentration in stock solution was measured as total organic carbon (TOC) by a TOC analyzer (TOC-V ws, Shimadzu Scientific Instruments, Columbia, MD), and as UV absorbance at 254 nm (UVA254) by a spectrophotometer (8453

UV/VIS spectrophotometer, Agilent Technologies, Inc., Santa Clara, CA). Once UVA254 was calibrated with TOC, only UVA254 was used to monitor humic acid concentration as UVA254 correlates to TOC measurements [22-24]. Before each UVA measurement, samples were filtered with 0.45 μm membrane filter (Supor®-450 PALL Life Science, Port Washington, NY). Turbidity was measured with a Hach Corporation's turbidimeter (Hach 2100N, Hach Company, Loveland, CO). Finally, pH was measured with Accumet AR50 pH meter (Fisher Scientific, Pittsburgh, PA).

Membrane Module

Schematic of the hollow fiber membrane module used in this experiment is shown in Figure 1. The module's housing is a transparent circular acrylic resin cylinder with openings at labels (1), (2), (3), (4). 20 hollow-fiber membrane fibers of approximately 20 cm in length run across openings (1) and (4). Real picture of a module appears in Figure 14 in Appendix.

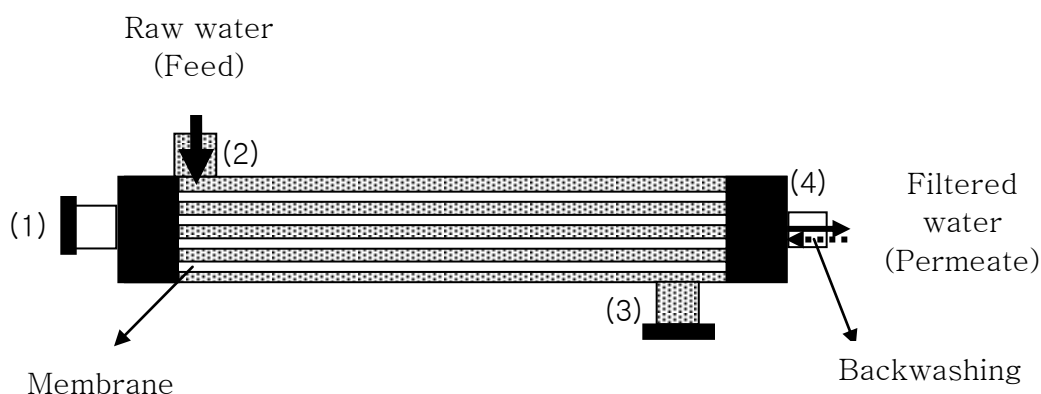


Figure 1: Schematic of Membrane Module

Membrane Pilot Plant and Operation

Schematic of membrane pilot plant is provided in Figure 2 (Real pictures of the plant appear in Appendix). Source water in the feed tank gets pumped by a peristaltic pump (Cole-parmer Instrument Co., Vernon Hills, Illinois) into the membrane. Water enters in opening (2) of the membrane module (Figure 1). Because opening (3) is blocked off, pressure is established in the membrane. Then, the pressure difference in and out of membrane (transmembrane pressure) forces source water to diffuse into the membrane and the water gets filtered. With opening (1) blocked off, filtered water exits out of opening (4) and flows into permeate reservoir. Digital pressure meter and flow meter record data directly to a LabView program on a PC computer. However, the flow rate of the experiment was too small for the digital flow meter to accurately measure. Therefore, all flow rates were measured manually with a stopwatch (to measure time) and a scale (to measure weight, therefore volume). Flow rates were maintained at constant to run all experiments in constant-flux mode. All experiments were run at a starting transmembrane pressure of 9.5 psi and they were stopped when the transmembrane pressure reached 20 psi. Samples were taken three times during the pilot plant operation: 1) After 0.5 hour 2) At 14.5 psi 3) At 19.5 psi. UVA254, turbidity and pH were measured for each sample.

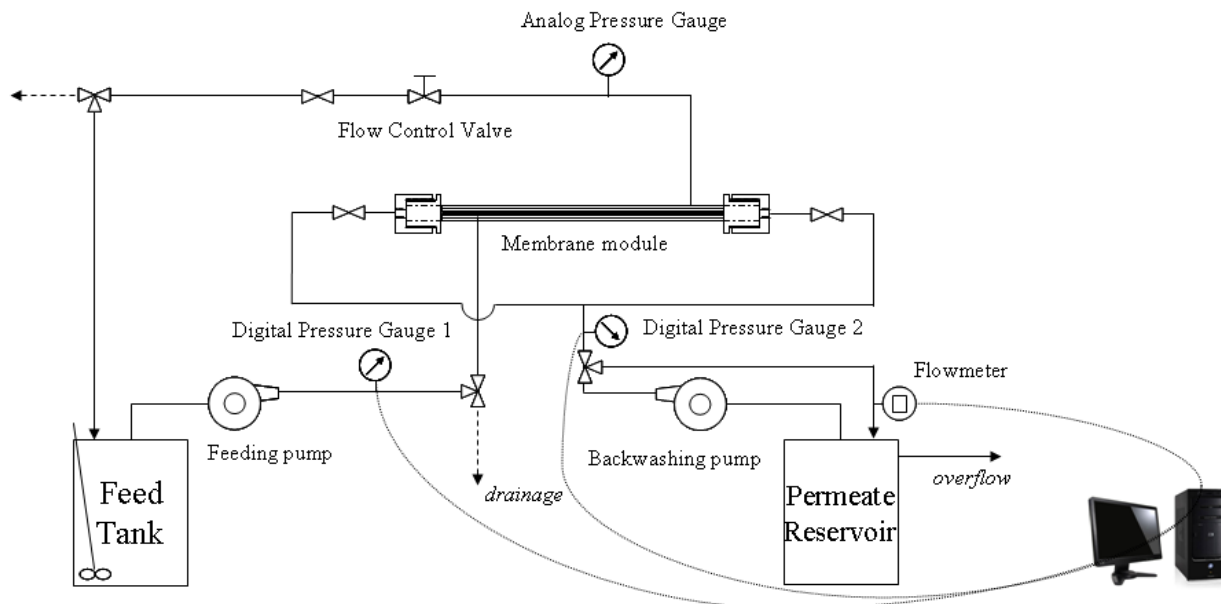


Figure 2: Schematic of Membrane Pilot Plant

Backwashing Procedure

The entire sequence of backwashing was programmed into a programmable logic controller (PLC) (Master K-80s, LS Industrial Systems, South Korea). In the first step of backwashing, a backwashing pump (Cole-parmer Instrument Co., Vernon Hills, Illinois) pre-pressurized a part of permeate water for backwashing. After 2 seconds, the 3-way valve connected to the backwashing line changed in the flow direction to start backwashing and the valve for drainage was opened. The backwashing pump was controlled to maintain a backwashing pressure of 25 psi. Therefore, water enters in opening (4) (Figure 1) and the backwashing pressure drives the water out of the membrane. As water diffuses out of the membrane, it washes off the cakes composited on the surface of the membrane. At the end of backwashing, the backwashing pump stopped 2 seconds before the direction of 3- way valve got changed back to the original position. Then, the normal filtration process begins. Backwashing was done at three different settings: 1) No backwashing; 2) 30 min/1 min setting: Filter for 29 minutes and backwash for 1 minute; 3) 15 min/30 sec setting: Filter for 14.5 minutes and backwash for 30 seconds.

Cleaning Procedure

Foulants in form of deposited cake (Type i) were eliminated by flushing; 8 sets of DI water (5 minutes) and air (1 minutes) were flushed from opening (2) to opening (3) (Figure 1) at a cross flow rate of 0.1 m/s through the outside of the membrane fibers to remove any cake composited on the surface of membrane. The direction of the flow was switched every 2 sets. After flushing, permeability of the membrane was measured by recording the volumetric flow rate and the corresponding pressure when the membrane was used to filter Milli-Q water. Reversible foulants (Type ii) were removed by backwashing with pH 10 water for 2 hours (1 hour in each direction) and backwashing with pH 4.5 water for an hour (0.5 hour in each direction) with a backwashing pressure of 25 psi. Each backwashing procedure was accommodated by a cross flow of 0.01 m/s to carry the foulants out of the module. In a regular

backwashing procedure, water enters in (4). Because opening (1) is blocked off, the backwashing pressure drives water out of membrane, removing any remaining foulants from membrane pores and surfaces. Because cross flow enters in opening (3), water exits out to opening (2) and goes out to drainage. The reverse direction proceeds in the same manner from opening (1) to opening (3) and cross flow from opening (2) to (3). Permeability of the membrane was measured the same way as described above.

Table 2: Summary of Important Water Quality Parameters

Run	Date	humic acid concentration (cm ⁻¹)	turbidity (NTU)	pH	Alkalinity (mg CaHCO ₃ /L)	calcium concentration (mM)	backwashing
1	02-Jul-09	0	4.95	6.94	50	0.5	none
2	04-Jul-09	0.0639	4.96	6.92	50	0.5	none
3*	07-Jul-09	0.3102	4.99	6.99	50	0.5	none
4	08-Jul-09	0.8811	4.99	7.02	50	0.5	none
5	10-Jul-09	0.2913	0.356	7.06	50	0.5	none
6	13-Jul-09	0.2856	20.4	7.06	50	0.5	none
7	14-Jul-09	0.2813	50.4	7.04	50	0.5	none
8	15-Jul-09	0.2702	5.08	4.94	50	0.5	none
9	15-Jul-09	0.2975	4.93	9.03	50	0.5	none
10	16-Jul-09	0.2811	4.99	7.05	0	0.5	none
11	17-Jul-09	0.2954	5.02	7.03	150	0.5	none
12	17-Jul-09	0.2826	4.96	7.00	50	0	none
13	20-Jul-09	0.1027	5.05	6.93	50	5	none
14	21-Jul-09	0.2931	5.03	6.99	50	0.5	30 min/1 min
15	21-Jul-09	0.2931	5.03	6.99	50	0.5	15 min/30 sec
16	29-Jul-09	0.3109	5.04	7.07	50	0	15 min/30 sec

* indicates the standard condition

IV. Results and Discussion

Table 2 summarizes some water quality parameters that were controlled in this experiment. In order to better understand the effect of each parameter on the membrane performance, the transmembrane pressure (psi) was graphed with respect to time (hour) and the total run time (time it took to get from 9.5 psi to 20 psi) was compared for each parameter. In all transmembrane pressure graphs, the white square path indicates the standard condition (2 mg C/L humic acid, 5 NTU, pH 7, 0.5 mM Ca, 50 mg CaHCO₃/L, no backwashing).

Humic Acid Effect

Figure 3 depicts the effect of different humic acid concentrations on membrane performance in terms of pilot plant run time. The graph shows that higher concentration of humic acid has quite a strong influence on shortening the run time. This result is consistent with a general belief that NOM, especially humic substances, fouls UF membrane by adsorption and pore blocking.

The resistance graph of humic acid effect in Figure 4 also supports the same idea that humic acid mainly contributes to membrane adsorption and pore blocking. As humic acid loading is increased, reversible and irreversible fouling resistances, both of which correspond to the amount of foulants that exist in the porous area in the membrane, increase while cake resistance, representing the amount of foulants on the surface of the membrane, decreases. Therefore, humic acid fouls the membrane by adsorbing on the surface and getting gelled in the pores of the membrane.

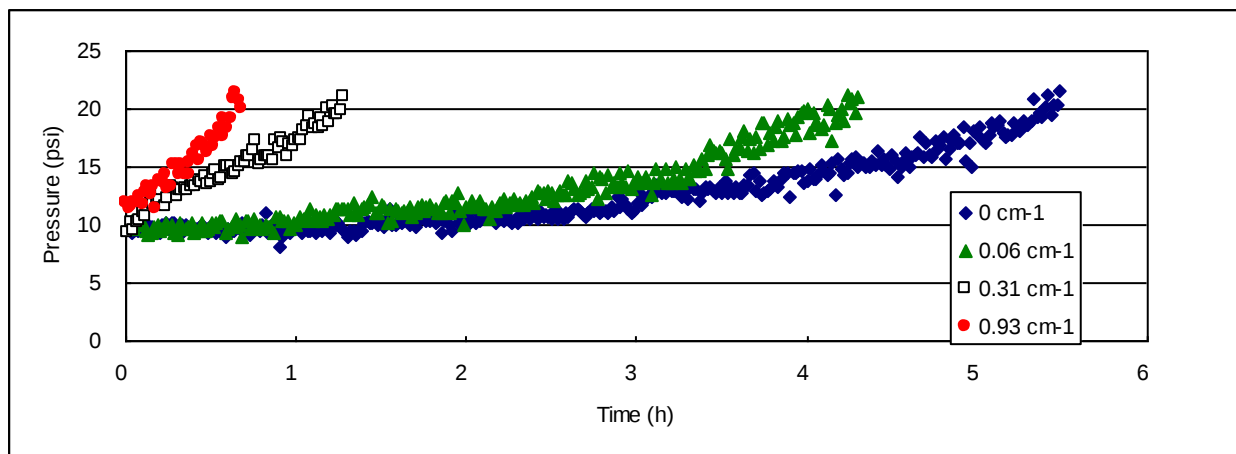


Figure 3: Effect of Humic Acid on Pilot Plant Run Time

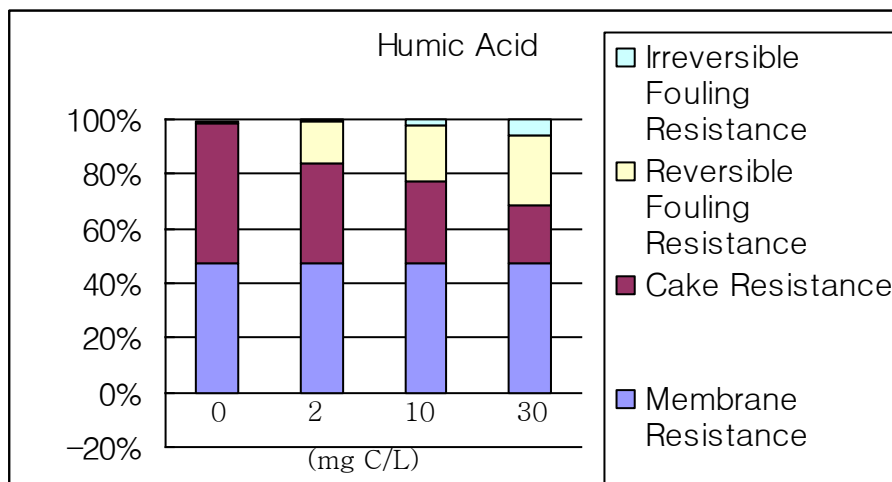


Figure 4: Hydraulic Resistances of Humic Acid Effect Experiments

Turbidity Effect

Figure 5 shows the effect of different turbidity on pilot plant run time. As shown by the graphs, increase in turbidity also decreases the total run time. Bentonite, which was used to control turbidity during the experiment, represents inorganic substances present in river. Because these inorganic substances are much bigger than the pore size of the membrane, they tend to accumulate on the outside of the membrane. In another study that investigated the interactions between NOM and kaolinite, it was found that NOM adsorbs onto the inorganic particle's surface and form a denser cake layer, resulting in synergistic fouling effect of NOM and kaolinite [5, 25]. A similar mechanism can be taking in place between humic acid and bentonite. Higher turbidity results in a formation of thicker cake layer which causes the transmembrane pressure to go up more quickly.

The hydraulic resistances graph of turbidity in Figure 6 shows increasing cake resistance in response to increasing turbidity. Although the exact cake formation models were not further explored, an increasing trend of cake resistance confirms that the cake layer is getting thicker as bentonite concentration increases. Therefore, it can be concluded that bentonite fouls the membrane by cake formation on the surface of the membrane.

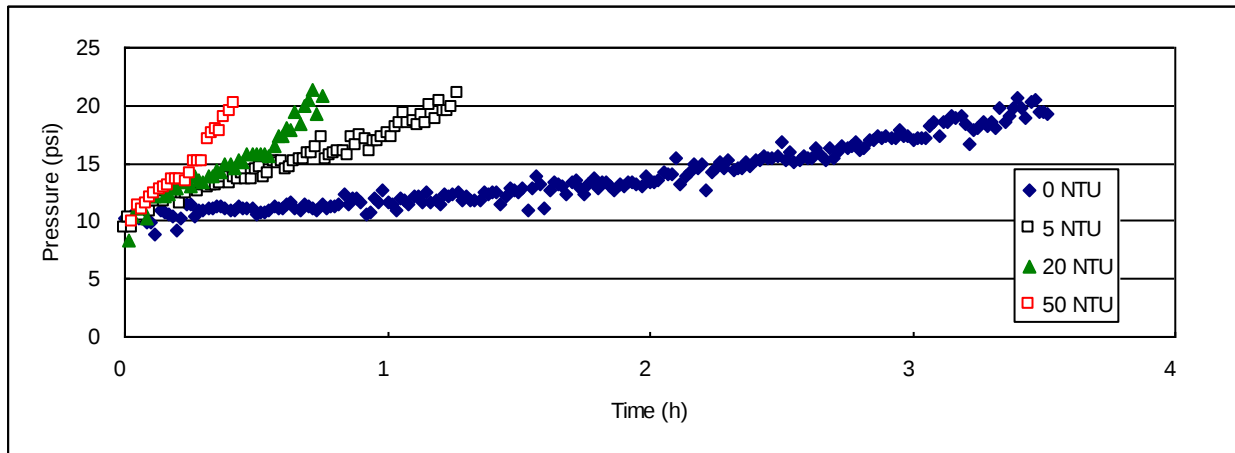


Figure 5: Effect of Turbidity on Pilot Plant Run Time

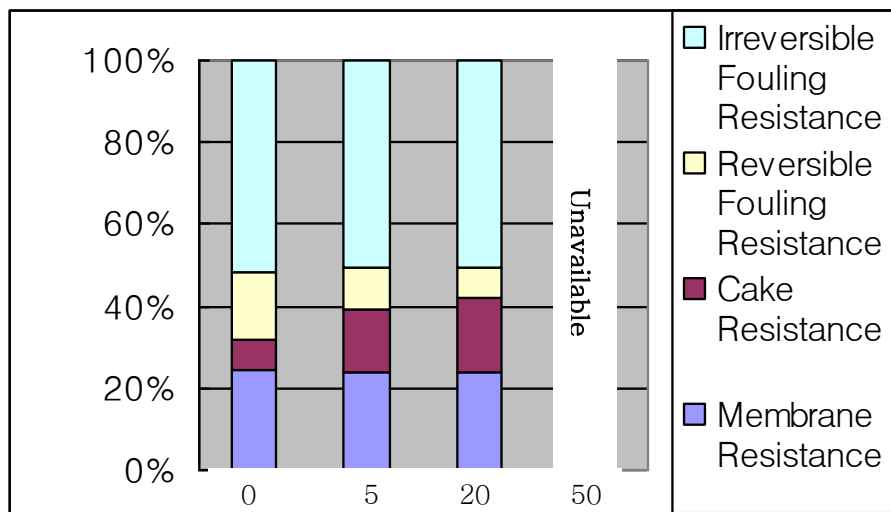


Figure 6: Hydraulic Resistances of Turbidity Effect Experiment

pH Effect

Figure 7 portraits the effect of pH on pilot plant run time. The graph shows no clear pattern that demonstrates pH effect on pilot plant run time. Small discrepancies among three graphs seem to be caused by random variation in source water synthesis. According to J. Cho et al. [24], pH only has a minor effect on NOM rejection. Their potentiometric titrations shows that NOM acidity—for both hydrophilic and hydrophobic acids—changes dramatically only in pH range of 3-4. In other words, pH does not really change the structure and characteristics of NOM in the pH range investigated in this study.

Hydraulic resistances graph of Figure 8 shows increasing tendency of irreversible fouling resistance with increasing pH. This tendency suggests that higher-pH source water fouls the membrane to a greater degree. However, the overall effect of pH on membrane fouling still seems very insignificant as shown by the run time graph in Figure 7.

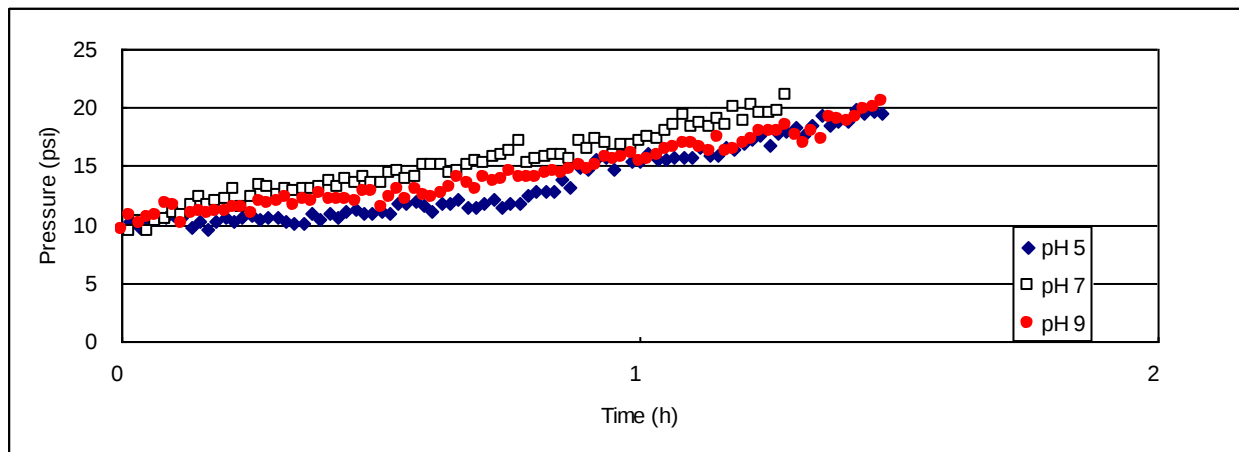


Figure 7: Effect of pH on Pilot Plant Run Time

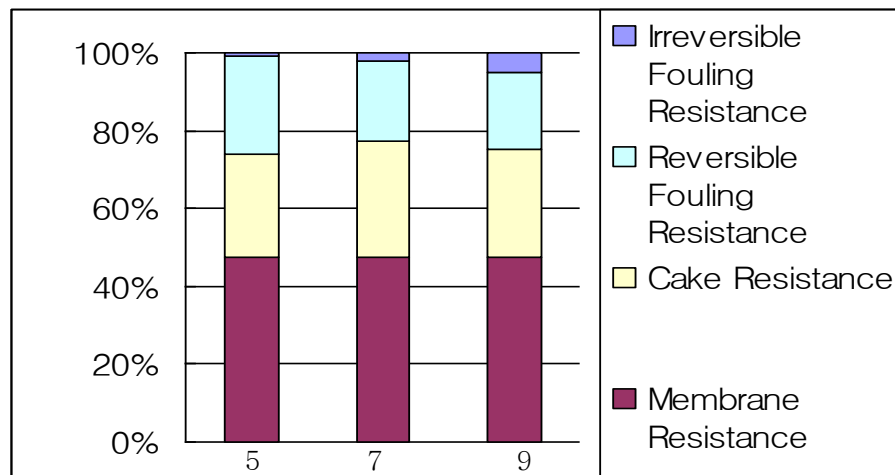


Figure 8: Hydraulic Resistances of pH Effect Experiment

Calcium Effect

Figure 9 shows the effect of calcium ion on pilot plant run time. When a small amount of calcium ion was present in the source water (0, 0.5 mM), calcium ion did not seem to affect the experiment a lot. However, when a substantial amount of calcium was added (5 mM), the experiment ran significantly longer than the other two. It was anticipated that higher concentration of calcium ion would decrease the electrostatic repulsive force of slightly-negatively-charged humic acid and the rejected molecules would bind together to form a more tightly-packed cake layer which would cause the transmembrane pressure to increase very rapidly and cut the run time very short. However, the experimental results indicate that some other mechanism is taking place.

Hong and Elimelech [13] also reported a similar unanticipated experimental result that higher calcium ion concentrations made almost no difference in NOM rejection through an aromatic-polyamide thin film composite (TFC) NF membrane. J. Cho et al. [24] presents a solution to this phenomenon by claiming that while intramolecular calcium ion binding is responsible for formation of thicker cake layer on the membrane surface, it also decreases the electrostatic interaction of the molecule and a pore mouth in which the molecule fits in. Therefore, decreasing the strength of the electrostatic repulsive force lets the molecule freely pass through the pores of the membrane, resulting in low NOM rejection.

It is important to note that UVA254 measurement of 5 mM-calcium synthetic source water was very low (Run 13, Table 1) compared to others even though a similar amount of humic acid was added each time. This fact suggests that high calcium concentrations changed the structure and characteristics of humic acid. Also, UVA254 measurements of feed and permeate samples of 5 mM-calcium water showed almost no difference, suggesting that the membrane was not effectively filtering out humic acid. Although intermediate calcium concentration conditions are missing due to time constraint, the experimental data correspond to J. Cho et al's claim that a high calcium concentration results in low NOM rejection.

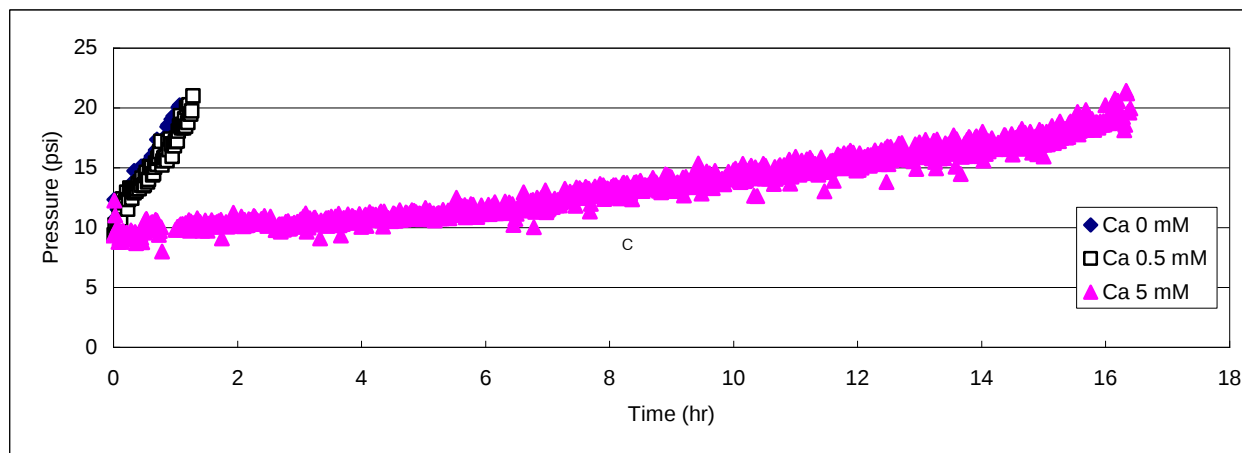


Figure 9: Effect of Calcium Ion on Pilot Plant Run Time

Alkalinity Effect

Figure 10 shows the effect of alkalinity on pilot plant run time. The three graphs of different alkalinity conditions show no apparent pattern in their relationships. Although the experiment with alkalinity 150 mg CaHCO_3/L ends faster than the other two conditions, it is not significant enough, as it was in the case of calcium effect experiment, to give it a special treatment besides random variation in source water synthesis.

Figure 11 also shows rather irregular variations in hydraulic resistances. Although the cake resistance of no alkalinity condition is lower than cake resistance of other two conditions, the graphs collectively as a whole do not offer any insight into why high alkalinity experiment stopped faster the other two. Therefore, it is concluded that alkalinity has no apparent influence on UF membrane performance.

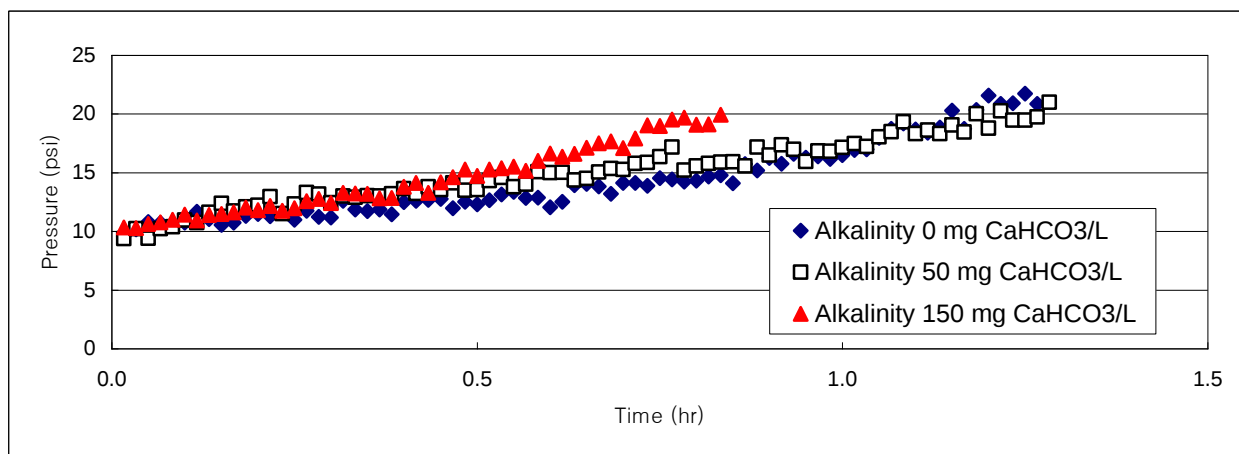


Figure 10: Effect of Alkalinity on Pilot Plant Run Time

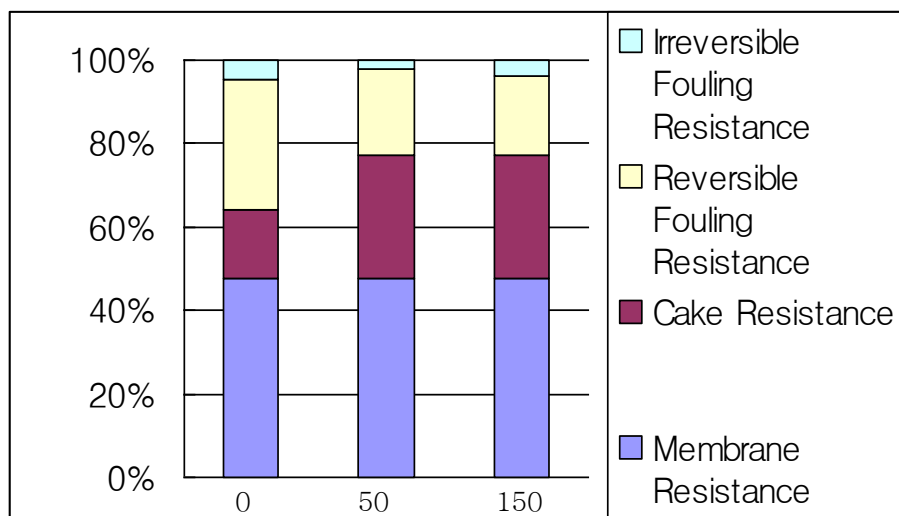


Figure 11: Hydraulic Resistances of Alkalinity Effect Experiment

Backwashing Effect

Figure 12 shows the pilot plant run times of no backwashing, 30 min/1 min backwashing, and 15 min/30 sec backwashing. For 30 min/1min and 15 min/30sec backwashing conditions, the recovery rate is the same because the ratio of the permeate volume to the feed volume is the same. However, the fact that 15 min/30 sec backwashing condition ran longer than 30 min/15 sec suggests that shorter backwashing interval is more effective than longer interval.

Figure 12 also shows that there is no apparent difference between 30 min/1 min backwashing condition and no backwashing condition. In other words, the backwashing was done at a very low efficiency. If the backwashing was done efficiently, the transmembrane pressure should drop at the moment of backwashing and the pilot plant run time graph should resemble a saw-tooth function, as observed in a previous experiment which was not included in this paper.

In order to find out the cause of the low-efficiency backwashing effect, source water with no calcium concentration was filtered at a backwashing rate of 15 min/30 sec since calcium seemed to have the greatest effect on pilot plant run time. Figure 13 shows that backwashing with no calcium condition ran significantly longer than normal backwashing condition did.

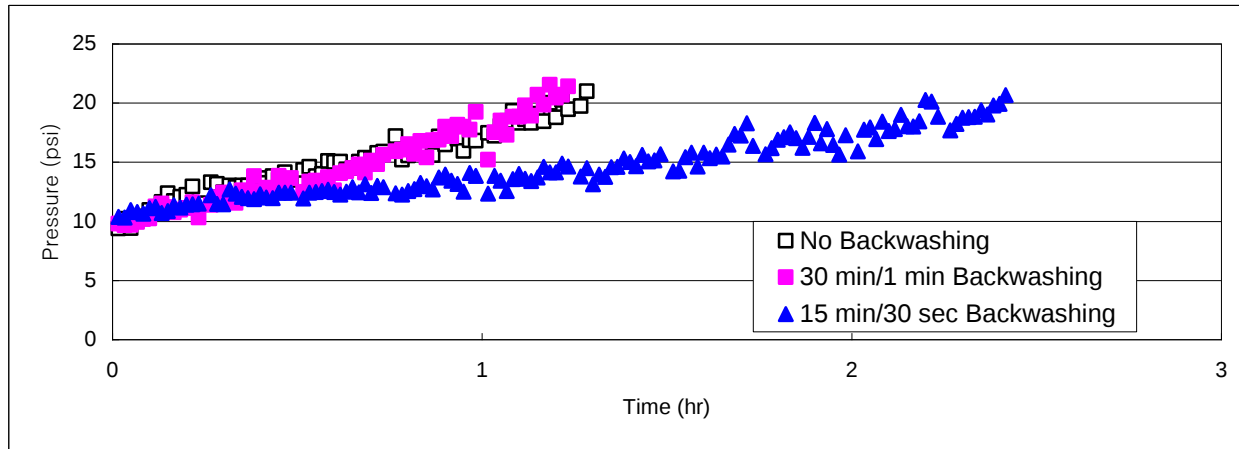


Figure 12: Effect of Backwashing on Pilot Plant Run Time

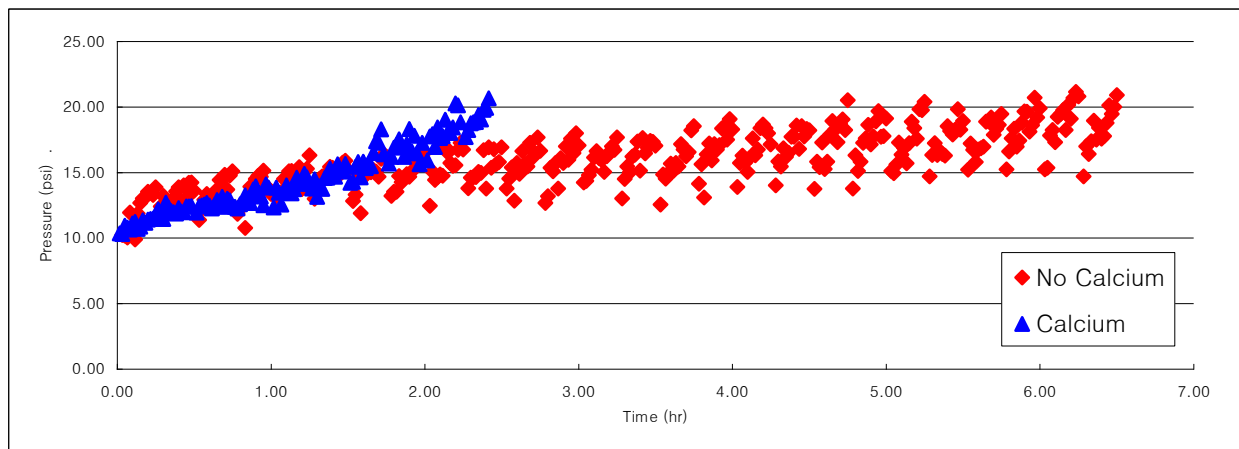


Figure 13: Effect of Backwashing with No Calcium on Pilot Plant Run Time

This result can also be explained by reduced electrostatic repulsive strength. When 0.5 mM of calcium was present in the source water, the cake layer was enhanced by reduced electrostatic repulsion. Because this cake layer was very tightly-packed, it was difficult to remove the cake very effectively as shown in the first backwashing experiment in Figure 12. However, when there was no calcium present, the cake layer could be easily removed by backwashing and the drop in transmembrane pressure at the moment of backwashing is clearly indicated in Figure 13. Therefore, shorter backwashing interval will always be a better choice for higher backwashing efficiency. However, if the source water contains high calcium concentrations, backwashing efficiency drops significantly. Therefore, a careful cost analysis of backwashing and membrane module change is necessary to find the cost-minimizing operating configuration.

V. Conclusion

The experimental results offer some insights into what the operating configurations should be for water sources of different qualities. If the source water contains humic substances of more than UVA254 0.31 cm^{-1} or if the turbidity is greater than 5 NTU, there may need to be some pre-treatment steps for a longer plant run time. The operating configurations should not depend on the pH or alkalinity of local water although there may be some complications if the pH drops below 4. When the source water contains high calcium concentrations, this particular membrane seems rather unable to filter out humic acid. Also, the backwashing efficiency drops significantly when calcium is present in the source water. If backwashing is to be done, shorter backwashing interval seems to be a better choice for backwashing efficiency. However, the long-term cost of shorter backwashing interval should be analyzed carefully to minimize the cost of operations.

There are two main areas that need further investigations for a complete evaluation of GE's UF 211/UFC 211 membrane. First, a more thorough picture of each parameter effect is necessary. Intermediate concentration conditions need to be tested for humic acid, turbidity, and calcium effects. Especially, calcium effect deserves a very meticulous investigation as calcium seems to complicate NOM characteristics dramatically.

Second, there needs to be a simple and non-costly method of analyzing the samples of local water to roughly estimate its humic substance concentrations, turbidity and calcium concentrations. Measuring devices used in lab facilities are very expensive and are not very portable. Therefore, this particular area may be the key to minimizing the cost of each filtering unit.

VI. Acknowledgements

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VII. Appendix

GE filtration systems

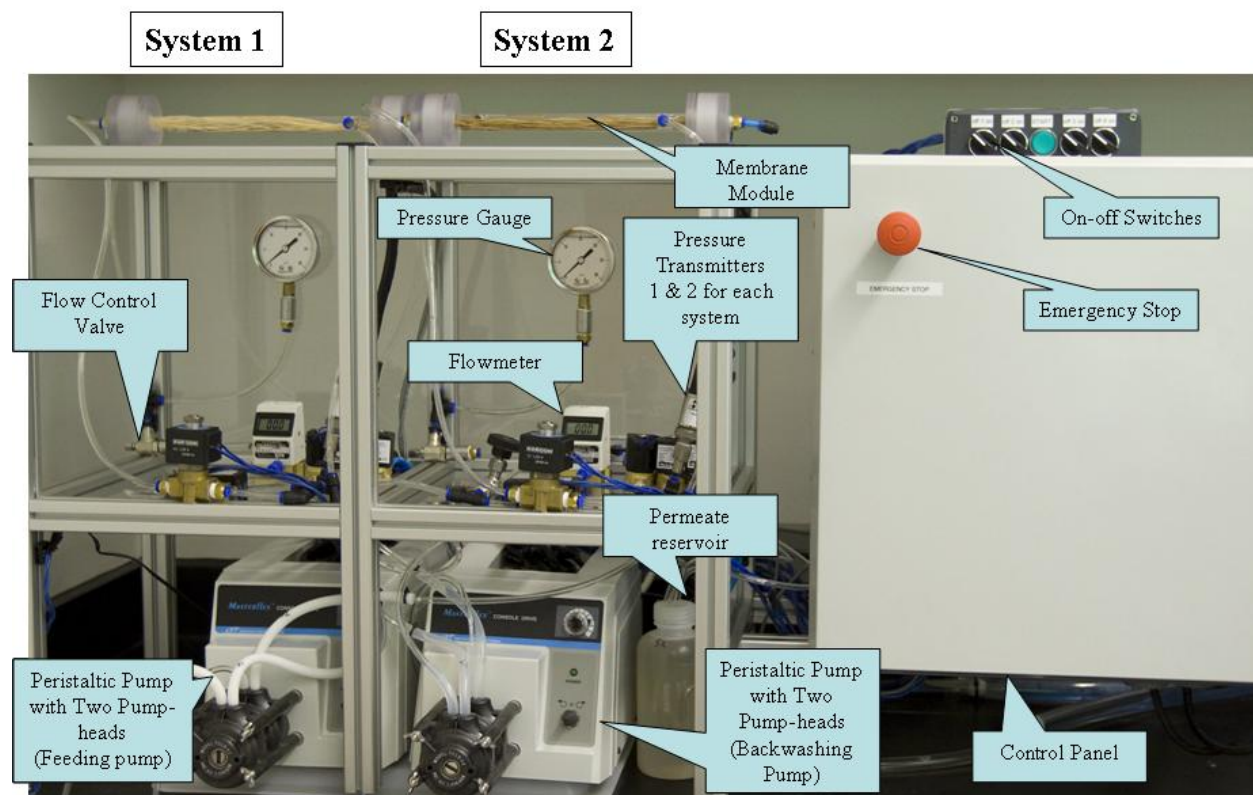


Figure 14: Overview of Filtration Systems

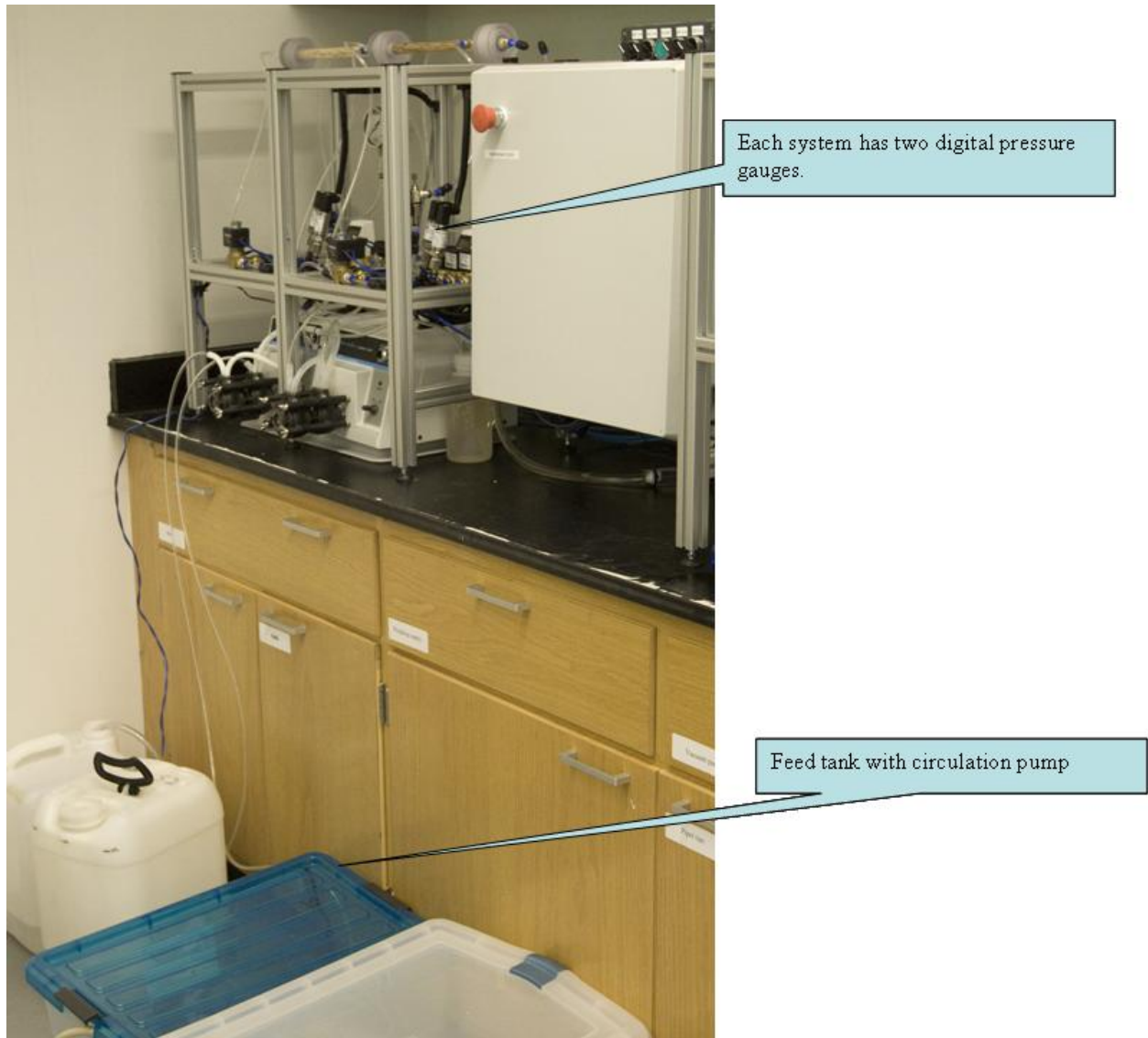


Figure 15: Filtration Systems with Feed Tank

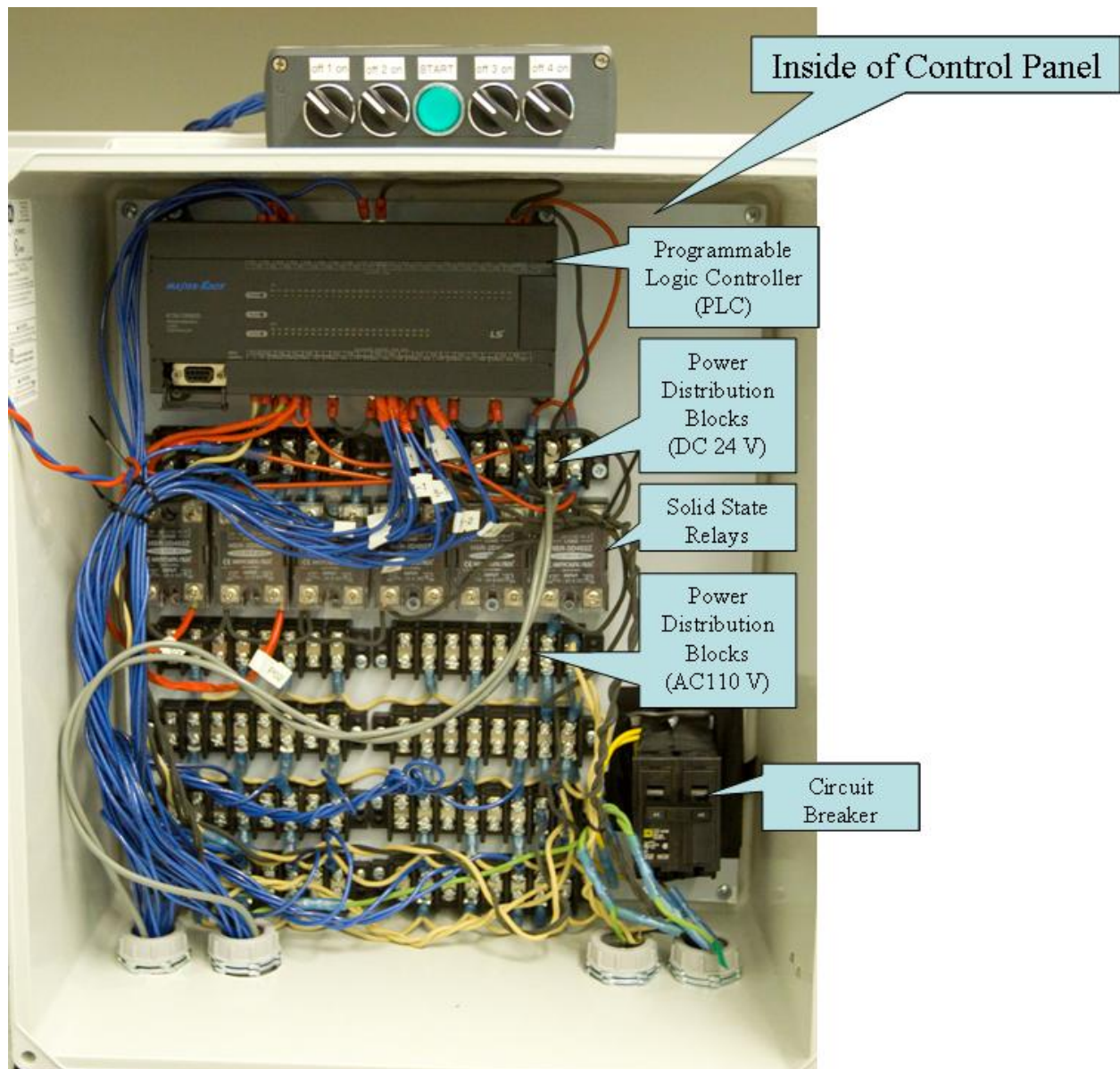


Figure 16: Inside of Control Panel

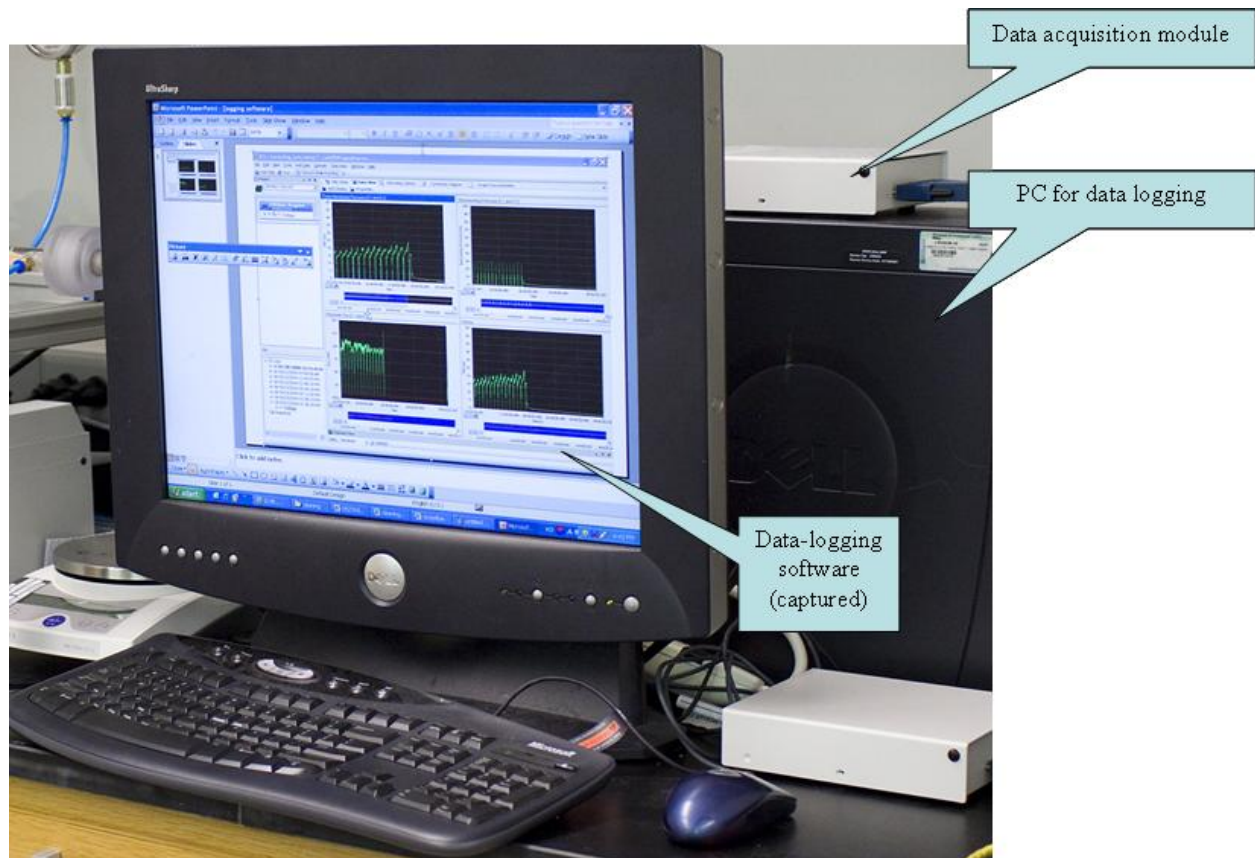


Figure 17: Data Acquisition and Analysis System