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Managing Water in the West

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No. 139

Electrocoagulation Pretreatment for Microfiltration: An Innovative Combination to Enhance Water Quality and Reduce Fouling in Integrated Membrane Systems

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14. ABSTRACT

A systematic and rigorous study was conducted with the aim of evaluating iron electrocoagulation for pretreatment of surface water prior to microfiltration. Companion experiments were also conducted using ferric chloride as a chemical coagulant in order to compare its performance with electrocoagulation. Fouling was not alleviated significantly by iron electrocoagulation in the entire range of experimental conditions investigated. Additionally, typically, conventional chemical coagulation outperformed electrocoagulation in terms of natural organic matter and virus removal since soluble ferrous iron was unexpectedly generated at the anode. It appears that the natural organic matter binds with Fe(II) and prevents its oxidation and subsequent hydrolysis to form $Fe(OH)_3(s)$ precipitates. Hence, conventional iron chemical coagulation is recommended over iron electrocoagulation for pretreatment of surface waters prior to low-pressure membrane filtration. It is also recommended to evaluate aluminum anodes in the future since they are expected to generate Al(III) during electrocoagulation producing $Al(OH)_3$ precipitates capable of destabilizing natural colloids and removing natural organic matter and viruses thereby improving membrane performance when used as a pretreatment process for microfiltration or ultrafiltration.

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Electrocoagulation Pretreatment for Microfiltration: An Innovative Combination to Enhance Water Quality and Reduce Fouling in Integrated Membrane Systems

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ACRONYMS AND ABBREVIATIONS

ATCC	American Type Culture Collection
AWWA	American Water Works Association chemical
CC	coagulation
CC-MF	chemical coagulation-microfiltration
CC-SF	chemical coagulation-syringe filtration
DAL	Double agar layer
DBP	disinfection by-products
D/DBP	Disinfectants/Disinfection By-Products
DI	de-ionized
DNA	deoxyribonucleic acid
DOC	dissolved organic carbon
EPA	U.S. Environmental Protection Agency
EC	electrocoagulation
EC-MF	electrocoagulation-microfiltration electrocoagulation-
EC-SF	syringe filtration
E. coli	*Escherichia coli*
GC/ECD	gas chromatography–electron capture detection
GC/MS	gas chromatography–mass spectrometry
HAA	haloacetic acids
IEP	isoelectric point
ICR	Information Collection Rule
IHSS	International Humic Substances Society
LRV	Log reduction value
MCLG	Maximum Contaminant Level Goal
MF	microfiltration
MTBE	methyl tert-butyl ether ()
NDIR	non-dispersive infrared gas analyzer
NF	nanofiltration
NIST	National Institute of Standards and Technology
NOM	Natural organic matter
NTU	Nephelometric Turbidity Unit
PFU	plaque-forming units
PTFE	polytetrafluoroethylene
PVDF	polyvinylidene fluoride
RO	reverse osmosis
RNA	ribonucleic acid
SDWA	Safe Drinking Water Act
SEM	scanning electron microscope
SUVA	specific ultraviolet absorbance
SWTR	Surface Water Treatment Rule
TDS	total dissolved solids
THMs	trihalomethanes
TOC	total organic carbon
TT	treatment technique
UF	ultrafiltration
UV	ultraviolet
UV254	ultraviolet absorbance at 254 nm and 1 cm path length

MEASUREMENTS

°C	Celsius
µL	microliters
µm	micrometer
A cm	amp centimeter
cm-^{1}	reciprocal centimeter
g	gram
Hz	hertz
kPa	kilopascal
L	liter
L/m^2-bar-h	liters per meter square hour bar
mA/cm^2	milliampere per square centimeter
mA	milliamps
mg	milligrams
mmole	millimole
mEq	milliequivalent
mg/L	milligram per liter
mL	milliliters
mL/d	milliliters per day
mL/min	milliliters per minute
mm	millimeters
mg/mmole	milligrams per millimole
mg/meq	milligrams per milliequivalent
meq/L	milliequivalent per liter
nm	nanometers
psi	pounds per square inch
psig	pounds per square inch gauge
rpm	revolutions per minute
s	seconds
V	volt

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Appendix A. Technical Notes

1. EXECUTIVE SUMMARY

1.1. Microfilter fouling and natural organic matter removal

The first part of this research considers a bench-scale study undertaken to evaluate iron chemical coagulation and electrocoagulation (EC) pretreatment for dead-end, constant pressure microfiltration (MF) of surface water. Fouling of a commercially available polyvinylidine difluoride (PVDF) membrane with conventional chemical coagulation pretreatment was found to:

(1) Be insensitive to pH in the range 6.4 – 8.3,

(2) Decrease only when greater than or equal to 10 mg/L Fe(III) were dosed, and

(3) Exacerbate with increasing transmembrane pressure.

These results were consistent with creation of larger flocs at high iron dosages that decreased cake specific resistance and formation of compressible cakes that increased hydraulic resistance at higher pressures. A substantial intermediate blocking stage was observed prior to cake formation during raw water microfiltration, whereas, cake filtration was the predominant flux decline mechanism for coagulated waters for almost the entire duration of microfiltration. Fouling was not alleviated significantly by iron electrocoagulation in the entire range of experimental conditions investigated since soluble ferrous iron was unexpectedly generated at the anode. Hence, conventional iron chemical coagulation is recommended over iron electrocoagulation for pretreatment of surface waters prior to low-pressure membrane filtration. Chemical- and electrocoagulation pretreatment decreased ultraviolet (UV) absorbance to a greater extent than dissolved organic carbon (DOC). Hence, disinfection by-product control by coagulation-microfiltration is achieved by decreasing the concentration of natural organic matter (NOM) precursors as well as their reactivity towards chlorine.

1.2. Virus removal

The main objective of the second part of the research was to study the performance of the electrocoagulation-microfiltration (EC-MF) process for virus removal in natural water containing NOM and particulate matter. EC was found to generate mostly Fe(II), not Fe(III), in the pH range 6.4-7.5. The presence of

NOM in natural water was found to have a detrimental effect on virus removal. It appears that the NOM binds with Fe(II) and prevents Fe(II) oxidation and subsequent hydrolysis to form $Fe(OH)_3(s)$ precipitates that can effectively remove the viruses. Unlike synthetic water where EC-MF achieved greater than 6-log removal at pH 6.4 using 12.8 milligrams per liter (mg/L) of iron, EC-MF was not efficient for virus removal from natural water where less than 2-log removal was observed at pH 6.4-7.5 even with high iron dosages up to 13 mg/L. Further, Fe(II) coagulation experiments indicated a significant decrease in log virus removal from 3.5 to 0.01 as the concentration of Suwannee River NOM increased from 0 to 2.7 mg/L DOC.

2. INTRODUCTION AND REPORT ORGANIZATION

Deteriorations in the quality of source waters, increased water demand, more stringent drinking water quality regulations, etc. have resulted in much interest in membrane technologies. In spite of the large economic market and technical interests related to membranes, every installation is influenced by ***fouling***, which is the bane of this technology. Fouling refers to the increased total resistance to water flow across a membrane—including any materials accumulated near its surface.

The addition of a coagulant prior to microfiltration (MF) or ultrafiltration (UF) membranes has been suggested to generally improve product water quality and reduce membrane fouling by coagulating natural colloidal matter and dissolved organic matter ahead of the filter (Lahoussine-Turcaud et al. 1990b and Wiesner et al. 1992). Usually, chemical coagulation with iron or aluminum salt is used, however electrocoagulation (EC) is another possibility. To our knowledge, this research is the first systematic investigation of electrocoagulation pretreatment to reduce MF fouling.

Further, MF alone is not an efficient barrier for viruses, because viruses are typically smaller than its pores. Several published studies have reported partial (0.2- to 3-log) virus removal by MF (Jacangelo et al. 1995a, Madaeni et al. 1995, and Urase et al. 1996), which is significantly less than the 4-log virus removal mandated by the Surface Water Treatment Rule (SWTR). Therefore, MF by itself cannot meet the SWTR virus removal requirement even though the turbidity and protozoa regulations can be met easily.

The first aspect of our research is membrane fouling wherein we compared an innovative EC pretreatment process with conventional chemical coagulation to reduce the fouling of MF membranes. Additionally, fouling of nanofiltration (NF) and reverse osmosis (RO) membranes was expected to be decreased while operating on water subjected to electrocoagulation-microfiltration (EC-MF) and chemical coagulation-microfiltration (CC-MF) pretreatment compared with simple microfiltration pretreatment. The second aspect of our proposed research considers improvements in microfiltered water quality in terms of NOM, colloids, and viruses using EC pretreatment compared to chemical coagulation pretreatment or MF alone.

In a paper from our laboratories that was published in *Water Research*, we demonstrated increased efficacy of virus removal from synthetic waters in the absence of particles and NOM using iron EC pretreatment (Zhu et al. 2005a). As shown in this paper, virus removal by MF alone was insignificant in the absence of coagulation. Adding iron (either EC or chemical coagulation), improved virus removal dramatically, demonstrating the superiority of both types of pretreatment to MF alone. Furthermore, EC out-performed conventional chemical coagulation over the entire range of iron dosages investigated. At > 10 mg/L iron dosages from EC, virus removal by the system was more than the SWTR requirement of 4-logs (99.99%) (Zhu et al. 2005a).

2.1. Project objectives

The explicit purpose of this research was to verify whether our promising results obtained earlier using synthetic waters would be valid in the presence of turbidity, NOM, and other contaminants typical of natural surface water so that iron EC can be adapted for municipal water treatment. The specific objectives of the proposed research investigating chemical coagulation and electrocoagulation pretreatment using iron for MF of surface water were to:

1. Quantify reductions in microfilter fouling as a result of electro- and chemical-coagulation pretreatment,
2. Investigate differences in virus and NOM removal due to EC pretreatment compared with conventional chemical coagulation using iron, and
3. Compare the EC process with CC-MF and MF alone as pretreatment for NF and RO membranes.

Experiments were performed on actual surface water (Lake Houston) that serves as one of the source waters for the City of Houston using iron as the coagulant species to answer the following questions:

- To what extent do chemical coagulation and electrocoagulation pretreatment reduce MF membrane fouling while simultaneously improving filtered water quality with respect to viruses and NOM?
- Can incorporating coagulation ahead of MF further reduce NF and RO membrane fouling rates in an integrated membrane system?
- What are the underlying mechanisms by which electrocoagulation and conventional chemical coagulation improve MF performance?

2.2. Report organization

The principal components of this report are two Masters' theses written by the respective research assistants. Chapter 4 reports on reductions in microfilter fouling and improvements in removal of natural organic matter with chemical- and electro-coagulation pretreatment. This aspect of research was completed by Ms. Ashima Bagga and was recently published in volume 309 issues 1-2, pages 82–93 of the *Journal of Membrane Science* under the title “Evaluation of Iron

Chemical Coagulation and Electrocoagulation Pretreatment for Surface Water Microfiltration". Chapter 5 documents enhancements in virus removal achieved by coagulation pretreatment and the effects of NOM on electrocoagulation performance. This constitutes research performed by Ms. Archana Venkataramanan. We are currently writing a manuscript for submission to a peer-reviewed journal such as *Water Research*.

Chapter 3 consists of an overall summary and conclusions drawn from this work as well as a few recommendations for future research in this subject area. Because the main body of the report is directly based on the thesis written on respective topics, the format of these chapters is essentially same as in the thesis. Chapters 4 and 5 begin with a brief introduction of the work, followed by experimental materials and methods, results and discussions, and finally the inferences from each set of experiments.

2.3. Consistency of the Lake Houston water sample

Lake Houston water was first sampled on January 12, 2006. A pump station feeds Lake Houston water to the City of Houston's East Water Purification Plant through a 14-mile open channel. Sampling was performed at the pump discharge into this channel. The cooperation and assistance of Mr. Joey Eickhoff and Mr. Naseem Jilani in sampling ~ 400 liters (L) of water is highly appreciated. Figure 2-1 shows University of Houston students and City of Houston staff who performed sampling. This water sample was stored at 4 degrees Celsius (°C) in a walk-in temperature controlled room and monitored regularly in terms of its water quality parameters to ensure integrity of our storage procedure. However, regular monitoring revealed a small drop in organic water quality parameters including dissolved organic carbon (DOC) and ultraviolet absorbance at 254 nm (UV_{254}). For example, DOC had decreased from ~ 5.3 to 4.6 mg/L and UV_{254} had decreased from 0.133 to 0.122 reciprocal centimeters (cm^{-1}). Additionally, we ran experiments using the original sample for approximately six-months. This was replaced by a new sample on July 7, 2006, obtained from the Lake Houston Canal at the City of Houston's East Water Purification Plant. Note that our second water sample was also stored at 4 °C and can be considered representative of Lake Houston water in the summer season, whereas the original one was obtained in the winter season.

Figure 2-1.—University of Houston students along with City of Houston employees near the Lake Houston sampling location. Ms. Ashima Bagga, Ms. Archana Venkataramanan, Dr. Yongki Shim, Mr. Joey Eickhoff, Mr. Naseem Jilani, and Dr. Ramesh Sharma (from left to right).

Figure 2-2 depicts several inorganic, physical, and organic parameters that were monitored approximately once a week. Data in Figure 2-2 indicate that the second water sample was lower in turbidity, hardness, conductivity, and buffering capacity (alkalinity), but higher in organic content compared with the original sample. Further, the water quality had been relatively consistent and has not deteriorated, at least in terms of the monitored parameters. This important observation allows the comparison of our results from experiments performed using these two water samples over the duration of the study.

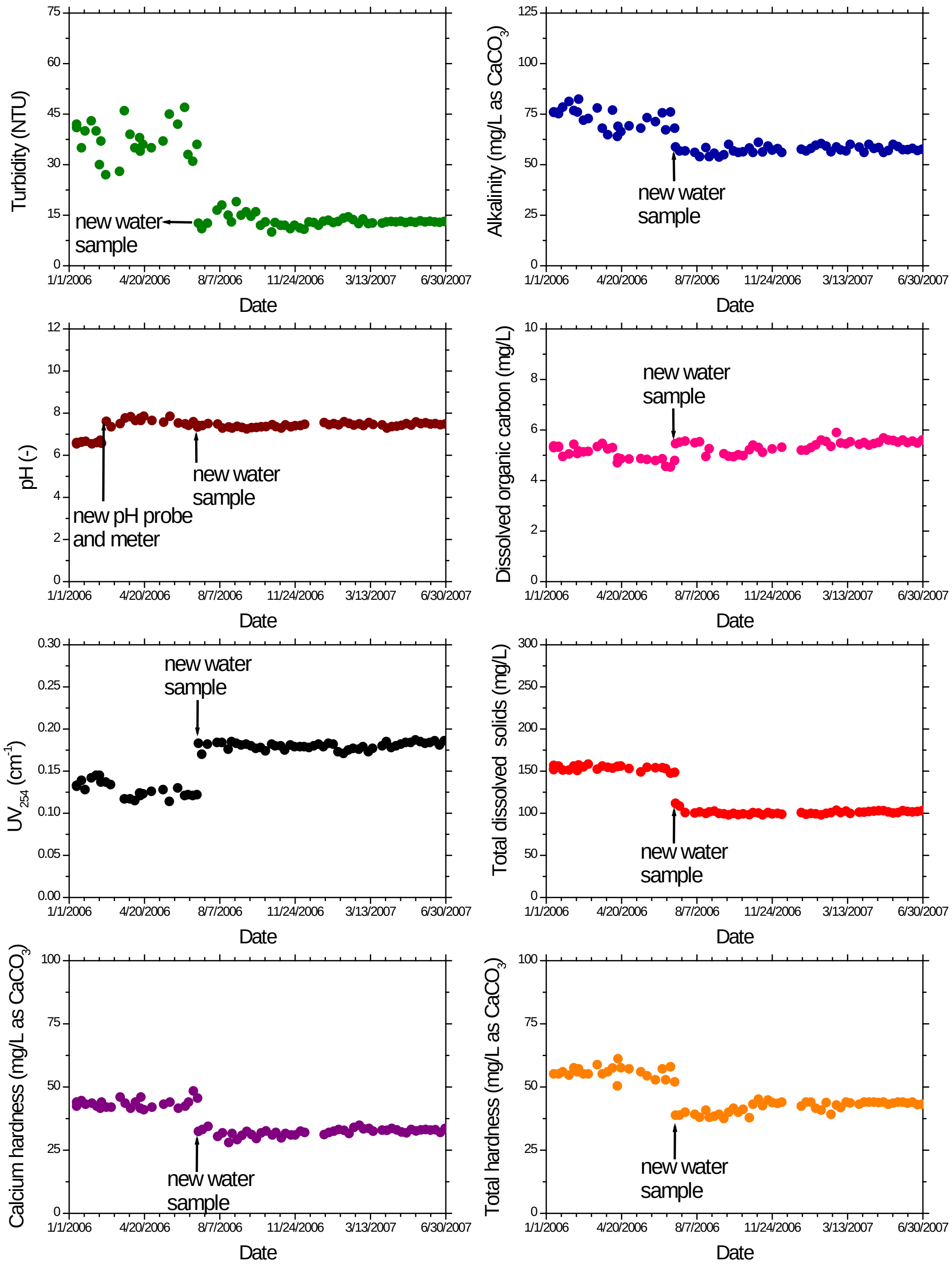


Figure 2-2.—Summary of Lake Houston water quality parameters for the two samples employed in this research.

3. SUMMARY AND CONCLUSIONS

3.1. Membrane fouling and natural organic matter removal

At the initiation of dead-end microfiltration of surface water, fouling is caused by the blocking of individual membrane pores by natural and flocculated colloids, with flux reduction quantitatively obeying the intermediate blocking law. Continued particle deposition results in blocking all available pores—resulting in the formation of a cake layer which increases frictional losses for water permeation. Hence, cake filtration theory may not be rigorously applicable to model short-term flux decline between backwashes especially for micro- and ultrafiltration of raw waters as has been assumed previously (e.g., Chellam et al. 1998, Kim and DiGiano 2006a, and Kim and DiGiano 2006b), but these assumptions hold true for MF and UF of coagulated waters. Difficulties in *a priori* predicting the effects of Fe(III) dose on fouling continue to necessitate empirical site-specific tests prior to implementing conventional chemical coagulation pretreatment for surface water MF.

3.2. Virus removal

Chemical coagulation using Fe(III) as a coagulant was found be very effective for virus removal in both test waters studied, namely lake water with NOM and synthetic water without NOM. For both test waters, in the absence of coagulation, MF alone achieved < 1-log virus removal. However, pretreatment with Fe(III) coagulation dramatically improved the membrane performance and virus removal. As coagulant dose increased, MS2 removal also increased, and > 5-log removal was achieved for both the test waters at pH 6.4 for a 5 mg/L Fe(III) dose. MS2 removal improved as pH decreased, which was presumably due to the increase in the concentration of positively charged iron floc particles at lower pHs causing better virus removal.

The mechanism of MS2 removal during CC-MF in both the test waters was attributed to adsorption of negatively charged virus particles to the positively charged iron hydroxide floc particles that were subsequently removed by microfiltration.

EC was found to generate Fe(II) iron in the pH 6.4-7.5, and the oxidation of Fe(II) to Fe(III) was strongly pH dependent. Deliberate oxidation/rusting of iron rods in the EC did not improve Fe(II) oxidation at pH 6.4 or 7.5. EC-MF was found to be an effective process for virus removal from synthetic water without NOM. Similar to CC-MF, in the absence of coagulant, MF achieved < 1-log virus removal which increased as iron dosages increased and > 5-log removal was achieved at pH 6.4 for 12.8 mg/L total iron generated. As observed with CC, virus removal increased as the pH decreased from 7.5 to 6.4. In-spite of Fe(II) being generated in EC at pH 6.4 and pH 7.5, effective virus removal was achieved in synthetic water due to the oxidation of Fe(II) to Fe(III) during several hours of continuous mixing and filtration. Hence, EC was not consistently effective for synthetic water due to the variable oxidation of Fe(II) that depends on pH and duration of filtration.

EC was not an effective pretreatment for MF when treating water containing NOM, such as lake water. Less than 2-log removal was observed in pH 6.4-8.3, even with iron concentrations as high as 13 mg/L. The poor performance of EC in lake water was attributed to:

(a) Generation of Fe(II) in EC that must be oxidized to Fe(III) to cause effective virus removal, and

(b) Presence of natural organic matter (NOM) in lake water, which complexed the Fe(II) and prevented its oxidation.

For a nominal iron concentration of 13 mg/L, EC-MF at pH 6.4 gave > 5-log removal in NOM free synthetic water while < 2-log removal was observed in lake water containing NOM. Similar results were observed at pH 7.5. As explained previously, although Fe(II) was generated in EC, high virus log removals were observed in synthetic water due to long hours of standing and filtration which caused effective Fe(II) oxidation. However, similar virus log removals were not observed in lake water in spite of providing several hours of standing and mixing for Fe(II) oxidation.

When comparing CC-MF and EC-MF, both processes were equally effective in removing viruses from synthetic water without NOM at both pH 6.4 and 7.5. In lake water with NOM, CC-MF consistently outperformed EC-MF at all pHs studied. CC-MF achieved > 5-log removal at pH 6.4 and > 4-log removal at pH 7.5 while < 2-log removal was observed at both the pHs by EC-MF. Further, Fe(II) coagulation-syringe MF experiments at pH 7.5 was found to achieve effective virus removal (up to 3.5 log) for a Fe(II) dose of 10 mg/L in NOM-free synthetic water when sufficient time was provided for Fe(II) oxidation. However, a similar performance was not observed in lake water, where < 0.5-log virus removal was observed for all the Fe(II) doses considered. The ineffectiveness of EC and Fe(II) CC for virus removal from lake water was concluded to be due to the rapid binding of NOM with Fe(II) and prevention of Fe(II) oxidation, hydrolysis, and subsequent sorption of viruses.

Fe(II) coagulation experiments in synthetic water spiked with varying concentrations of NOM (Suwannee River NOM) showed a detrimental effect on the removal of viruses. As the DOC concentration increased, the MS2 virus log removal decreased significantly from 3.5 to 0.01. Also, the ratio of DOC/Fe in the feed had a significant effect in the iron and NOM (i.e. DOC) present in the filtrate samples. No iron was present in the microfiltered effluent when the DOC/Fe ratios were low (≤ 1.7), whereas significant Fe(II) passed through the microfilter when the DOC/Fe ratios were relatively high (≥ 3.1). This suggests that the formation of NOM and Fe(II) complex depended on the initial concentrations of NOM and Fe(II) in the water sample. Hence, we concluded that in water containing significant concentrations of DOC, the NOM binds with Fe(II) and prevents its oxidation to Fe(III) and subsequent hydrolysis to $Fe(OH)_3$ (s) that can adsorb the viruses and remove them effectively in MF.

3.3. Future work and Recommendations

In this research, it was found that the presence of NOM in natural water had a detrimental effect in virus removal using EC due to its hindering effect on Fe(II) oxidation to Fe(III) and subsequent hydrolysis and adsorption of viruses. It was observed that the DOC/Fe ratio in the feed had a significant effect in the presence of iron and DOC in the filtrate and high DOC concentrations reduced the removal of viruses.

Future studies can focus on the studying the mechanisms involved in the DOC-Fe(II) interactions and their effect on virus removal. This will help to better understand the basic mechanisms involved in these reactions that will help to develop a method to overcome the hindering effect of NOM and Fe(II) complexation and improve virus removal in these experiments.

Iron electrocoagulation is not recommended for pretreating surface water prior to MF, since iron electrocoagulation generated soluble Fe(II) rather than insoluble Fe(III). A transition metal, such as iron, exhibits multiple common oxidation states. In contrast, aluminum, the other common metal-ion coagulant, is predominantly present as trivalent Al(III) in aqueous environments, since it belongs to main group 13 in the periodic table. Hence, it is hypothesized that employing aluminum anodes will generate Al(III) during electrocoagulation producing $Al(OH)_3$ precipitates capable of destabilizing natural colloids and potentially improving membrane performance when used as a pretreatment process for MFMembrane Fouling and Natural Organic Matter Removal

3.4. Research background

Deteriorations in the quality of source waters and increased demand, coupled with more stringent water quality regulations have resulted in increasing implementation of pressure-driven membrane technologies to purify drinking water, wastewater, and for water reuse applications (Mallevialle et al. 1996). Microfiltration (MF) and ultrafiltration (UF) are highly effective for turbidity, bacteria, and protozoa removal (Jacangelo et al. 1995a), also indirectly assisting in disinfection by-product (DBP) control by lowering chemical disinfection requirements for the filtered water. However, wider application of these technologies continues to be impeded by fouling.

A common method to reduce fouling and simultaneously improve filtered water quality is to chemically coagulate the feed water using aluminum or iron salts prior to MF/UF (Choi and Dempsey 2004, Howe and Clark 2006, Howe et al. 2006, Lahoussine-Turcaud et al. 1990a, and Wiesner et al. 1989). One example of a full-scale facility is the 38 milliliters per day (mL/d) Western Canyon Regional Water Treatment Plant operated by the Guadalupe Blanco River Authority near San Antonio, Texas where ferric sulfate is used as the coagulant followed by ~10 minutes of flocculation and direct MF. Coagulant dose is adjusted to meet the filtered water quality disinfection by-products (DBP) regulations and membranes are backwashed every 20 minutes.

Even though several large-scale water treatment plants already employ coagulation pretreatment, the initial stages of MF fouling mechanisms are still not fully understood. All microfilters employed for municipal applications are inherently operated in an unsteady mode to obtain higher fluxes by frequent backwashing. Hence, it is necessary to quantify both the transient and pseudo steady-state fluxes. To date, only a few studies have elucidated the initial stages of fouling wherein fluxes are transient and pore blocking is responsible for flux decline e.g. (Bowen et al. 1995, Taniguchi et al. 2003, Tracey and Davis 1994, Xu and Chellam 2005, and Yuan et al. 2002). Importantly, all these studies have employed synthetic waters incorporating model colloids or purified macromolecules (e.g., pure cultures of bacteria, humic acids, natural organic matter (NOM), and proteins). Therefore, their findings are not readily extrapolated to municipal water treatment applications. Unsteady fluxes during the initial stage of microfilter fouling caused by natural colloidal matter present in raw waters have been considered only very recently (Kim et al. 2007).

Electrocoagulation pretreatment is an alternative to conventional chemical coagulation using Fe or Al salts prior to MF. In electrocoagulation, the coagulant (Fe or Al) is generated by electrolytic oxidation of an anode. The advantages of electrocoagulation over conventional chemical coagulation include:

(1) There is no alkalinity consumption,

(2) No change in bulk pH,

(3) The direct handling of corrosive chemicals is nearly eliminated, and

(4) Can be easily adapted for use in portable water treatment units especially during emergencies.

Even though it has been reported to be efficient for contaminant removal (Caňizares et al. 2006 and Hu et al. 2003), very limited information is available regarding electrocoagulation as a MF pretreatment process to reduce fouling e.g. (Al-Malack et al. 2004). Because these studies have focused on crossflow filtration and synthetic waters, they are not directly applicable to dead-end MF of surface waters encountered in municipal applications. Further, the effects of electrocoagulation and MF operating conditions on system performance have not yet been delineated.

3.5. Goals and objectives

The overall goals of this research were to evaluate iron electrocoagulation and conventional iron chemical coagulation to treat natural surface water prior to microfiltration. Specific objectives were to:

1. Delineate the dominant fouling mechanisms during early and later stages of microfiltration of raw and coagulated waters,
2. Quantify the effects of coagulation and filtration conditions on microfilter fouling,
3. Compare the effects of electrocoagulation and chemical coagulation pretreatment to improve membrane performance (reduction in membrane fouling and enhancements in natural organic matter removal), and
4. Identify the underlying mechanisms responsible for differences in the performance of chemical coagulation and electrocoagulation pretreatment.

To achieve these objectives, chemical- and electrocoagulation experiments were conducted at different coagulant dosages and pH using Lake Houston water as a representative surface water. NOM concentrations and transient flux decline data for constant pressure, dead-end microfiltration of raw and coagulated suspensions were recorded and analyzed. Particle size distribution measurements along with visual inspection of surface deposits using scanning electron microscopy enabled better interpretation of experimental fouling data.

3.6. Experimental Work

3.6.1. Source water

All experiments were performed using a water sample from the Lake Houston Canal at the City of Houston's East Water Purification Plant obtained on July 7, 2006. This sample was stored at 4°C in a walk-in temperature controlled room and brought to room temperature (23 ± 1°C) prior to experimentation. Regular monitoring of physicochemical water quality parameters demonstrated limited change in the integrity of this water sample during the entire duration of laboratory work. The Lake Houston water sample could be characterized as being near neutral (pH 7.3-7.5), moderately turbid (11-14 Nephelometric Turbidity Unit [NTU]), soft (total hardness 39-44 mg/L as $CaCO_3$ and calcium hardness 29-

34 mg/L as $CaCO_3$), with low alkalinity (54-60 mg/L as $CaCO_3$), low total dissolved solids (TDS) (98-103 mg/L), and having moderate NOM concentrations (DOC 5.0-5.6 mg/L and ultraviolet absorbance at 254 nm at 1 cm path length UV_{254} 0.175-0.185 cm^{-1}).

3.6.2. Chemical coagulation

Conventional chemical coagulation was performed using a programmable jar tester (Model PB-900, Phipps & Bird Inc., Richmond, Virginia). Different volumes of $FeCl_3$ stock solution (1,000 milligrams [mg] Fe(III)/L) were added to six separate Gator jars during 1 minute of rapid mixing at 300 rpm (mean gradient velocity = 495 s^{-1}) to achieve Fe(III) concentrations between 0 to 15 mg/L. Flocculation was performed at a mean velocity gradient = 32 s^{-1} (40 revolutions per minute [rpm]) for 30 minutes. Importantly, a separate sedimentation step was not employed to simulate the most common application of coagulation pretreatment for MF during municipal water/wastewater treatment. Coagulation was evaluated at pH values of 6.4, 7.5, and 8.3 with a precision of ±0.2 pH units. Because of the low buffering capacity of Lake Houston water, 0.5 N NaOH was added to maintain the pH at its target value during coagulation/flocculation.

3.6.3. Electrocoagulation

Water samples were coagulated electrochemically using a custom built unit consisting of 940 milliliters (mL) volume fitted with a single 25 centimeter (cm) long rod-shaped iron anode (industrial grade mild steel 1018 CF Bar ASTM A108, 98.52% iron) with 38 cm^2 of active surface area and a porous cylindrical stainless steel cathode (stainless steel-316). The desired iron concentration (1-20 mg/L) was achieved by operating the unit in a batch mode under constant current (I = 0.8 A) and variable generation time (t) mode in accordance with Faraday's law as shown in Equation 1:

$$m_{Fe} = \frac{55.86 \times I \times t}{Z \times F} \qquad \text{(Equation 1)}$$

Where:

- m_{Fe} is the mass of Fe generated (g)
- I is constant current
- t is variable generation time
- Z is the number of electrons transferred per Fe atom (2 for ferrous ions and 3 for ferric ions)
- F is Faraday's constant (96,486 C eq^{-1}).

The cell contents were vigorously mixed using a magnetic stirrer for 2 minutes to simulate rapid mixing employed in chemical coagulation and to increase the rate of mass transfer of the electrochemically generated iron away from the electrode surface into the bulk solution. Importantly, unlike chemical coagulation, pH adjustment was not required because electrocoagulation did not consume alkalinity and alter the bulk pH. To maintain the iron generation efficiency, the anode was mechanically scrubbed using a waterproof silicon paper sanding sheet (100 Grit, C-Weight, McMaster-Carr Supply Co., Atlanta, Georgia) before each experiment to remove any precipitated ferric hydroxide and maintain a similar anodic surface for all experiments. Occasionally, the entire electrocoagulation unit was rinsed with dilute HNO_3 to dissolve any deposited ferric hydroxide.

3.6.4. Iron oxidation state measurements

Chemical speciation tests were conducted to determine the relative concentrations of ferrous and ferric ions generated electrochemically over the entire range of total iron dosages (0 to 20 mg Fe/L) at the three pH values investigated (6.4, 7.5, and 8.3). First, the total iron concentration was measured using flame atomic absorption spectroscopy (Analyst 300, Perkin-Elmer Corporation, Connecticut) using Standard Method 3111. Prior to analysis, samples were acidified to $pH < 2$ with 2% HNO_3 to solubilize particulate iron.

Second, the ferrous ion concentration was measured using the 1, 10-Phenanthroline method. The electrocoagulated samples were acidified to 0.1% HCl and treated with 0.2 M $CH_3COONa \cdot 3H_2O$ to adjust the pH between 3.5 and 4.5. The addition of 4 mL of 13.9 mM 1, 10-Phenanthroline (Sigma Aldrich, St. Louis, MO) resulted in the formation of a reddish orange complex $[(C_{12}H_8N_2)_3Fe]^{2+}$ with ferrous ions. The concentration of ferrous ions generated was estimated by measuring the absorbance of this complex at 510 nanometers (nm) wavelength using a glass cell with one-inch (2.54 cm) path length and a spectrophotometer (DR/4000 Spectrophotometer, Hach Company, Loveland, Colorado). Finally, ferric iron was calculated as the difference between total iron and ferrous iron.

3.6.5. Microfiltration procedure

Because coagulation pretreatment for MF would be more cost competitive without a separate sedimentation step, all experiments were performed in the direct MF mode (no settling) as noted in §4.3.2. Unstirred, dead-end, constant pressure MF experiments were conducted using a cell with 4.1 cm^2 of effective area (Model 8010, Millipore Corporation, Bedford, Massachusetts). Modified PVDF membranes rated at 0.22 micrometer (μm) (Durapore, GVWP02500, Millipore Corporation, Bedford, Massachusetts) were employed to approximate three commercial chlorine-tolerant micro/ultrafilters popular for large-scale municipal drinking water applications (MEMCOR® CS, Pall Microzoa, and

ZENON ZeeWeed). The Darcy resistance of all the membranes employed in this study was $5.96 \pm 0.53 \times 10^{10}$ m^{-1}.

The cumulative permeate volume was monitored by collecting the filtered water on a weighing balance (Ohaus Navigator N1H110, Fisher Scientific, Houston Texas) connected to a personal computer. The pressure was monitored using an analog pressure transducer (PX303-050G5V, Omega Engineering Company, Stamford, Connecticut). Data were acquired at a rate of 2 hertz (Hz) using a program written in LabVIEW (version 5.1, National Instruments, Austin, Texas). Prior to filtering raw and coagulated water samples, 100 mL of ultrapure water was filtered to evaluate clean membrane resistance and to rinse it of any preservative chemicals. The time-series of cumulative volume filtered per unit membrane area was exponentially smoothed using a damping factor of 0.3 prior to numerical differentiation to obtain the instantaneous water flux. Filtration was terminated after filtering ~150 mL water. After this, the membrane was carefully removed from the cell and dried to a constant weight at 60°C to measure cake mass. Separate experiments were performed in the range 2-30 pounds per square inch (psi) to determine cake compressibility.

3.6.6. Particle-size distributions

Particle size distributions of the raw and coagulated water samples were determined using an electrical sensing zone device (Multisizer III, Beckman Coulter Inc., Miami, Florida). A 100 μm aperture was used to measure floc sizes in the range of 2 to 60 μm. Both the raw and coagulated water samples were diluted 100 fold with prefiltered 2% NaCl solution to avoid aperture clogging and reduce potential flocculation. Also, the suspension was gently stirred during the analysis period to avoid settling. An average of triplicate analysis of each sample is reported. Each run was completed in ~ 30 seconds (s), wherein 1 mL of the suspension was drawn through the aperture and particle size distributions were obtained by passing a current of 3200 μA with a gain of 1. The instrument performance was verified by using NIST-certified nominal 10 μm polystyrene latex beads provided by the manufacturer.

3.6.7. Scanning electron microscopy

Scanning electron micrographs (SEM) of membrane coupons before and after filtration were obtained to visualize the foulant layers. The membranes were dried overnight in an oven at 60°C and observed under a field emission scanning electron microscope (LEO 1525, Carl Zeiss, Thornwood, New York) after sputter coating with a thin (10 nm) layer of gold.

3.6.8. Natural organic matter

DOC concentrations in raw and coagulated waters were quantified in terms of non-purgeable organic carbon using Standard Method 5310B and UV_{254} using

Standard Method 5910. Samples were acidified to $pH < 2$ using 2N HCl, purged with ultra-zero air, and analyzed using a total organic carbon (TOC) analyzer (TOC 5050A, Shimadzu Scientific Instruments, Columbia, Maryland). An average of 4 runs with < 2% coefficient of variation is reported in this manuscript. The instrument was calibrated with standards prepared using 4.80 mM stock solution of potassium hydrogen phthalate (Kanto Chemical Co. Inc., Japan). UV_{254} was measured using a spectrophotometer (DR/4000 Spectrophotometer, Hach Company, Loveland, Colorado) fitted with a 1 cm path length quartz cell.

3.7. Results and Discussion

3.7.1. Quality control and quality assurance statements

As reported in § 4.3.1 and Figure 2-2, the Lake Houston water sample showed only small variations in several physical, inorganic, and organic water quality parameters. Additionally, ~15% of experiments were repeated over the course of this study at varying pH, and Fe(III) dosages which demonstrated no statistical differences ($\alpha=0.05$) in flux decline profiles for duplicate experiments conducted on different days under identical experimental conditions. These results demonstrate that:

(1) The composition of the water sample remained highly consistent and

(2) Our coagulation, flocculation, and microfiltration procedures and protocols were highly reproducible

Results obtained over the entire duration of this research can be quantitatively compared. Quantitative details and statistical analysis of experimental reproducibility and consistency of the raw water sample can be obtained elsewhere (Bagga 2007).

3.7.2. Overall effects of chemical coagulation and microfiltration parameters

3.7.2.1. *Observations for fouling reduction*

As shown in Figure 4-1, fouling was reduced only at iron dosages higher than 10 mg Fe(III)/L, whereas lower dosages of ≤ 5 mg Fe(III)/L resulted in very similar flux profiles compared with the raw water. Additionally, in the range of our experimental conditions, similar flux decline profiles were obtained at pH 6.4, 7.5, and 8.3 demonstrating that chemical coagulation pH had a negligible impact on fouling (Figure 4-2). Further, as previously reported for dead-end MF of synthetic and untreated natural waters, fouling increased with transmembrane pressure (Chellam and Jacangelo 1998, Chellam et al. 1998, Lee et al. 2003, Opong and Zydney 1991) (Figure 4-3). Similar results were generally obtained for all combinations of pH, iron dose, and transmembrane pressure. These experimental observations are investigated in more detail next.

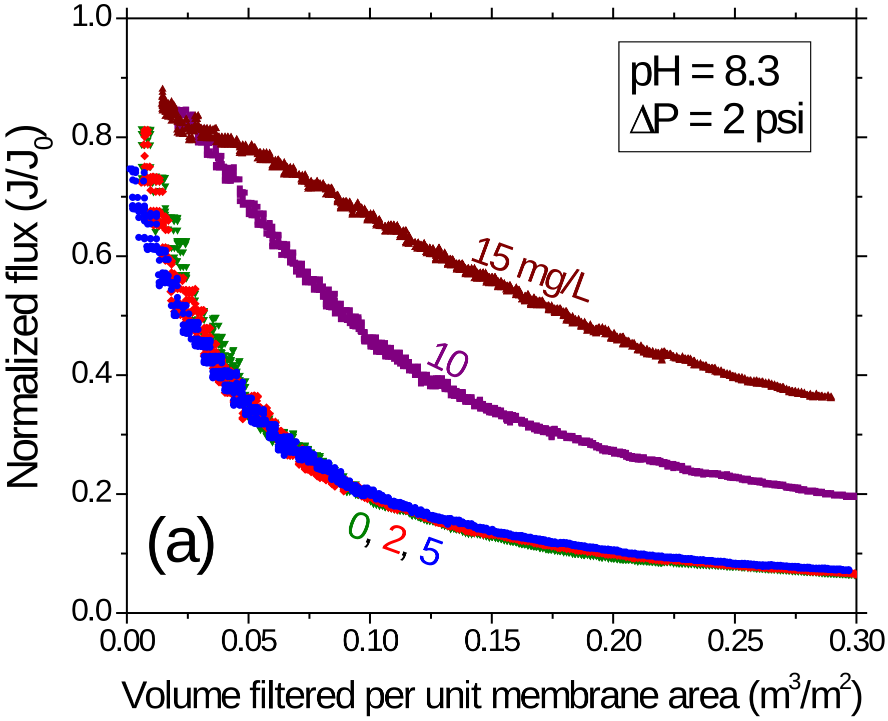


Figure 4-1.—Effects of iron coagulant dose on membrane fouling.

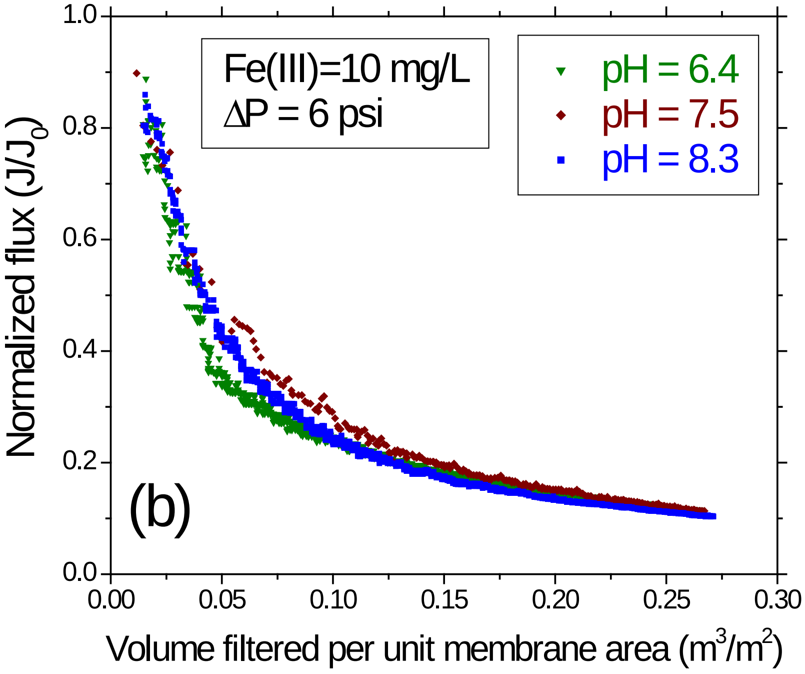


Figure 4-2.—Effects of coagulation pH on membrane fouling.

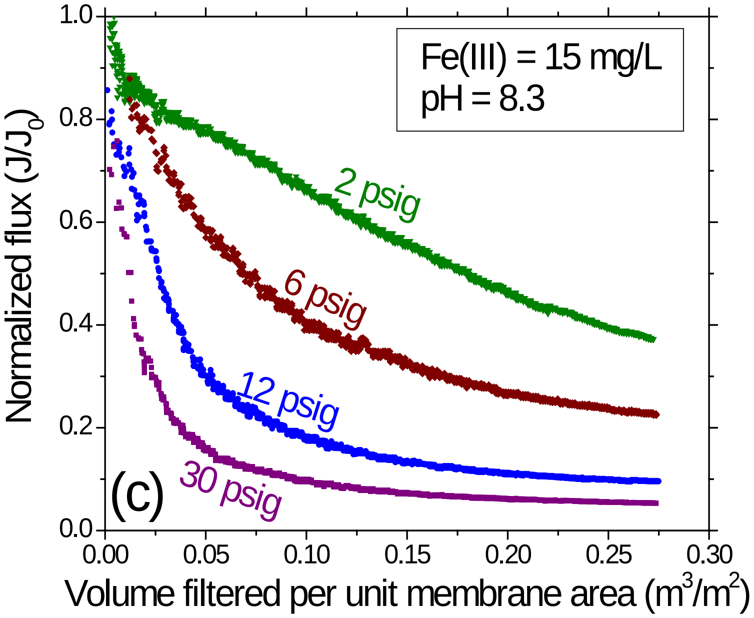


Figure 4-3.—Effects of transmembrane pressure on membrane fouling.

3.7.2.2. *Evaluation of dominant fouling mechanisms with chemical coagulation pretreatment*

Best-fits of blocking laws were obtained to evaluate fouling mechanisms as depicted in Figure 4-4. For raw water, a distinct intermediate blocking stage was observed initially wherein the inverse instantaneous flux profile exhibited a concave-upward behavior (Figure 4-4) obeying the expression (Bowen et al. 1995 and Hermia 1982) as shown below in Equation 2:

$$\frac{dt}{dV} = \frac{1}{Q_o} e^{k_i V} \qquad \text{(Equation 2)}$$

Where:

- Q_o is the initial flow rate,
- t is time,
- V is the cumulative volume of filtered water, and
- k_i is a parameter that describes the kinetics of flux decline during intermediate blocking.

Similar results were obtained for untreated raw water at all pH values and pressures investigated. The duration of initial intermediate blocking was significantly reduced by iron coagulant addition as seen in Figure 4-5. Also, the duration of intermediate blocking for raw water microfiltration decreased with pressure. Empirical deposition factors during intermediate blocking decreased with pressure for both raw water and coagulated suspensions suggesting differences in deposit morphology with initial permeate flux (Xu and Chellam 2005).

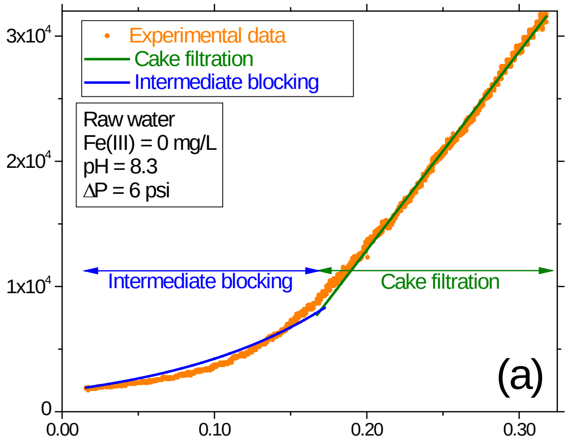


Figure 4-4.—Initial intermediate blocking followed by cake filtration for raw water microfiltration (no pretreatment).

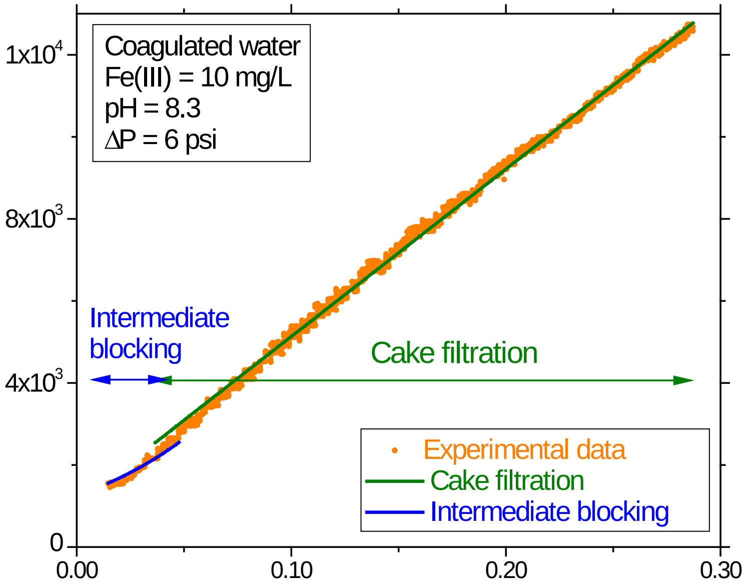


Figure 4-5.—Dominant cake filtration for almost the entire duration of microfiltration of coagulated water.

At the end of the intermediate blocking stage, a straight-line relationship between inverse instantaneous flux and cumulative permeate volume was established (Figure 4-6 and Figure 4-7) following filtration of sufficient water volume, as predicted for cake-dominated constant-pressure microfiltration (Bowen et al. 1995, Chellam and Xu 2006, Ho and Zydney 2000, Lee et al. 2003, Tiller 1990, Tracey and Davis 1994, Wiesner et al. 1989) as shown in Equation 3:

$$\frac{dt}{dV} = k_c V + \frac{1}{Q_o} \qquad \text{(Equation 3)}$$

Where the cake growth constant is shown in Equation 4:

$$k_c = \frac{\mu \alpha^* c_b}{\Delta P A_m^2} \cdot \qquad \text{(Equation 4)}$$

Where:

- A is area
- ΔP is change in pressure
- α^* is the specific cake resistance on a mass basis
- μ is the absolute viscosity
- c_b is the bulk particle concentration

In other words, once previously removed particles have completely obstructed all available membrane pores, microfilter fouling was quantitatively described by cake filtration. Under these conditions, flux decline is caused by the increasing resistance to water flow by a surficial colloidal deposit that grows in thickness as more water is filtered. Data presented in Figure 4-6 suggest that the higher feed water mass concentration for coagulated suspensions caused by $Fe(OH)_3$ precipitation greatly increased the kinetics of cake layer build-up—dramatically reducing the duration of intermediate blocking.

Specific cake resistances obtained by fitting Equation 3 to experimental data are summarized in Figure 4-6. As observed in Figure 4-6, specific resistances were highest for the raw water but declined with increasing iron dosage. However, comparable fouling profiles were obtained for Fe(III) dosages ≤ 5 mg/L (Figure 4-1) indicating that decreasing specific resistances were offset by increasing cake mass resulting in similar total hydraulic resistances for low iron dosages. In contrast, improved fluxes for Fe(III) dosages ≥ 10 mg/L demonstrate that both the specific and total cake resistances were decreased at high iron dosages. Increasing average particle sizes were measured at all pH conditions as more iron was dosed (inset of Figure 4-6) causing lower specific resistances. Larger particles are formed as natural colloids and organic matter are swept into flocs of freshly precipitated ferric hydroxide. Further, specific resistances and particle sizes at all doses in Figure 4-6 were independent of pH, underscoring the negligible impacts of coagulation pH on fouling shown earlier in Figure 4-4 at a single iron dose of 10 mg Fe(III)/L.

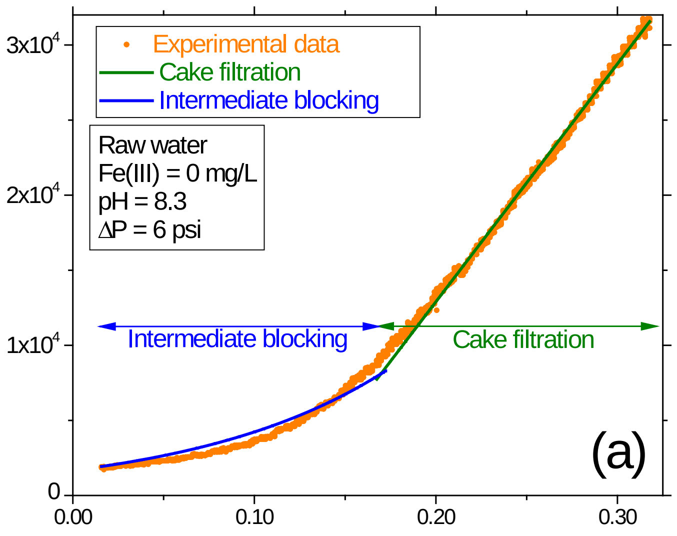


Figure 4-6.—Decreasing cake specific resistance with increasing iron dosage at a fixed pressure.

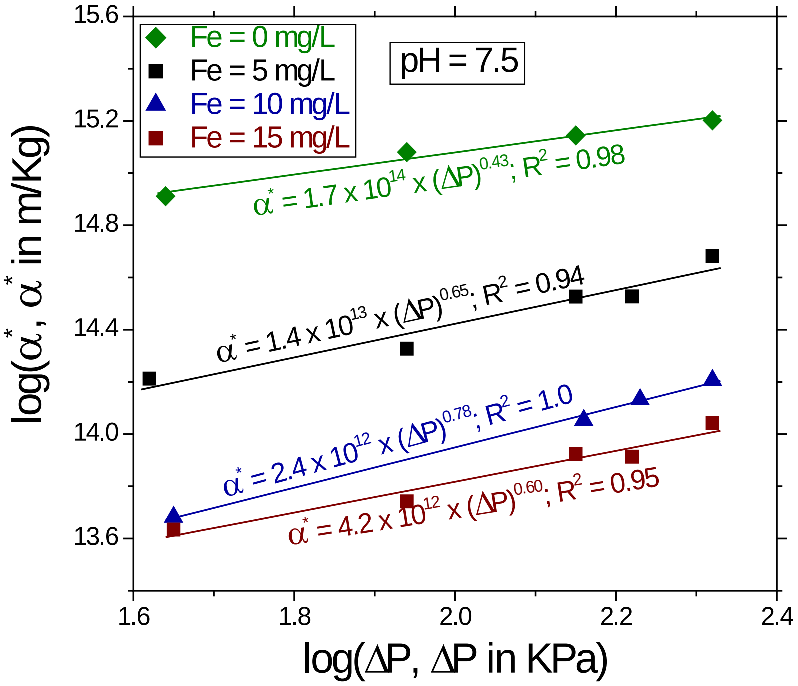


Figure 4-7.—Power-law compressibility of natural and iron-coagulated colloids.

Increasing stresses caused by higher transmembrane pressures generated more compact cakes having higher specific resistances. For example, Figure 4-7 depicts that at pH 7.5, natural and coagulated colloids compressed in a power-law manner with compressibility indices between 0.4 to 0.8, which is in the range reported earlier for other water and wastewater treatment studies (Chellam and Jacangelo 1998, Sørensen and Sorensen 1997, Tiller and Kwon 1998). Similar results were obtained at other pH values. Highly compressible cakes non-linearly increased hydraulic resistance to water permeation—causing discernibly greater fouling at higher pressures reported in Figure 4-2.

3.7.3. Overall effects of electrocoagulation and microfiltration parameters

As shown in Figure 4-8, increasing the dosage of electrochemically-generated iron only resulted in small reductions in membrane fouling. Unlike chemical coagulation, similar fluxes were obtained even at high iron dosages of 14 and 21 mg/L. Also in contrast to chemical coagulation, the transmembrane pressure did not influence membrane fouling during filtration of electrocoagulated suspensions (Figure 4-9). Interestingly, fouling was incrementally worse at pH 6.4 compared with pH 7.5 and 8.3 for a fixed iron dose (Figure 4-10). Hence, in the range of our experimental conditions, electrocoagulation pretreatment did not substantially improve fluxes compared with raw water. These observations are investigated in more detail next.

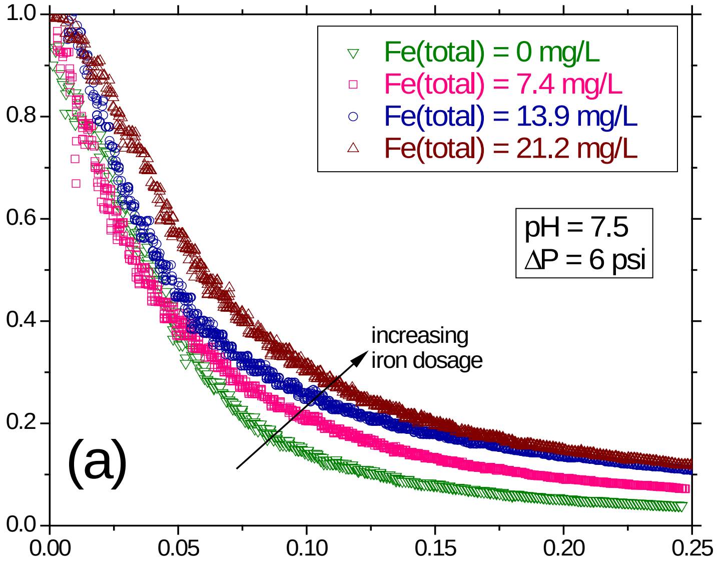


Figure 4-8.—Effects of electrochemical iron addition on microfilter fouling.

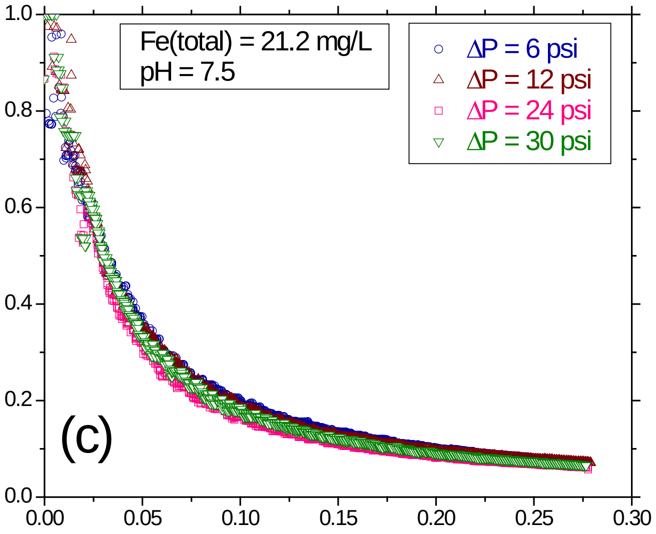


Figure 4-9.—Effects of pressure on microfilter fouling with electro-coagulation pretreatment.

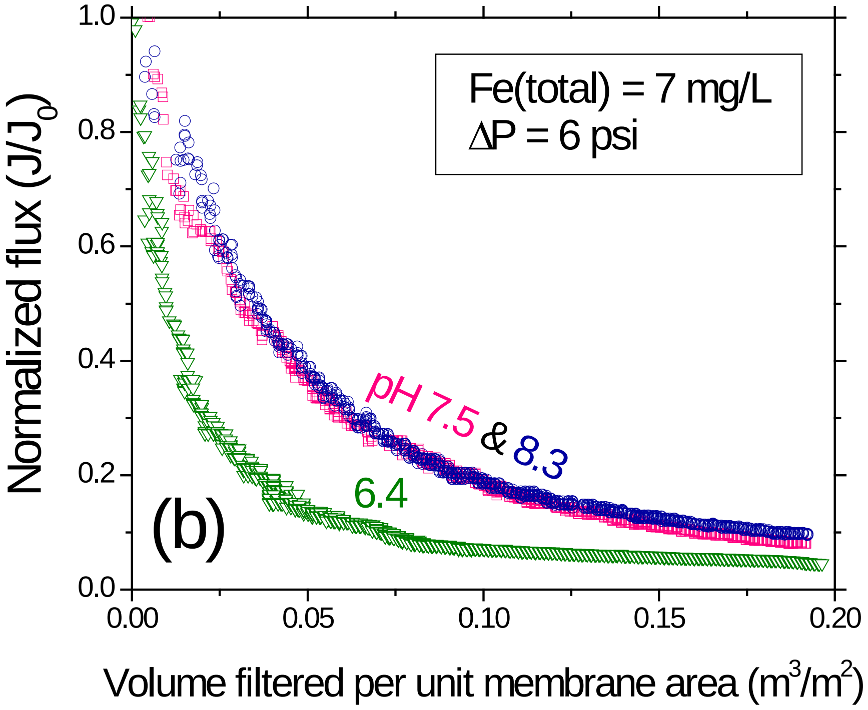


Figure 4-10.—Effects of electrochemical cell pH on microfilter fouling.

3.7.4. Iron speciation in electrocoagulated waters

Because electrocoagulation did not reduce MF fouling in the entire range of experimental conditions investigated, the emphasis of this aspect of our research was on electrochemical iron generation rather than MF fouling mechanisms. The electrocoagulation system performance was evaluated by varying generation times for a fixed total rapid mixing period of 2 minutes at a constant operating current of 0.8 A and comparing with Faraday’s law (Equation 1) for ferrous (Z=2) and ferric (Z=3) iron (Figure 4-11, Figure 4-12, and Figure 4-13). As observed from Figure 4-11, experimental data accurately agreed with a 2-electron transfer at all pH values. In other words, ferrous ions (rather than ferric ions) were produced at the anode during electrocoagulation of Lake Houston water. Note that it is highly disadvantageous to generate Fe(II) since it is highly soluble in nature and therefore, is not capable of colloid destabilization by sweep flocculation thereby causing poor electrocoagulation performance.

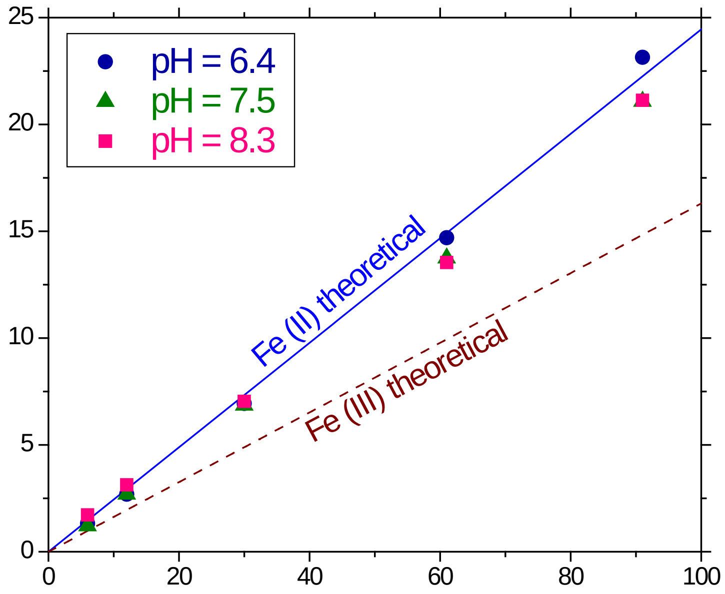


Figure 4-11. Larger particles are formed during electrocoagulation at pH 7.5 and 8.3 compared with 6.4 and with increasing total iron dose.

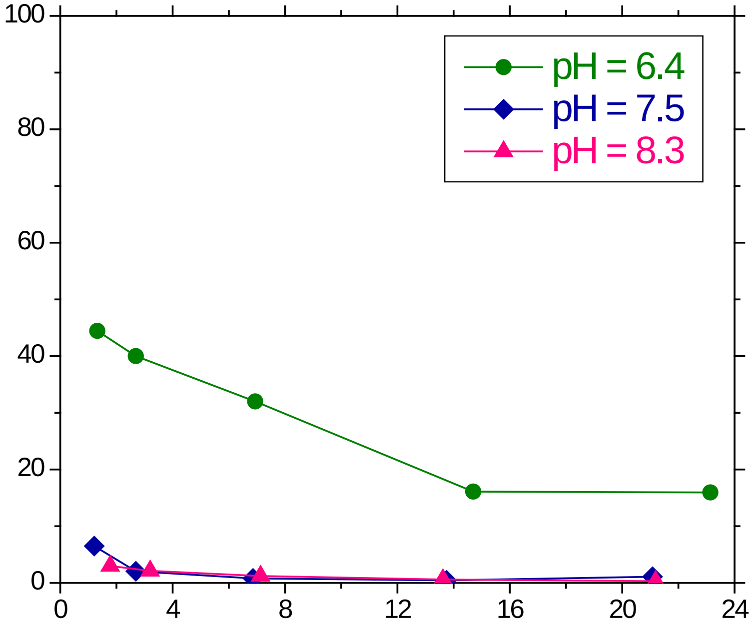
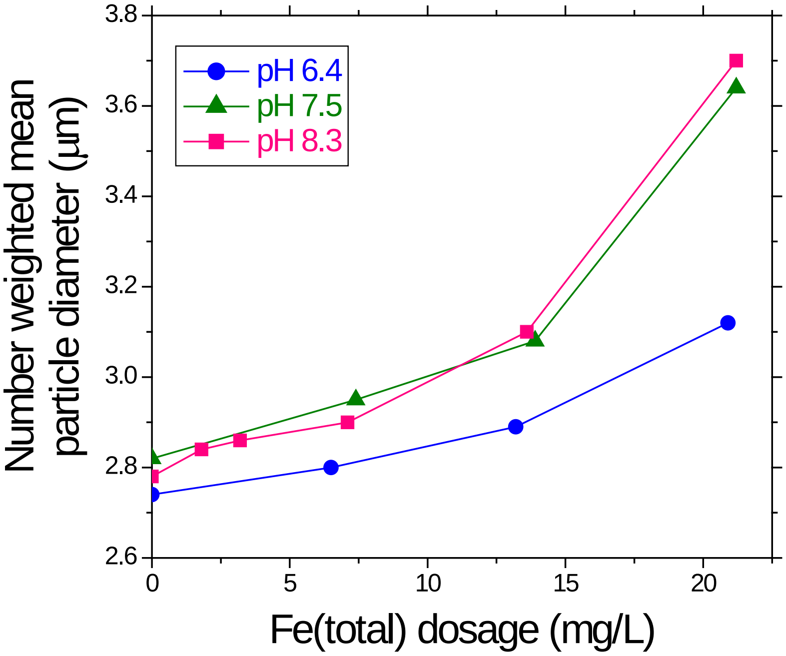


Figure 4-12.—Oxidation of electro-chemically generated Fe(II) as a function of pH values and total iron dose following 2-minutes rapid mixing.



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Our results agree with other recent results from our laboratories (Lakshamanan 2007) that also have documented electrochemical generation of ferrous ions at the anode rather than the anticipated ferric ions. Ferrous ions generated at the anode get oxidized during the rapid mixing period, whose rate at pH ≥ 5 is first order in Fe(II) concentration and oxygen partial pressure (p_{O2}) and inverse second order in solution pH (Stumm and Morgan 1996) as shown in Equation 5:

$$-\frac{d[Fe(II)]}{dt} = k[Fe(II)][OH^-]^2 p_{O_2} \qquad \text{(Equation 5)}$$

The rate of Fe(II) oxidation in natural waters is complicated by NOM chelation, which tends to inhibit the rate provided by Equation 5 (Liang et al. 1993 and Theis and Singer 1974). Nevertheless, as shown in Figure 4-12, substantial levels of reduced iron were present only at low pH and low dosages as predicted by Equation 5. Higher coagulant dosages have been shown to increase pH transiently during the electrolysis period (Lakshamanan 2007), and are considered responsible for faster oxidation of Fe(II) at higher iron concentrations. Therefore, Fe(II) concentrations decreased with total iron generated in Figure 4-12. One limitation of our iron speciation measurements is that Fe(III) was calculated as the difference between Fe(total) obtained by atomic absorption spectrometry and Fe(II) through colorimetry. Fe(II) complexed with NOM may potentially not be measured by the phenanthroline method and will therefore count towards Fe(III). Hence, we recommend direct measurement of Fe(III) in future studies. Also, given the second order increase in the rate of Fe(II) oxidation with [OH^-], only Fe(III) was essentially present in the system after two minutes of mixing at higher pH values. Therefore, based on this and other recent research in our laboratory (Lakshamanan 2007), the overall electrochemical reactions taking place inside the cell can be written as:

Anode: $Fe(0)(s) = Fe(II) + 2e^-$

Oxidation: $4Fe(II) + O_2 + 4H^+ = 4Fe(III) + 2H_2O$

Hydrolysis: $Fe(III) + 3H_2O = Fe(OH)_3 + 3H^+$

Cathode: $2H_2O + 2e^- = 2OH^- + H_2$

Overall: 4Fe(0)(s) + 10H2O + O2 = 4Fe(OH)3 (s) + 4H2

In other words, during iron electrocoagulation of natural water, in situ oxidation of Fe(II) results in destabilization and aggregation, whereas in chemical coagulation, it is due to hydrolysis of added Fe(III) and hydrolysis products that form Fe(OH)3 precipitates. The significant presence of soluble Fe(II) at pH 6.4 resulted in smaller flocs (Figure 4-13) that formed more compact cakes with higher hydraulic resistance. Another likely contributor to fouling is the oxidation of Fe(II) to Fe(III) in the membrane pores. These two phenomena together explain the sharper fouling at 6.4 pH in Figure 4-11 compared with pH 7.5 and 8.3. At higher pH values, electrolytically generated Fe(II) was nearly completely oxidized (as predicted by Equation 5) resulting in larger flocs and consequently more permeable cakes. Very similar flux decline profiles observed in Figure 4-12 at pH 7.5 and 8.3 are consistent with flocs of similar sizes at these pH values. Poor microfilter performance with EC pretreatment indicates that the *in situ* oxidation of Fe(II) to Fe(III) and subsequent hydrolysis is a less efficient pretreatment process compared with hydrolysis of Fe(III) added directly in chemical coagulation. Fe(II) oxidation in solution has been shown to produce a colloidal and less polymeric precipitate (Stumm and Morgan 1996) that is consequently also more soluble than freshly precipitated $Fe(OH)_3$ partially explaining the poor performance of iron electrocoagulation.

More importantly, it has been found in a related study in our laboratory (Venkataramanan 2007) that the Fe(II) generated by electrocoagulation complexes quickly with NOM and prevents Fe(II) oxidation to Fe(III) and subsequent $Fe(OH)_3$ precipitation and floc formation. These results are in agreement with literature reports on the interference of NOM on the oxidation of ferrous iron (Liang et al. 1993 and Theis and Singer 1974). Thus, the presence of NOM in surface waters is highly detrimental to iron electrocoagulation because, even with increasing pH and long coagulation time, the immediately formed Fe(II)-NOM complex prevents $Fe(OH)_3$ precipitation and floc formation.

3.7.5. Comparison of microfilter performance with chemical- and electrocoagulation pretreatment

Under conditions that favored both chemical- and electrocoagulation (higher Fe(III) dosages, high pH, and lower pressures), chemical coagulation out-performed electrocoagulation for fouling control (Figure 4-14). However, under conditions unfavorable to chemical coagulation (high pressures, low iron dosages, and high pH), both types of pretreatment did not improve fluxes compared with raw water (Figure 4-15). These are consistent with the formation of larger flocs at high $FeCl_3$ dosages compared with electrochemically generated iron and the effects of cake compressibility. Greater productivity achieved at low pressures in this study is similar to earlier pilot-scale results of surface water microfiltration (Chellam and Jacangelo 1998). These results underscore that MF and UF systems commonly operated at constant flux during water and wastewater treatment disproportionately lose productivity when the pressure is increased to maintain the permeate flux due to power-law cake compression.

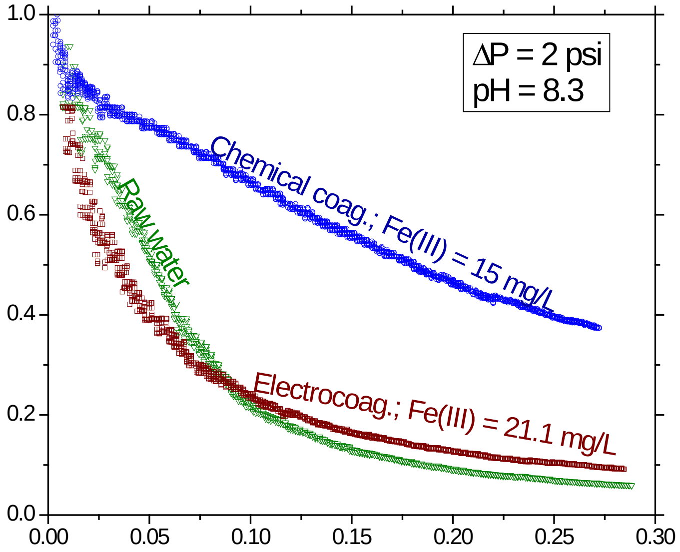


Figure 4-13.—Comparison of chemical- and electro-coagulation pretreatment under conditions favoring the former (low pressure and high iron dosage).

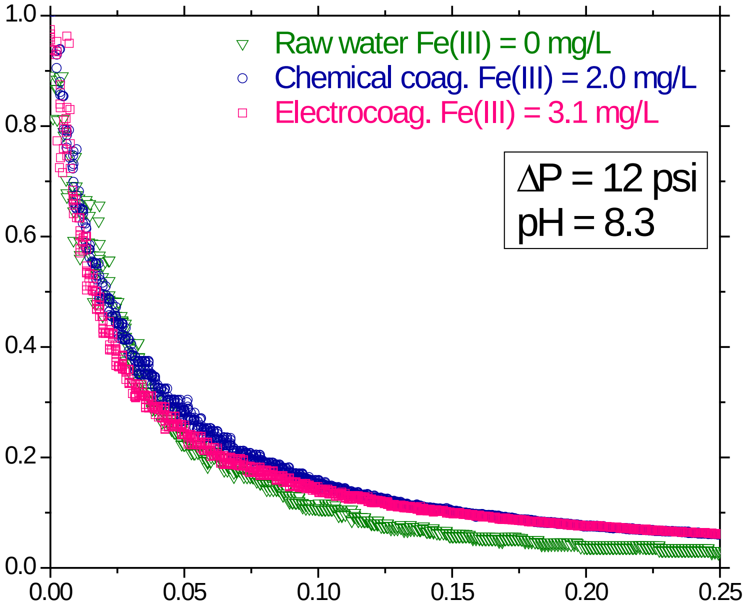


Figure 4-14.—Comparison of chemical- and electro-coagulation pretreatment under conditions not favoring the former (higher pressure and lower iron dosage).

Chemical- and electrocoagulation pretreatment was also compared in terms of improvements in permeate water quality. As shown in Figure 4-16a, b, d and e, NOM removal (measured as DOC and UV_{254}) increased monotonically with iron dosages for both chemical- and electrocoagulation. Additionally, greater removals were obtained at pH 6.4 than at 7.5 and 8.3 with both types of pretreatment. Higher iron dosages at lower pH values increase the concentration of positively charged Fe hydrolysis products that enhance charge neutralization of NOM resulting in the observed monotonic increase in DOC and UV_{254} removal with decreasing pH and increasing iron dosage. These observations are similar to the enhanced coagulation requirements of the U.S. Environmental Protection Agency (EPA) to increase NOM removal by lowering the pH while simultaneously increasing chemical coagulant dose. Further, as seen in Figure 4-16a, b, d and e, coagulation preferentially removed the aromatic components of NOM and compounds with unsaturated carbon bonds (which is measured as UV_{254}) over DOC. In other words, higher molecular weight and hydrophobic NOM components are preferentially removed during both chemical and electrocoagulation.

Inefficient electrochemical NOM removal at pH 6.4 (Figure 4-16) is attributed to significant generation of soluble Fe(II). However, since Fe(II) oxidation was rapid at higher pH values, both pretreatment methods achieved similar NOM removals at higher pH values (Figure 4-16d and e). Results presented in Figure 4-14 and Figure 4-15 demonstrate that under optimized conditions, iron chemical coagulation pretreatment would improve MF performance both in terms of filtered water quality and permeate fluxes compared with iron electrocoagulation.

Specific ultraviolet absorbance ([SUVA] defined as UV_{254} expressed in m^{-1} divided by the DOC concentration expressed in mg/L) is a good surrogate for the humic content of water (Croué et al. 1999) and can also provide an indication of the reactivity of NOM to form DBPs. As summarized in Figure 4-16a, b, d, and e, chemical coagulation and electrocoagulation pretreatment removed UV_{254} to a greater extent than DOC. Therefore, microfiltered waters with coagulation pretreatment had a lower SUVA than the feed water (Figure 4-16c and f). Further, SUVA values decreased with coagulant dosage. Hence, DBP control by chemical coagulation pretreatment to MF is achieved simultaneously by reducing the reactivity of NOM with chlorine and removing precursors measured as DOC. Lower SUVA values observed for chemically coagulated samples than electro-coagulated samples provide additional evidence for the better performance of chemical coagulation over electrocoagulation.

Figure 4-17 depicts representative scanning electron micrographs of the clean membrane as well as those fouled by raw, chemically coagulated, and electrocoagulated water.

The porous structure and tortuous nature of pores in the microfilter employed is evident from Figure 4-17a. The heterogeneous nature of natural colloidal matter in terms of size and shape (and potentially composition) can be seen in Figure 4-17b. The membrane surface was completely covered and a thick cake layer was formed by particles present in Lake Houston water after 100 mL was filtered. Chemical coagulation produced a thick cake layer of ferric hydroxide flocs which appeared to have completely enveloped all particles in the raw water (Figure 4-17c). In contrast, natural colloids were found to be interspersed with ferric hydroxide flocs for all electrocoagulated samples (e.g. Figure 4-17d). This is consistent with enhanced *in situ* oxidation of adsorbed Fe(II) to form more soluble Fe(III) species (as shown in Stumm and Morgan 1996) resulting in poor electrocoagulation performance summarized in Figure 4-16

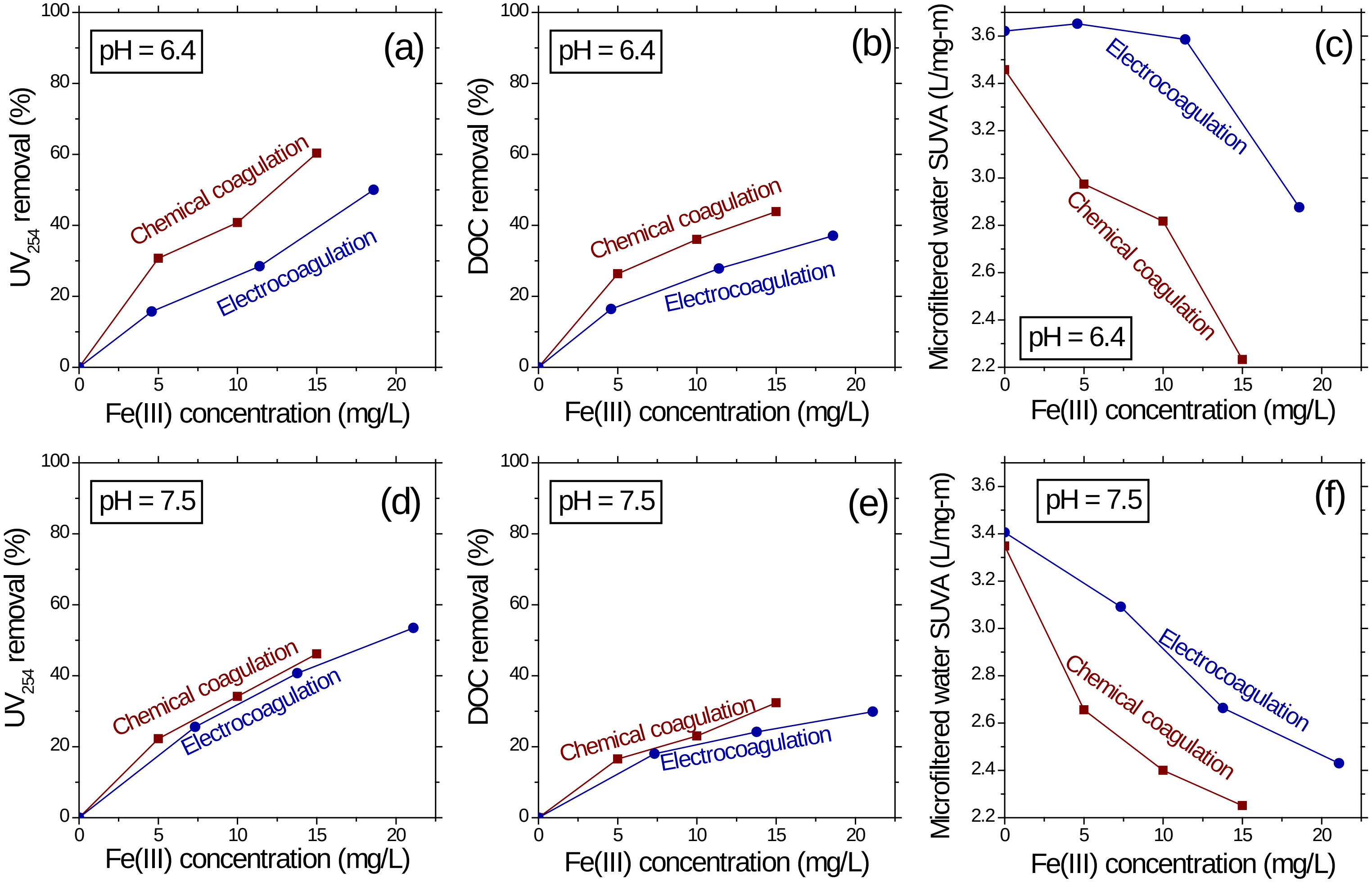


Figure 4-15.—Comparisons of NOM removal by chemical- and electrocoagulation.

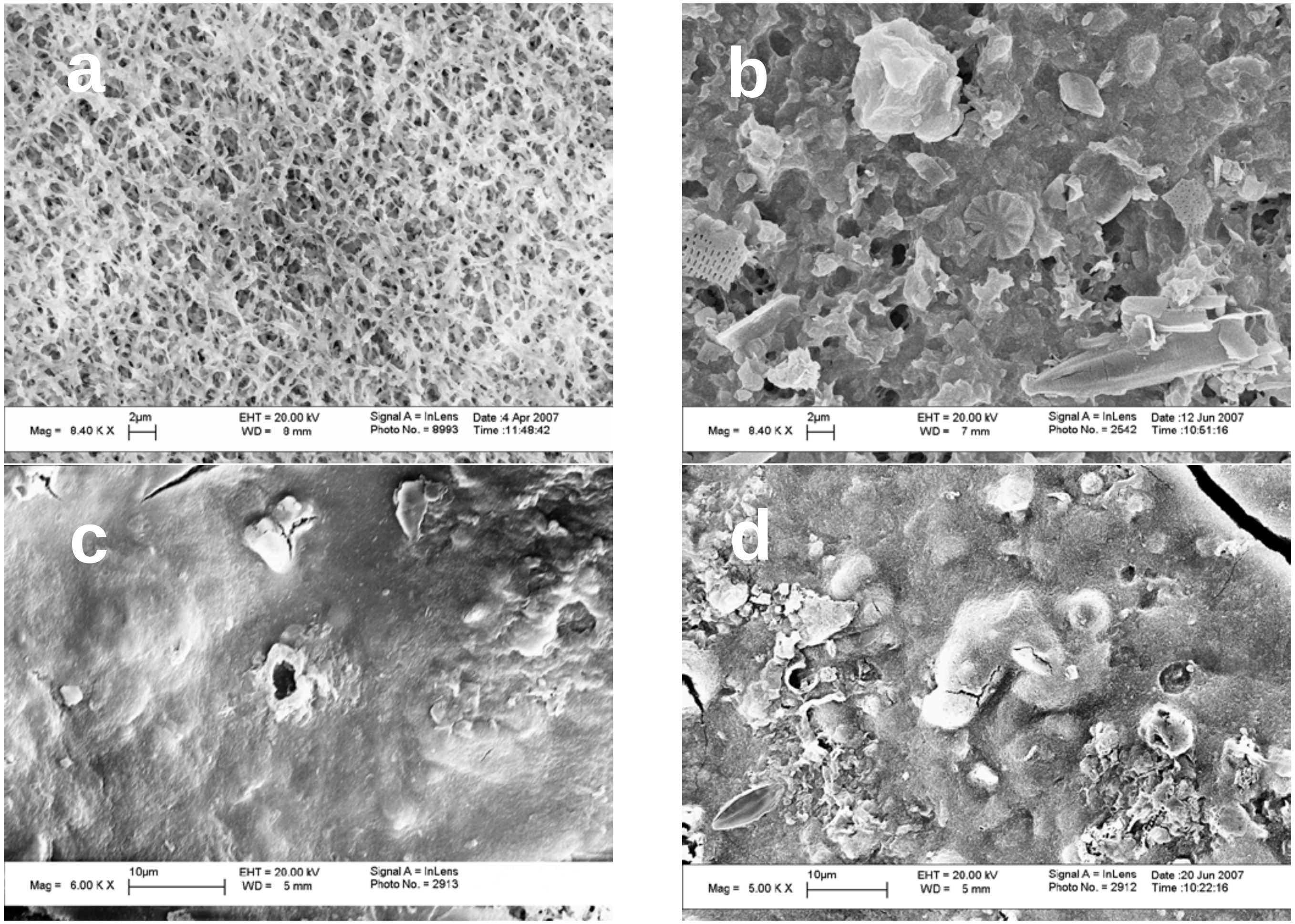


Figure 4-16.—Scanning electron micrographs of a clean membrane (a), cakes formed by filtering 100 mL raw water (b), chemically coagulated water (c), and electrocoagulated water (d) at pH 8.3 and 10 mg Fe(III)/L.

4. VIRUS REMOVAL

4.1. Introduction

4.1.1. Viruses: Occurrences, Health effects and Regulations

The presence of pathogenic microorganisms in drinking water poses a serious threat to human health. These pathogens include various types of viruses, bacteria, protozoa and other microorganisms. In 1990, the EPA concluded that exposure to pathogens was likely the greatest remaining health risk management challenge for drinking water suppliers (EPA nd a). The contamination of water systems with pathogens is often due to floodwaters, fecal matter from sewage discharges, leaking septic tanks and run-off from animal feedlots into bodies of water. Ingesting these harmful pathogens can cause severe disorders in the human digestive system. Of all pathogenic microorganisms, viruses are the major cause of concern for the outbreak of waterborne diseases. Viruses have been shown to be responsible for approximately 80% of disease outbreaks for which infectious agents were identifiable (Ryan et al. 2002). Especially important with respect to health are the viruses that infect the gastrointestinal tract of man and are excreted with human feces. The most common route of virus exposure is through fecal-oral route although exposure through dermal contact and respiratory intake are also other possible routes of exposure. This is related to use of reclaimed water for domestic, agricultural, and recreational purposes. The various types of viruses excreted with feces include polioviruses, coxsackieviruses, echoviruses, adenoviruses, reoviruses, rotaviruses, hepatitis A viruses, and Norwalk-type viruses. These viruses are readily transmitted through water and cause several diseases on exposure including: gastrointestinal illness, brain, eye and liver infections, heart anomalies, diarrhea, vomiting and respiratory diseases. Thus, the potential for viral infections makes virus removal the most important task of water treatment.

The increasing concern over the spread of water-borne diseases due to microbial contamination prompted the EPA to propose stringent rules to regulate the presence of microbial pathogens in drinking water. The Safe Drinking Water Act (SDWA) was promulgated by the EPA in 1974 to enforce lawful standards on drinking water contaminants (EPA nd b). Under this act, EPA established treatment technique (TT) requirements for microbial contaminants which are generally expressed as Log reduction value (LRV) in Equation 6:

$$LRV = \log(C_F/C_P) \quad \text{(Equation 6)}$$

Where:

- C_F is the feed virus concentration
- C_P is the process effluent virus concentration.

Later in the 1989 SDWA amendments, EPA implemented the Surface Water Treatment Rule (SWTR) which mandates against these water-borne pathogens that cause diseases. The rule requires water treatment plants to use disinfection and filtration techniques on surface water to reduce the occurrence of unsafe levels of pathogenic microorganisms (EPA nd b). Under the SWTR, EPA has set Maximum Contaminant Level Goals (MCLG) of zero for viruses, *Giardia*, and *Legionella*, since any amount of exposure to these pathogens causes some health risk. SWTR mandates at least 99.99% removal or 4-log removal of viruses in all drinking water treatment plants in the U.S. Following the SWTR, EPA implemented the Interim Enhanced SWTR in 1998 and the Long Term 1 Enhanced SWTR in 2002, which ensured further control of microbial pathogens, including the protozoan *Cryptosporidium*, in drinking water. This further strengthened the monitoring of microbial contaminants in drinking water supplies thereby reducing public health risk.

4.1.2. Properties of viruses

Viruses are associations of macromolecules made up of nucleic acid, either deoxyribonucleic acid (DNA) or ribonucleic acid (RNA) encapsulated within a protein capsid. These are small infectious agents, usually 20 to 200 nm in size. Viruses are obligate intracellular parasites that cannot replicate outside a living cell. They are completely dependent on host cells for metabolism and growth. The capsid surrounding the nucleic acid core is made up of polypeptides that contain weak ionizable acidic and basic groups. The ionization of the protein coat causes the viruses to behave as charged particles in aqueous media. The net charge is a function of pH, ionic composition and ionic strength of the solution. In general, viruses are negatively charged under most natural environmental conditions, which enable them to favorably bind or adsorb onto positively charged particles through electrostatic attraction.

Since viruses replicate only inside living cells, they need appropriate hosts for their growth. For laboratory studies, most viruses are grown in cell or tissue cultures prepared in a culture media consisting of rich nutrients such as amino acids, salts, glucose etc. Among the various methods of quantification of viruses, the agar overlay technique (Adams 1959) with some modifications has been commonly implemented by several researchers (Jacangelo et al. 1995a, You et al. 2005). In this technique, molten agar is mixed with the virus sample and a culture medium of the host bacterium and is overlaid on a thin layer of agar medium adhered to a petri dish, and then incubated at a suitable temperature. When a virus particle infects a host cell growing on the layer, a zone of lysis called a *plaque* is formed. By counting the number of *plaque-forming units* (PFU), the concentration of the viruses in the original sample can be calculated.

4.1.3. Bacteriophages

Bacteriophages (phages) are viruses that infect bacteria. They are the most common type of viruses used as models or surrogates for enteric viruses in water. In drinking water treatment, phages are used as indicator organisms to measure the effectiveness of the treatment processes in removing pathogenic viruses. Phages are good model viruses because they closely resemble the structure, composition, morphology, size, and site of replication of the enteric viruses (Sundram et al. 2002). Further, compared with enteric viruses, phages are detectable by simple, inexpensive, and rapid techniques. In addition, phages also closely meet the basic requirements of an indicator organism as listed below.

1. They should always be present in water environments when enteric viruses are present.
2. They should be of fecal origin and present in large numbers.
3. They should be easy to isolate, identify, and enumerate compared to enteric viruses.
4. They should respond to treatment processes in a similar manner as enteric viruses.

MS2 bacteriophage has been commonly used as a model organism in several water treatment studies (Jacangelo et al. 1995a, You et al. 2005, Zhu et al. 2005a, Zhu et al. 2005b). MS2 is a RNA virus that can only replicate within its host bacterium, *Escherichia coli*. It is a good surrogate for pathogenic enteric viruses such as Hepatitis A and polio virus because it is similar in size, shape, and nucleic acid composition to them. It can be easily cultured and enumerated using the agar overlay technique as described previously.

4.1.4. Membrane processes for virus removal

Membrane technology has provided water and wastewater treatment utilities with attractive alternative treatment options to meet such strict regulations as the new rules proposed on water quality standards. The water treatment industry has used membranes for several years as membranes are highly effective in removing particles, turbidity, bacteria, viruses, and other microorganisms from untreated water. Low pressure membrane processes such as MF and UF have been widely used for removing particulates and microbes. According to reported literature, over 300 water treatment facilities in U.S. and Europe use MF and UF modules (Gitis et al. 2006).

Several reported studies have focused on MF and UF technologies for microbial pathogen removal (Jacangelo et al. 1995a, Jacangelo et al. 1991, Madaeni et al. 1995, Urase et al. 1996, Zhu et al. 2005a, and Zhu et al. 2005b). UF was found to be highly effective in removing viruses, *Giardia*, and bacteria as they are much larger than the membrane pore size (Jacangelo et al. 1995a, Jacangelo et al. 1991, and Madaeni et al. 1995). Though MF was efficient in the removal of bacteria and Giardia (Jacangelo et al. 1995a), only partial removal of viruses was observed (Jacangelo et al. 1995a, Madaeni et al. 1995, Urase et al. 1996, Zhu et al. 2005a, and Zhu et al. 2005b). MF membranes alone are not effective barriers for virus removal because the sizes of the viruses are often smaller than membrane pore sizes. The MS2 virus removal in the above studies ranged from 0.2-3 logs, which is significantly less than 4-log virus removal mandated by the SWTR. Hence, MF by itself cannot meet the SWTR requirements for virus removal but MF can easily meet bacteria and protozoa standards.

Although MF is highly efficient for removing bacteria and protozoa, its application is hindered largely due to membrane fouling. Fouling refers to the deposition of solid material from the feed stream on or into a membrane that increases the total resistance to water flow across the membrane. The fouling of membranes has been primarily attributed to the presence of colloids and dissolved natural organic matter in raw water (Fan et al. 2001, Hong and Elimelech 1997, Howe and Clark 2002, and Nilson and DiGiano 1996). Adding coagulation pretreatment before microfiltration has been suggested to improve membrane performance by reducing fouling and improving the removal of dissolved organic matter and pathogens. Several published results have shown chemical coagulation to be effective in reducing membrane fouling and providing high quality water (Howe and Clark 2002, Howe and Clark 2006,and Mallevialle et al. 1996). The two common coagulation techniques used in water treatment are chemical coagulation and electrocoagulation.

4.1.5. Chemical coagulation pretreatment

Chemical coagulation (CC) is the process of destabilizing colloidal particles so that they can combine and aggregate to form larger particles during flocculation. The four main destabilization mechanisms are:

(1) Compression of the diffuse layer

(2) Adsorption and charge neutralization

(3) Enmeshment in a precipitate (or) sweep flocculation

(4) Adsorption and inter-particle bridging

All the above mechanisms are important in drinking water treatment. Which mechanism predominates depends on the type and dosage of coagulant used. Aluminum and iron salts have been widely used as coagulants in several studies

(Han et al. 2002 and Zhu et al. 2005b). Both of these salts can effectively destabilize particles using two different mechanisms: adsorption and charge neutralization at low concentration and sweep flocculation at higher concentrations. Aluminum is effective in the pH range of 5 to 9, whereas iron is effective over a wider pH range from 2 to 10. This is primarily due to the difference in the solubilities of the two metals. However, both coagulants have been effectively used in various drinking water treatment systems.

Both chemical coagulation with and without filtration have been shown to be highly effective for virus removal. Recent studies have shown that iron coagulation alone played an important role in removing viruses; i.e., coagulation followed by sedimentation was as effective as coagulation followed by microfiltration in removing the viruses (Zhu 2004).(Zhu et al. 2005b) reported no inactivation of MS2 virus using aluminum coagulation with aluminum doses of 1.0 and 5.0 mg/L. This is in agreement with the virus concentration procedure in Standard Methods (Clesceri et al. 1998), in which viruses are concentrated by precipitation with aluminum hydroxide. Up to 99% recovery of viruses using this standard method was reported by (Wallis and Melnick 1967). Shelton and Drewry (1973) reported aluminum can remove more than 99.0% of viruses from both surface water and wastewater. Shelton and Drewry also found ferric chloride to be reasonably effective for removing viruses with 94.6 and 99.1% removals in wastewater and surface water respectively. Chaudhuri and Engelbrecht (1970) showed 98.0% virus removal using an optimum dose of 40 to 50 mg/L of alum.

Several studies have also reported significant virus removals using coagulation-filtration. (Wang et al. 2005) concluded that coagulation-filtration using polyaluminum chloride could achieve substantial virus removal (> 2-log) but better removal was observed using ultrafiltration. (Matsushita et al. 2005) had reported > 7-log virus removal using 1.6 mg/L polyaluminum chloride dosage in coagulation-ceramic microfiltration system. (Zhu et al. 2005b) used $FeCl_3$ as the coagulant and they found that iron coagulation before microfiltration dramatically improved virus removal and > 4-log removal of viruses was observed with a 10 mg/L iron dose.

4.1.6. Electrocoagulation pretreatment

EC has been used in the water industry since the 19th century when a treatment plant was successfully built in London (Vik et al. 1984). In the early 1900s, EC plants with steel electrodes were installed in the U.S. An EC treatment plant called the “Electronic-coagulator” using aluminum anodes was used in the year 1940 (Matteson et al. 1995). A similar process to treat river water was used in Britain in 1956 using iron electrodes (Matteson et al. 1995). EC has been effectively used for several applications such as treatment of potable water (Holt et al. 2002 and Vik et al. 1984), treatment of urban wastewater (Kobya et al. 2006 and Pouet and Grasmick 1995), treatment to remove heavy metals (Hansen et al. 2006, Mills 2000, and Parga et al. 2005), treatment to remove turbidity (Han et al.

2002) and microorganisms (Mills 2000 and Zhu et al. 2005a) and treatment to remove dyes (Can et al. 2003 and Caňizares et al. 2006). Kobya et al. (2006) reported 93 % removal of COD with aluminum electrodes and 98 % removal of oil-grease with iron electrodes. Parga et al. (2005) reported up to 99% removal of arsenic and chromium from well water in a pilot-plant EC study in Mexico.

Several researchers have also compared EC and CC for various applications. Han et al. (2002) found EC to be more efficient than CC for turbidity removal using aluminum electrodes. More recently, researchers at the University of Houston reported iron EC to be better than CC for virus removal in synthetic water without particulates or NOM (Zhu et al. 2005a). These studies reported that regardless of the pH, EC required less iron dosage compared to CC to achieve a given MS2 log removal. At > 10 mg/L iron dosages, EC achieved more than the SWTR requirement of 4-log removal (99.99 %). The excellent virus removals observed in synthetic water should be verified in natural water to be more beneficial for the water treatment industry. This formed the basis of the present work, which focuses on EC for virus removal in the presence of turbidity, NOM, and other contaminants typical of natural surface water.

4.1.7. Objectives

The overall objective of this aspect of research was to study the performance of electrocoagulation-microfiltration (EC-MF) process for virus removal in natural surface water with NOM, turbidity and other contaminants present. The specific objectives were to:

(1) Determine the effect of coagulant dose and pH on virus removal in synthetic water and natural water using CC-MF and EC-MF

(2) Compare EC-MF performance for virus removal in synthetic water and natural water

(3) Compare EC-MF and CC-MF for virus removal in synthetic water and natural water

To accomplish the above objectives, the following specific tasks were carried out using non-pathogenic MS2 bacteriophage as the indicator virus:

i. Determine the effect of Fe(III) dose and pH on MS2 virus removal by CC-MF in synthetic water without NOM.
ii. Determine the effect of Fe(III) dose and pH on MS2 virus removal by CC-MF in natural water with NOM.
iii. Study the deliberate oxidation of iron rods during electrolysis in the EC unit and examine the oxidation state (ferrous and ferric) of the iron produced.

iv. Determine the effect of EC-syringe microfiltration (EC-SF) on virus removal from natural water with NOM, as a rapid screening test for EC-MF.

v. Determine the effect of iron generated and pH on MS2 removal by EC-MF in natural water with NOM.

vi. Determine the effect of iron generated and pH on MS2 removal by EC-MF in synthetic water without NOM.

vii. Determine the effect of Fe(II) dose on MS2 removal by CC-SF at pH 7.5 in natural water with NOM and in synthetic water without NOM

viii. Study the effect of increasing NOM concentration on MS2 removal at pH 7.5 and constant Fe(II) dose.

ix. Determine the Fe(Total) removal as a function of NOM concentration and Fe(II) dose.

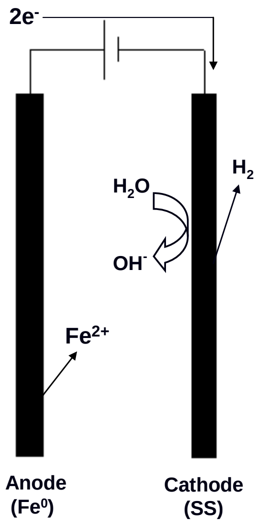
The above list of tasks was initially much shorter but expanded during the course of the research, because the initial EC experiments indicated ineffective removal of viruses from natural water at pH 6.4-7.5. However as discussed before, (Zhu et al. 2005a) had reported high virus removal using EC-MF. These researchers explained that such high removals were due to the generation of Fe(III) iron in the EC unit that oxidizes to form ferric hydroxides and precipitates with the viruses attached to it. However, in our work and in recent studies, generation of Fe(II)—and not Fe(III)—was observed in the EC unit at pH 6.5-8.5. Furthermore, we found the oxidation of Fe(II) to Fe(III) was highly pH dependent (Lakshamanan 2007). Due to the ambiguity in the mechanisms involved in EC, it was important to understand the fundamentals of the EC theory and the electrolytic oxidation of iron in the EC unit to better interpret its performance. Thus all the tasks outlined above were necessary. The next section discusses the process fundamentals occurring in the EC unit in detail.

4.2. Process fundamentals of the electro-coagulation unit

The EC unit comprises an electrochemical cell consisting of an electrolysis chamber, electrolyte, cathode, and anode. The energy needed for electrolysis is provided by the application of electric current, which drives the chemical reactions. Two common kinds of electrodes can be used in the EC cell: inert electrodes and sacrificial electrodes. Inert electrodes do not undergo any change during electrolysis and do not participate in the chemical reaction. Some inert electrodes that are frequently used include platinum, carbon, mercury, and graphite. Sacrificial electrodes, such as iron and aluminum, undergo chemical change during electrolysis and hence are self-destructive in nature. Both are commonly used as sacrificial anodes in EC applications. This research studies electrocoagulation using iron anodes to form the $Fe(OH)_3(s)$ coagulant species for virus adsorption and removal.

The basis of electrolysis was developed by the famous chemist and physicist Michael Faraday. According to the Faraday's law of electrolysis, the theoretical amount of species generated (i.e., iron in this research) is a function of current and generation time and it can be calculated using Equation 1.

When iron is used as the sacrificial anode, as in this research, the electrolytic oxidation of iron (Fe^0) can theoretically result in the generation of ferric (Fe[III]) or ferrous (Fe[II]) ion. No generally accepted conclusion is available on the electrolytic oxidation of iron from the literature. Various opinions exist in the literature regarding the oxidation state of iron being produced at the anode. While few authors have reported the electrolytic oxidation of iron to produce Fe(III) (Kobya et al. 2006 and Zhu et al. 2005a), others have reported the production of Fe(II) (Hansen et al. 2006 and Mollah et al. 2001). During a study of iron electrocoagulation for virus removal from synthetic water, (Zhu et al. 2005a) reported the generation of Fe(III) in the EC unit, with subsequent hydrolysis to form ferric hydroxides that adsorbed the viruses. These ferric hydroxides then got removed by settling or membrane filtration. As this research was the driving force behind the present work, we expected that Fe(III) would be generated in the EC unit and also cause efficient virus removal in natural water containing NOM and particulate matter. However as explained previously, both in this work and in recent studies (Lakshamanan 2007), Fe(II) rather than Fe(III), was observed to be generated in the EC unit at pH range 6.5-8.5. Also, the oxidation to Fe(II) was found to be highly pH dependent. The summary of the basic electrochemical reactions occurring in the EC unit is shown in Figure 5-1.



1. Anode

   $Fe^0{}_{(s)} = Fe^{2+} + 2e^-$

2. Cathode

   $2H_2O + 2e^- = 2OH^- + H_2$

3. Oxidation of Fe^{2+} to Fe^{3+}

   $4Fe^{2+} + O_2 + 4H^+ = 4Fe^{3+} + 2H_2O$

4. Hydrolysis

   $Fe^{3+} + 3H_2O = Fe(OH)_{3\,(s)} + 3H^+$

5. Combined oxidation and hydrolysis

   $4Fe^{2+} + O_2 + 2H_2O + 8OH^- = 4Fe(OH)_{3(s)}$

Figure 5-1.—Reactions in EC unit.

As reported by (Lakshamanan 2007), the electrolytic oxidation of iron anode results in generation of ferrous iron with the release of 2 electrons (equation 1 in Figure 5-1). The electrolytic reduction of water produces hydrogen gas and hydroxides at the cathode (equation 2 in Figure 5-1). The formation of hydroxides would result in an increase in pH during electrolysis, however (Lakshamanan 2007) reported insignificant pH change at the end of 2 minute of mixing, because the protons resulting from the hydrolysis of Fe(III) ions, produced by prior oxidation of Fe(II) to Fe(III) using dissolved oxygen (DO; equation 3 in Figure 5-1) were consumed by the hydroxides to form ferric hydroxides (equation 4 in Figure 5-1). The overall reaction of Fe(II) oxidation and hydrolysis is represented by equation 5 in Figure 5-1. Importantly, the rate of oxidation of Fe(II) to Fe(III) depends on the pH and the DO concentration. As explained in literature (Singer and Stumm 1970 and Stumm and Lee 1961), the oxidation rate of Fe(II) follows first order kinetics with respect to Fe(II) and oxygen concentrations and is second order with respect to hydroxide concentration. The kinetic relationship for Fe(II) oxidation is shown in Equation 5.

From this rate expression, it can be seen that a unit increase in the pH results in 100-fold increase in the rate of Fe(II) oxidation. Since Fe(II) is highly soluble compared with Fe(III), the ferrous ions generated in the EC unit will easily pass through a 0.2 µm filter and not cause any virus removal. However, the ferric hydroxides $Fe(OH)_3(s)$ will precipitate and the resulting flocs will adsorb the viruses and be retained on a 0.2 µm membrane filter.

Therefore, the key to effective electrocoagulation is the oxidation of Fe(II) to Fe(III) and the subsequent hydrolysis of Fe(III). With this understanding of the fundamentals of the electrolytic oxidation of iron in the EC unit, we conducted the EC experiments to study the efficiency of electrocoagulation on virus removal.

4.3. Materials and methods

4.3.1. Test waters

Lake Houston water and synthetic water were used as the test waters for all the experiments. The lake water was sampled on January 12th, 2006 and July 7th, 2006. The older water sample can be considered to be representative of the winter season while the newer water sample represents the summer season. Several water quality parameters have already been summarized in Figure 2-2.

Synthetic water of the composition reported elsewhere (Zhu et al. 2005a and Zhu et al. 2005b) was used in the coagulation experiments as a benchmark for comparison purposes. The synthetic water consisted of de-ionized (DI) water spiked with 3.0 mM reagent grade $NaHCO_3$ (EM Science, New Jersey) and 1.0 mM reagent grade $CaCl_2$ (EM Science, Darmstadt, Germany). The water sample had an unadjusted pH of 7.5 ± 0.1. It should be noted that the synthetic water did not contain any natural organic matter (NOM).

4.3.2. Virus

The MS2 virus, a bacteriophage, was used as a surrogate organism for all the experiments. MS2 is 0.025 µm in size, icosahedral in shape, and contains a single strand of ribonucleic acid with 3569 nucleotides (Valegård et al. 1990). As explained previously, MS2 is an ideal model virus to study because it is non-pathogenic and is similar in shape, size, structure and nucleic acid composition to pathogenic human viruses such as hepatitis A virus and polio virus. Being one of the smallest viruses, MS2 is harder to filter. Therefore using MS2 presents a worst-case challenge to MF membranes. Furthermore, MS2 bacteriophage assays are easy to perform, inexpensive and the waste can be easily disposed. MS2 virus stock was purchased from American Type Culture Collection (American Water Works Association [AWWA] # 15597-B1).

MS2 viruses were propagated and enumerated by implementing the EPA's Information Collection Rule Method (ICR) for Male-Specific Coliphage assay (EPA, 1996). In this method, *Escherichia coli (E. coli)* (American Type Culture Collection [ATCC] #15597) was used as the ATCC-recommended host organism in place of *E. coli* Famp. The entire procedure as adapted from EPA's ICR method (EPA, 1996) is described below with necessary modifications.

4.3.3. Apparatus, materials, media and reagents

The following apparatus and materials were used during the virus enumeration experiments:

1. Centrifuge
2. Sterilizing syringe micro-filter - 0.2 µm and 0.45 µm
3. Water bath and dry bath - set at 44.5 ± 1 °C
4. Incubator - set at 37 ± 0.5 °C
5. Autoclave - set at 121 °C
6. Inoculating loop
7. Laboratory balance
8. Pipets-Pipettors -100 microliters (µL), 1 mL, 5 mL and 10 mL
9. Petri dishes, 100 × 15 millimeters (mm)
10. Glass bottles (autoclaved), capped, 100 mL and 1 L capacity
11. Centrifuge tubes - 1.5 mL,15 mL and 50 mL

The following media and the reagents were prepared to conduct the bacteria and virus assays. The amount of media prepared was increased proportionally based on the number of samples to be analyzed.

1. **Tryptone bottom agar**. To prepare bottom agar, 1.0 gram (g) Bacto tryptone, 0.1 g yeast extract, 0.1 g glucose, 0.8 g NaCl, 0.022 g $CaCl_2$, and 1.5 g of Bacto agar were added to 100 mL of de-ionized (DI) water in a 250 mL flask. The contents were dissolved thoroughly in the autoclave. After autoclaving, 15 mL of the agar was poured into sterile petri dishes and allowed to harden. The plates were prepared one day prior to sample analysis and the unused plates were discarded.
2. **Tryptone top agar.** The top agar was prepared using the same ingredients listed for bottom agar except that 0.7 g of Bacto agar was used. The contents were mixed and dissolved by autoclaving at 121°C for 15 minutes, and the agar was kept in the water bath set at 44.5 ± 1°C. The top agar was prepared on the day of the sample analysis, and the leftover agar was discarded to avoid contamination during virus enumeration.
3. **Tryptone broth.** The broth was prepared using the same ingredients listed for top agar without the addition of Bacto agar. After autoclaving at 121 °C for 15 minutes, the broth was allowed to cool to room temperature (23 ± 2 °C) before usage.
4. **Saline-calcium solution.** 8.5 g NaCl and 0.22 g $CaCl_2$ were dissolved in 1 L of DI water. A 4.5 mL aliquot of the solution was dispensed in 16 × 100 mm capped test-tubes and sterilized by autoclaving at 121 °C for 15 min. These saline-calcium tubes were used as dilution tubes to prepare several dilutions of the virus samples.
5. **Beef extract solution.** A 1.5% beef extract solution was prepared by dissolving 1.5 g of beef extract powder and 0.375 g of glycine in 100 mL DI water. The pH of the solution ranged from 7.0 to 7.5. The solution was autoclaved at 121 °C for 15 min and cooled to room temperature before use. The solution was stored for one week at 4 °C. The solution was primarily used to pretreat the syringe micro-filters just before their usage, to prevent phage adsorption onto the filter.

The host *E. coli* was propagated using the ICR method as described below. The host culture of *E. coli* was inoculated onto tryptone agar plates using a sterile inoculating loop by streaking the inoculum on the agar surface. The streaked plates were incubated for 20 to 24 hours at 37 °C. The culture was stored for up to 2 weeks at 4 °C.

E. coli culture from the streaked agar plates was inoculated into tryptone broth using a sterile inoculating loop and incubated in a shaking incubator for a period of 18-20 hours. The host culture obtained was used for MS2 enumeration and propagation. The amount of broth and inoculum used was altered proportionally based on need. The procedure for *E. coli* propagation is depicted in Figure 5-2.

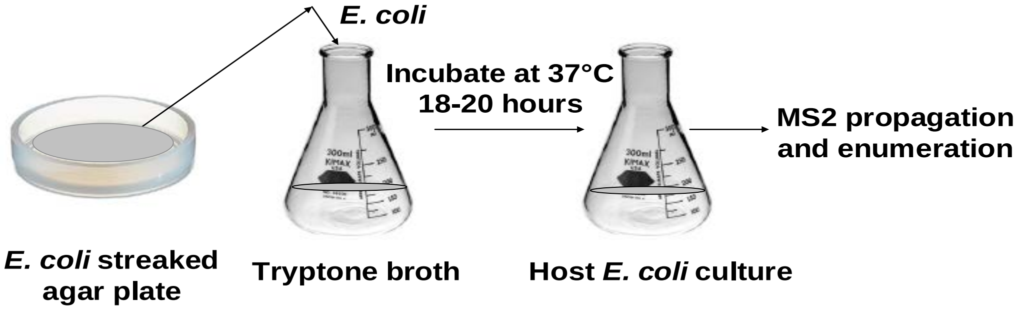


Figure 5-2.—Procedure for E. coli propagation.

The re-hydrated MS2 stock was added to the host culture and the solution was incubated for 18-20 hours at 37 °C in a shaking incubator. The propagated viruses were separated from the cellular debris by centrifugation at 5000 rpm at 4 °C for 10 min. The supernatant containing the viruses was filtered using 0.22 µm syringe filters. The filtrate was the MS2 stock solution, and it was refrigerated at 4 °C and used as needed. The procedure for virus propagation is depicted in Figure 5-3.

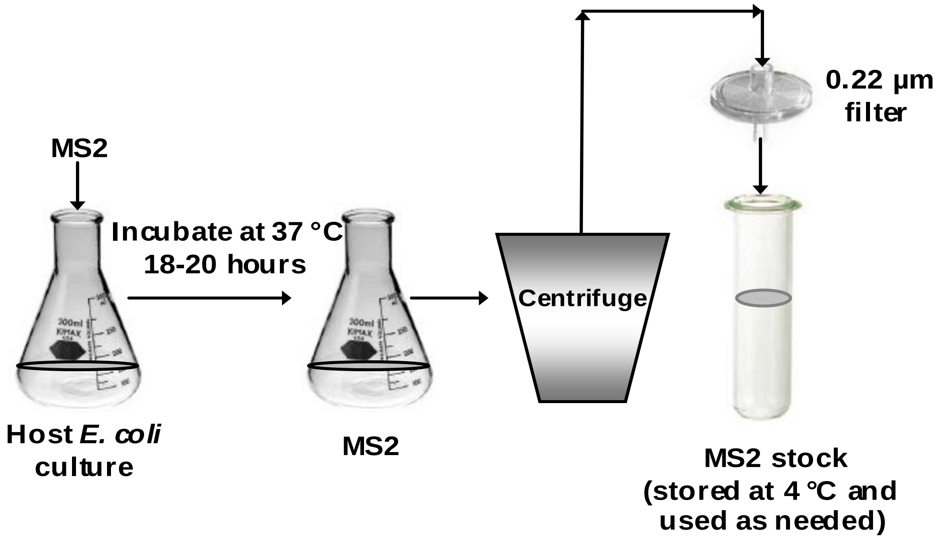


Figure 5-3.—Procedure for MS2 propagation.

MS2 viruses were enumerated using the double agar layer (DAL) procedure reported by the EPA under ICR (EPA, 1996). The procedure for MS2 enumeration is shown in Figure 5-4. The tryptone agar plates were labeled with the sample identification information. The MS2 sample was serially diluted using saline-calcium (SC) dilution tubes, and 1 mL of each dilution was added to 0.1 mL of host *E. coli* culture. Some coagulated samples had low virus concentrations, and for these samples, 0.5 mL of the raw sample was added to the host without any dilution. Three mL of molten tryptone top agar, maintained at 44.5 ± 1 °C in a water bath, was added to the above mixture and immediately mixed well and poured over the labeled agar plates. The plates were rotated to spread the suspension evenly over the surface of the tryptone bottom agar. The plates were left on an even surface to allow the top agar to solidify. The inoculated plates were incubated at 37 ± 0.5 °C overnight and the plaques were examined and counted the following day. The results were expressed in *plaque forming units* (PFU). An agar plate with the MS2 plaques grown on the bottom agar after incubation is shown in Figure 5-4 and Figure 5-5.

To avoid interference from bacteria during plaque counting, the MS2-spiked lake water samples were pre-filtered using 0.45 µm syringe filters, pre-treated with 1.5% beef extract solution. The beef extract pre-treatment helped to minimize phage adsorption to the filter. To quantify the removal of MS2 due to the 0.45 µm pre-filtration step, preliminary experiments were conducted in synthetic water and the concentration of MS2 before and after filtration was obtained, and log removal was calculated. It should be noted that the synthetic water samples were devoid of bacteria and other microorganisms and, therefore, could be easily enumerated for viruses without pre-filtration.

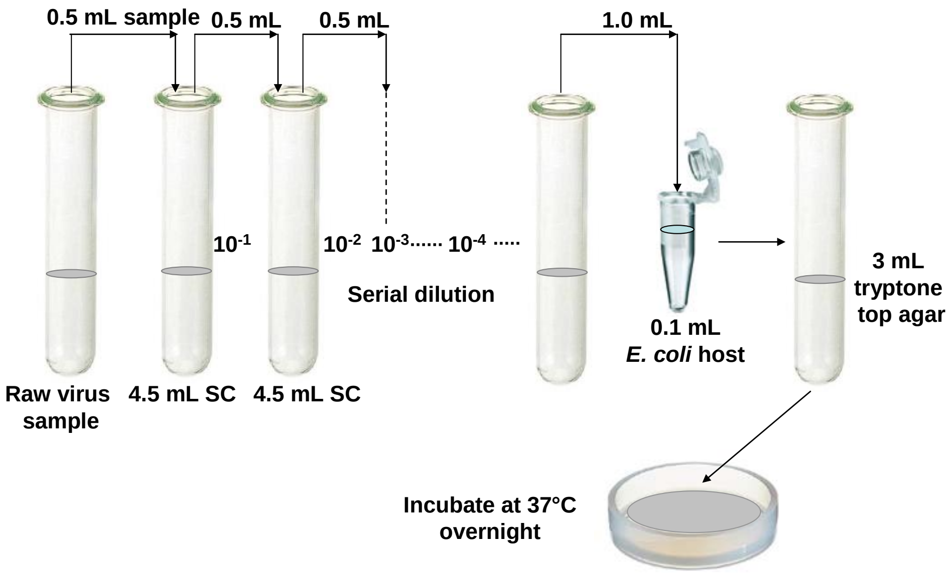


Figure 5-4.—Procedure for MS2 enumeration.

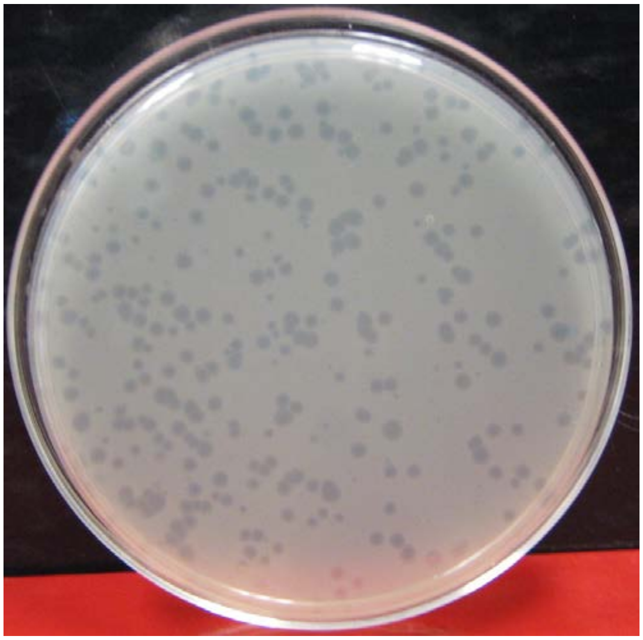


Figure 5-5.—Agar plate with MS2 plaques.

The waste, including used plates, pipette tips, and centrifuge tubes, generated from the virus enumeration experiments was autoclaved in a biology bag at 121°C for 15 minutes. The glassware was disposed of in separate glass disposal containers. The autoclaved bag and the glass disposal containers were picked up by the University of Houston Safety and Risk Management Office for secure off-site disposal.

The virus removal is expressed as log reduction value (LRV) was calculated using the ratio of feed and the permeate concentration of the MS2 virus (Equation 6).

4.3.4. Procedure for chemical coagulation using ferric iron as the coagulant

Two types of iron coagulant: ferric and ferrous were used in this study. The ferric coagulant was prepared by dissolving reagent-grade ferric chloride into DI water to prepare 1.0 g/L Fe(III) solution. The ferrous coagulant was prepared by dissolving reagent-grade ferrous sulfate into DI water to give 1.0 g/L Fe(II) solution. Fresh coagulant solutions were prepared before the start of every experiment.

The coagulation experiments were conducted using a conventional bench-scale jar tester having a six-position flat-blade stirrer (Model PB-900, Phipps and Bird Inc., Richmond, Virginia). One liter of pH-adjusted source water spiked with a known concentration of MS2 was placed in each 2-L jar and dosed with a predetermined concentration of $FeCl_3$ stock solution (0 – 10 mg/L Fe (III)). The samples were rapidly mixed for 1.0 min at 300 rpm (mean velocity gradient = 495 s^{-1}). Since the coagulant used was highly acidic, it reduced the pH of the water samples after dosing. Hence, during the rapid mix stage, the pH was adjusted to the desired value (6.4, 7.5 or 8.3) by adding 0.5 N NaOH. Pre-titrations were conducted, before the experiment, using varying amounts of Fe(III) to determine the amount of NaOH needed to achieve the desired pHs at the different dosages. After mixing, the samples were flocculated by slow mixing for 30 minutes at 40 rpm (mean velocity gradient = 32 s^{-1}). The coagulated/flocculated samples were then transferred to the microfiltration system.

4.3.5. Electrocoagulation unit

A custom-designed bench-scale EC unit, fabricated at University of Houston, was used to perform the electrocoagulation experiments. This unit is a modified version of a similar unit reported elsewhere (Zhu et al. 2005a). The original unit was re-designed to reduce its dead volume and improve its operational stability and ease of electrode cleaning and assembly. The redesigned unit consists of a 260 mL active volume, flow-through electrode chamber with three rod-shaped iron anodes surrounded by three porous cylindrical stainless steel cathodes. The total active surface area of an iron anode is 110 cm^2. As shown schematically in Figure 5-6 and detailed in Table 5-1, the recycle pump discharged the water into the annular area between the cathode and anode of each of the three anode-cathode pairs in order to flush the anode where iron coagulant was generated.

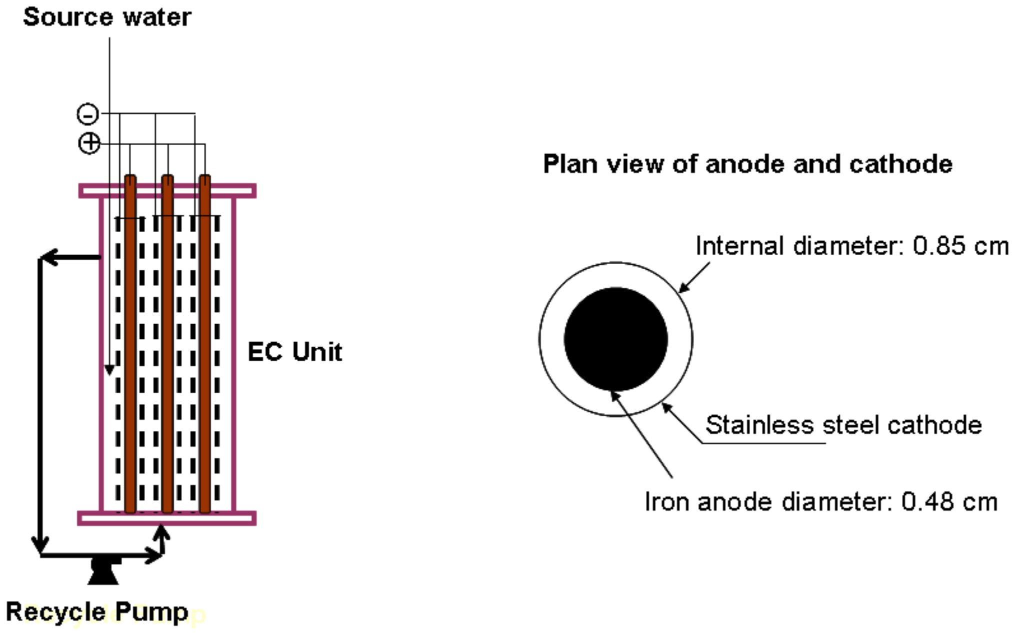


Figure 5-6.—Schematic representation of the bench-scale EC unit.

Table 5-1.—Anode and cathode compositions

	Anode (Industrial grade)	Cathode
	1018 CF Bar ASTM A108	**Stainless steel-316**
Chemical Composition		
Iron (wt %)	98.52(*)	61.08 ~ 69.00
Carbon (wt %)	0.17	< 0.08
Manganese (wt %)	0.78	1.5 ~ 2.0
Phosphorous (wt %)	0.019	< 0.04
Sulfur (wt %)	0.025	< 0.30
Silicon (wt %)	0.16	< 1.0
Nickel (wt %)	0.07	12 ~ 14
Chromium (wt %)	0.05	17 ~ 18
Copper (wt %)	0.21	< 0.5

The compositions of the iron anode and stainless steel cathode are shown in Table 5-1. As it is common practice in bench-scale electrocoagulation experiments (Han et al. 2002, Holt et al. 2002 and Zhu et al. 2005a), the EC unit was operated in batch mode for each virus removal experiment under non-steady state conditions. A photograph of the EC unit specially designed for this research is shown in Figure 5-7.

Figure 5-7.—Electrocoagulation Unit.

4.3.6. Procedure for electrocoagulation

Source water spiked with MS2 viruses was poured into the top of the EC unit. The pH of the source water was adjusted to the pre-determined value (6.4, 7.5, or 8.3) as needed. By varying the generation time and keeping the current density constant at 0.3 milliampere per square centimeter (mA/cm^2), the amount of iron generated was calculated using the Faraday's law as explained in §4.3.3 and §5.2. The final iron doses generated were in the range of 0 to 10 mg/L. The water in the EC unit was recycled at the rate of 500 milliliters per minute (mL/min) for 2 minutes to ensure that the electrodes remained free of surface deposits and that the iron generated was quickly released from the surface, well-mixed, and hydrolyzed. The electrocoagulated water sample was transferred to the microfiltration system. To simplify the experiments and to obtain preliminary results with the modified EC unit, the coagulated samples were filtered using 0.2 µm syringe filters in the initial-stage experiments. Before filtration, samples were taken for total iron and ferrous iron analysis. It should be noted that the iron rods were thoroughly scrubbed using sand paper (Waterproof Silicon Paper Standing Sheet 100 Grit, C-Weight) before every experiment to remove the rust and hydroxides deposited on the rod thus preventing the effect of electrode passivation on iron generation efficiency (Lakshamanan 2007).

4.3.7. Microfiltration system set-up

After 3 to 10 hours of chemical or electrocoagulation, the coagulated/ flocculated sample was transferred into a feed bottle where it was continuously and gently mixed for up to 2 to 8 hours during membrane filtration depending on the type of sample water used and the coagulation dose. Mixing was necessary to avoid settling of flocs in the feed bottle during filtration. Thus, the total standing time and filtration time for the coagulated samples varied from 3 to 18 hours. The bench-scale membrane system was operated at a constant trans-membrane pressure of 20 pounds per square inch gauge (psig) applied by pressure-regulated compressed air. The water samples were filtered using 0.22 µm pore size, polyvinylidene fluoride (PVDF) membrane filter (Durapore, Millipore Corp, Bedford, Massachusetts) with an effective membrane filtration area of 4.1 cm^2. A fresh membrane was used for each filtration experiment. The filtered water was continuously collected in a glass beaker, and its weight was monitored using a weighing balance. The samples were collected at three different times during filtration for virus enumeration, and the log removal was calculated. The schematic representations of CC-MF and EC-MF are shown in Figure 5-8 and Figure 5-9 respectively.

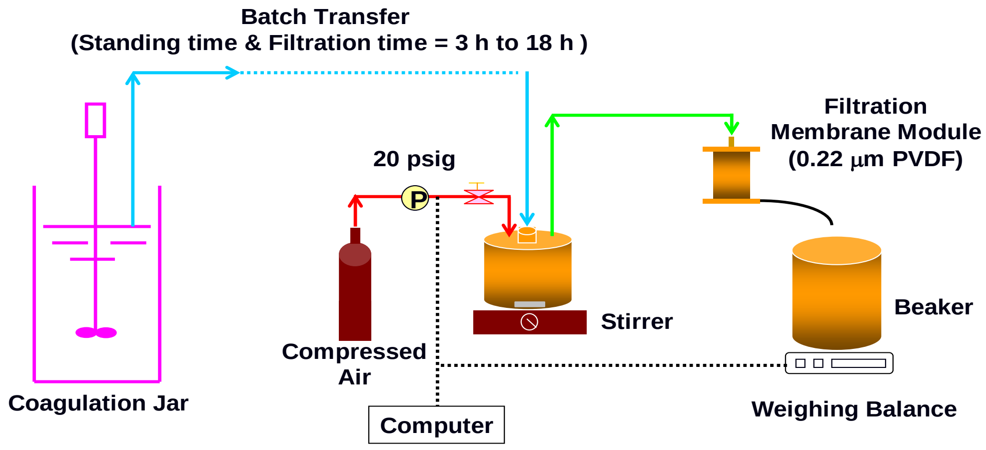


Figure 5-8.—Chemical coagulation – Microfiltration set up.

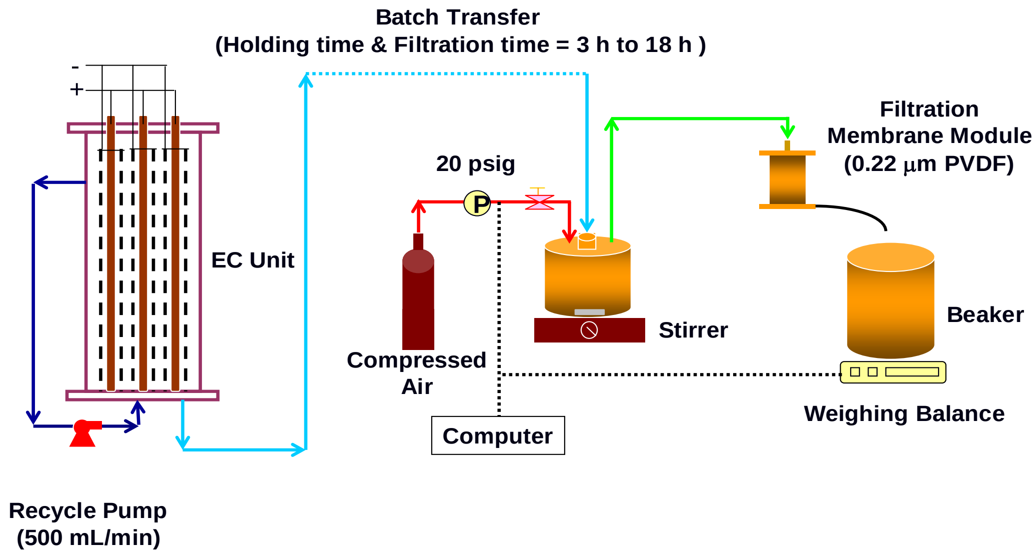


Figure 5-9.—Electrocoagulation – Microfiltration setup.

4.3.8. Procedure for chemical coagulation using ferrous iron as the coagulant

The source water was coagulated with $FeSO_4$ (0-10 mg/L as Fe(II)) to compare chemical coagulation with ferrous iron and electrocoagulation with ferrous iron. As in previous chemical coagulation experiments, one liter of MS2-spiked source water adjusted to pH 7.5 was taken in each 2-L jar and varying concentrations of ferrous were dosed in each of them. The samples were rapidly mixed for 1 min at 300 rpm (mean velocity gradient = 495 s^{-1}). As explained previously, the reduction in pH due to the acidic nature of the coagulant was prevented by the addition of 0.5 N NaOH. After mixing the samples were flocculated by slow mixing for 60 min at 100 rpm. It has been reported that at pH 7.5, one hour flocculation provided sufficient time for the nearly complete (>90%) oxidation of ferrous to ferric (Lakshamanan 2007). Immediately after flocculation, the sample was filtered using 0.2 µm syringe filter. A sample was taken before coagulation and after filtration for virus enumeration, and the log removal was calculated.

4.3.9. Procedure for preparation of Suwannee River NOM solution

Suwannee River NOM purchased from International Humic Substances Society (IHSS) was used as an NOM dose to study the effect of NOM on virus removal in the chemical coagulation experiments. Suwannee River NOM is an aquatic NOM, which is reported to have the elemental composition shown in Table 5-2.

Table 5-2.—Elemental composition % (w/w) of Suwannee River NOM.

Element	% (w/w) values*
C	52.47%
H	4.19%
O	42.69%
N	1.1%
S	0.65%
P	0.02%

* % of a dry ash-free sample

Fifty milligrams of the Suwannee River NOM powder was dissolved in 500 mL of DI water to prepare a 100 mg/L stock solution of NOM. The pH of the solution was raised, by adding 40 mg of NaOH, and mixed for 2-3 hours to solubilize the NOM and form a visually clear solution. The solution was vacuum filtered using a 0.45 µm cellulose acetate filter and the filtrate obtained was stored as stock solution at 4°C. The pH of the stock NOM solution was about 10.4. Several dilutions of the stock solution were made, and the concentration of the DOC was measured using a Total Organic Carbon Analyzer (Model TOC-5050, Shimadzu Corporation). The average DOC concentration of the stock was 40 mg/L.

4.3.10. Procedure for chemical coagulation using ferrous iron in the presence of NOM

To study the influence of NOM on ferrous coagulation to remove MS2 viruses, synthetic water spiked with a known concentration of MS2 virus was coagulated with $FeSO_4$ (9-10 mg/L as Fe(II)) by adding varying concentrations of NOM (0-3 mg/L DOC). The following procedure was used: One liter aliquots of pH-adjusted synthetic water (pH 7.5) were poured into square jars, and dosed with 0-3 mg/L DOC. Samples were collected from each jar for DOC and UV-254 measurements. The water samples were spiked with a known concentration of

MS2. After this, we took additional samples for DOC and UV-254 measurements. This was necessary because the addition of MS2 caused an increase in the DOC concentrations due the organics present in the virus culture. Each jar was dosed with a relatively constant (9-10 mg/L) dose of ferrous iron and rapid mixed and flocculated as explained previously. Immediately after flocculation, the samples were filtered using 0.2 µm syringe filter. Filtrate samples were analyzed for viruses and LRV was calculated. Filtrate samples were also used for DOC and UV-254 measurements.

4.4. Results and Discussion

4.4.1. Chemical coagulation – Microfiltration

4.4.1.1. *MS2 removal by CC-MF using Fe(III) as the coagulant in synthetic water*

The results for the chemical coagulation – microfiltration (CC-MF) experiments conducted on MS2-spiked synthetic water using ferric chloride as the coagulant are presented here. It should be noted that the synthetic water is devoid of NOM and particulate matter. The feed MS2 concentrations for the spiked water were 10^5 and 10^7 PFU/mL for pH 7.5 and 6.4, respectively. The effects of the coagulant dose on the removal efficiency of MS2 from synthetic water by CC-MF at different pHs is shown in Figure 5-10 and Figure 5-11. The results show that chemical coagulation pretreatment to MF dramatically improved virus removal at both pH 6.4 and 7.5. As can be observed from the figures, without the coagulant and at 0 mg/L Fe(III) dose at both the pHs, MF alone achieved < 1 log removal, which increased slightly as more water was filtered. As coagulant dose increased, MS2 removal also increased, and > 4-log removal was achieved at pH 7.5 using a 10 mg/L Fe(III) dose. Virus log removals were much higher at pH 6.4, and with an initial virus concentration of 10^7 PFU/mL, > 7 log removal was achieved for 5 mg/L Fe(III) dose; number of countable plaques in the effluent was less than the detection limit.

These removals were comparatively higher than those observed by (Zhu et al. 2005b) who reported > 4-log removal at pH 6.3 for a higher Fe(III) dose of 10 mg/L. The possible reason for differences in our results is the variation in the initial virus concentration which was low (10^5 to 10^6 PFU/mL) in our experiments compared with 10^7 PFU/mL for (Zhu et al. 2005b). Due to low initial viral concentration, no viruses were detected in the effluent for high Fe(III) doses.

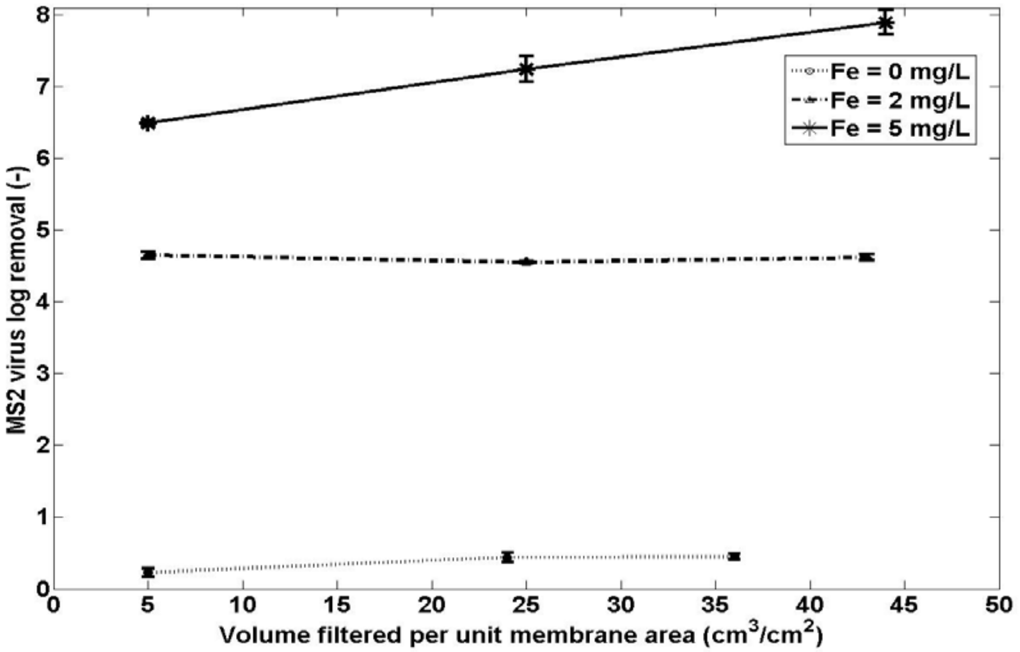


Figure 5-10.—Effect of Fe(III) dose and volume filtered on MS2 virus removal by CC-MF in synthetic water at pH 6.4, MS2 feed concentration = 9.8 x 107 PFU/mL.

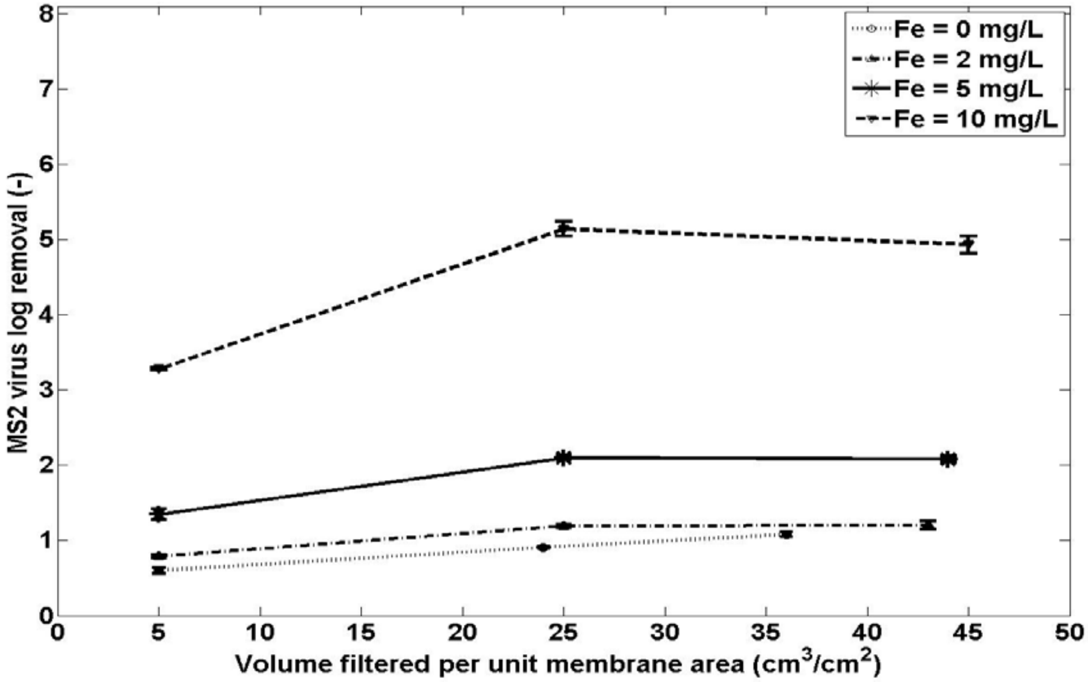


Figure 5-11.—Effect of Fe(III) dose and volume filtered on MS2 virus removal by CC-MF in synthetic water at pH 7.5, MS2 feed concentration = 8.6 x 105 PFU/mL.

4.4.1.2. *Effect of pH on MS2 removal by CC-MF using Fe(III) as a coagulant in synthetic water*

The effect of pH on MS2 removal by CC-MF using Fe(III) as a coagulant in synthetic water is shown in Figure 5-12 for a total volume of 20 mL filtered. The log removal of viruses increased with increasing iron dose and decreasing pH. For a given dose of Fe(III), more log removal of viruses was achieved at pH 6.4 than at pH 7.5. This is due to the increase in the positive surface charge on iron floc particles at lower pH that will cause better adsorption of viruses and their subsequent removal. With no coagulant, log removal of viruses was not influenced by pH. This is expected based on the consistent negative charges on both membrane and MS2 in this pH range. Similar observations were made for 100 mL and 175 mL filtered as shown in Figure 5-13 and Figure 5-14 respectively.

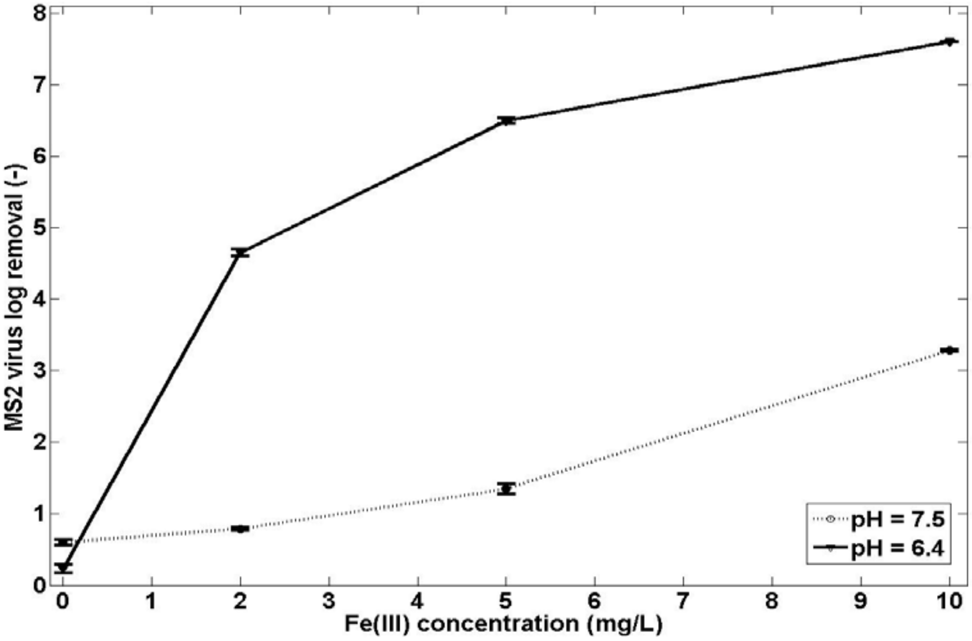


Figure 5-12. Effect of pH and iron dose on MS2 virus removal by CC-MF using Fe(III) as a coagulant in synthetic water after 20 mL of volume filtered.

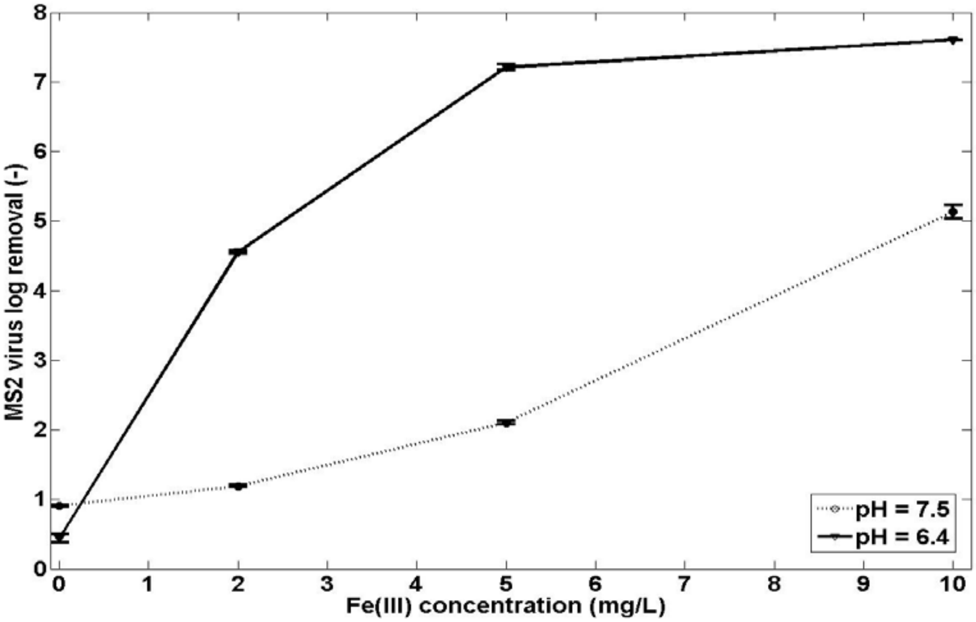


Figure 5-13.—Effect of pH and iron dose on MS2 virus removal by CC-MF using Fe(III) as a coagulant in synthetic water after 100 mL of volume filtered.

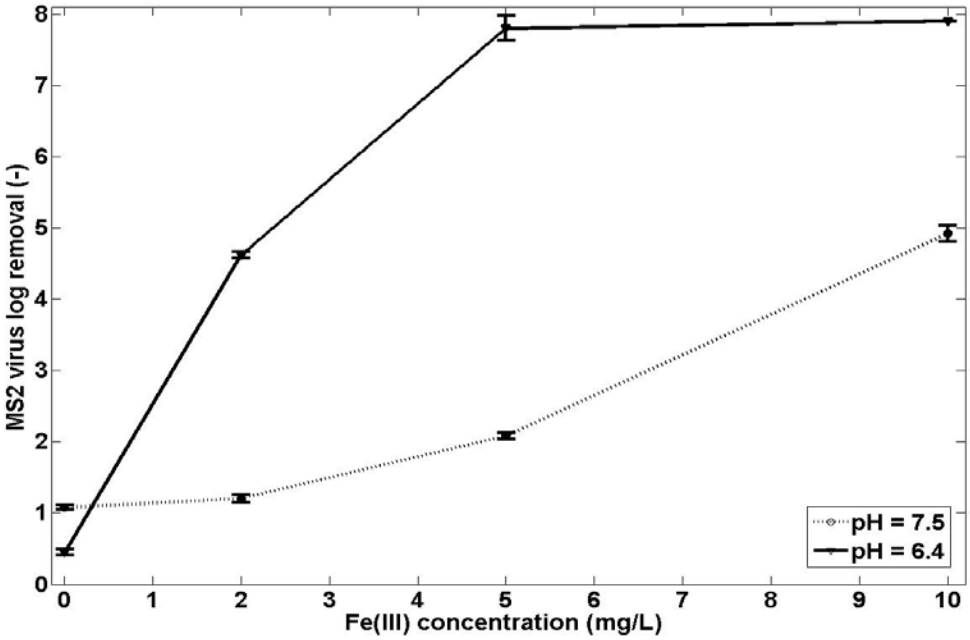


Figure 5-14.—Effect of pH and iron dose on MS2 virus removal by CC-MF using Fe(III) as a coagulant in synthetic water after 175 mL of volume filtered.

4.4.1.3. *Effect of volume filtered on MS2 removal by CC-MF using Fe(III) as a coagulant in synthetic water*

The effect of volume filtered on MS2 removal by CC-MF in synthetic water using Fe(III) as a coagulant is presented here. The feed MS2 concentrations for the spiked water were 10^6 PFU/mL for both pH 6.4 and 7.5, respectively. The effect of volume filtered at various coagulant dosages, on the removal efficiency of MS2 from synthetic water by CC-MF at different pHs is shown in Figure 5-15 and Figure 5-16 respectively. It can be seen that a negligible impact of volume filtered was observed at both the pHs studied. The log MS2 removal remained almost constant during filtration at a fixed iron dose even as cake-layer thickness increased during the filtration as more volume of water was filtered. Hence virus removal did not improve as more volume was filtered. Similar results were reported by (Zhu et al. 2005b), who had observed constant log removal of viruses with increase in cake layer thickness.

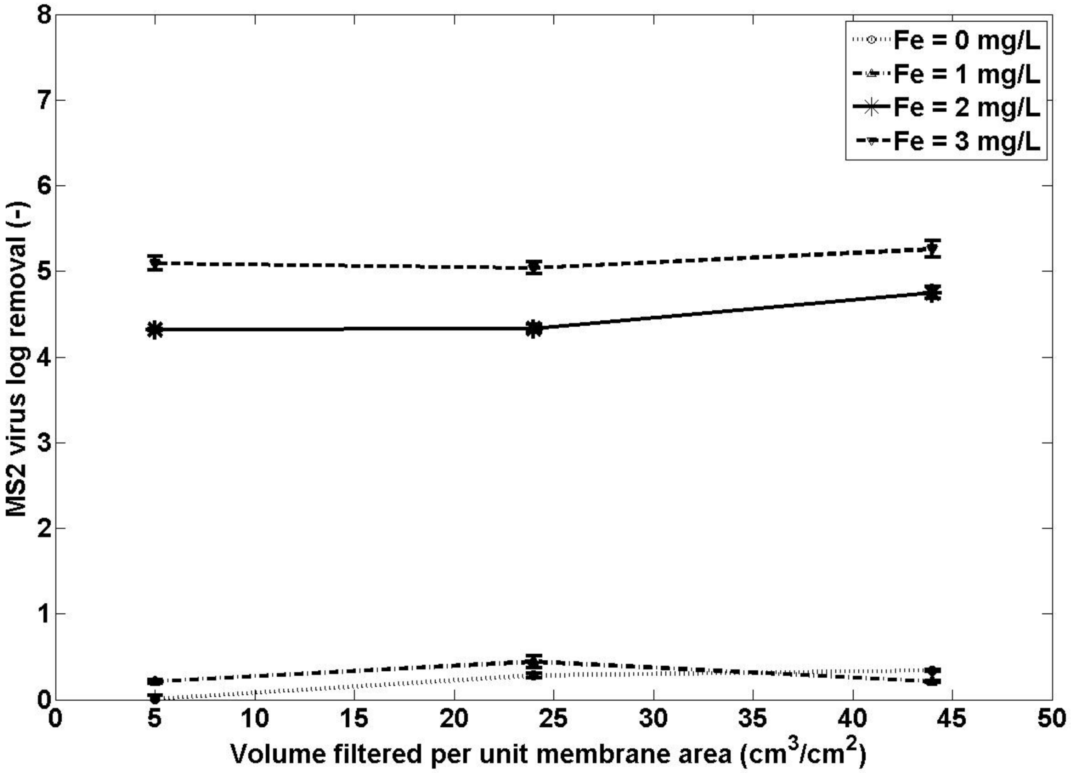


Figure 5-15.—Effect of volume filtered on MS2 virus removal by CC-MF in synthetic water at pH 6.4, MS2 feed concentration = 3.5 x 106 PFU/mL.

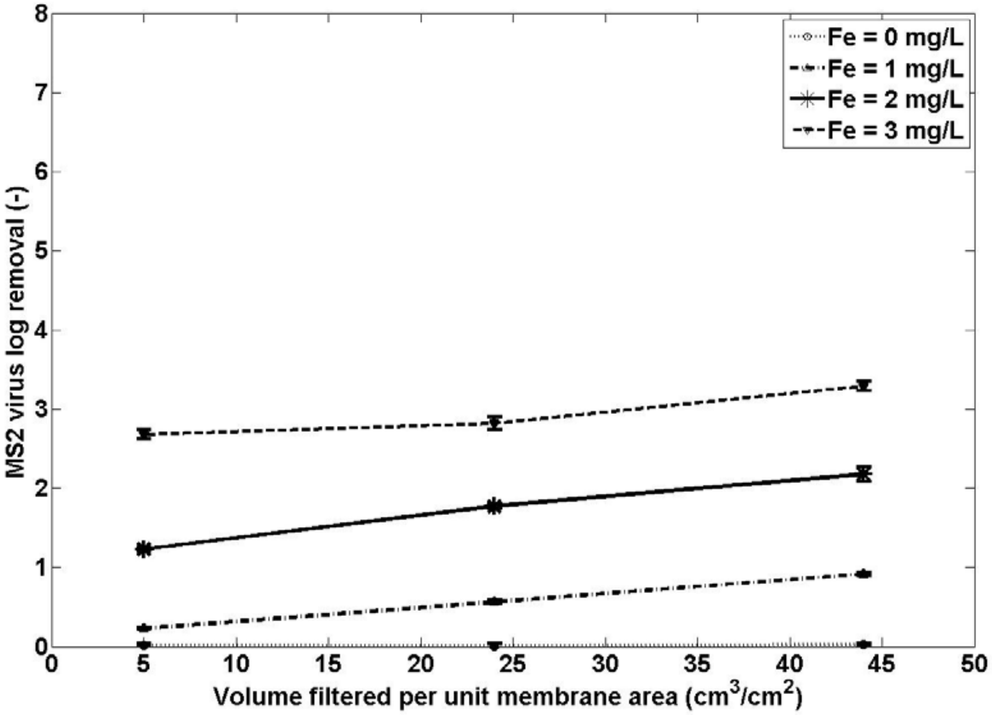


Figure 5-16.—Effect of volume filtered on MS2 virus removal by CC-MF in synthetic water at pH 7.5, MS2 feed concentration = 1 x 106 PFU/mL.

4.4.1.4. *MS2 removal by CC-MF using Fe(III) as the coagulant in lake water*

The results for the CC-MF experiments conducted on MS2-spiked lake water containing NOM, turbidity and particulates are presented here. The target MS2 feed concentration for the spiked water was 10^6 PFU/mL. The effect of the Fe(III) dose on the removal efficiency of MS2 by CC-MF at different pHs is shown in Figure 5-17, Figure 5-18 and Figure 5-19. As observed previously in synthetic water, coagulation pretreatment with Fe(III) was also effective for virus removal in lake water. With no coagulant (0 ppm Fe(III) dose) added, MF alone achieved < 1 log removal, which seemed to increase slightly as more volume was filtered. At all the three pHs of 6.4, 7.5 and 8.3, an increase in coagulant dose caused an increase in the log removal of viruses and > 5-log removal was achieved at pH = 6.4 using a 5mg/L Fe(III) dose.

The results for the effect of volume filtered on MS2 removal from lake water were slightly different from observed results for synthetic water (Figure 5-15 and Figure 5-16). As shown in Figure 5-17, Figure 5-18 and Figure 5-19, a steady increase in log removal for lake water was observed as more volume was filtered at all Fe(III) dosages and pHs studied. This is believed to be due to the presence of NOM in lake water that causes considerable amount of membrane fouling resulting in clogging of the filter pores (due to the formation of cake), thereby improving the removal of viruses.

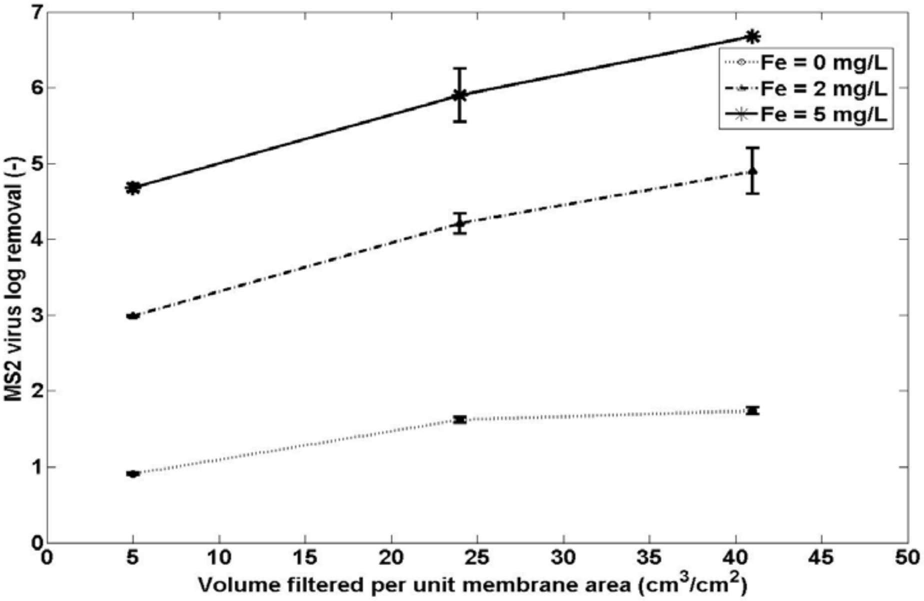


Figure 5-17.—Effect of Fe(III) dose and volume filtered on MS2 virus removal by CC-MF in lake water at pH 6.4, MS2 feed concentration = 1.6 x 106 PFU/mL.

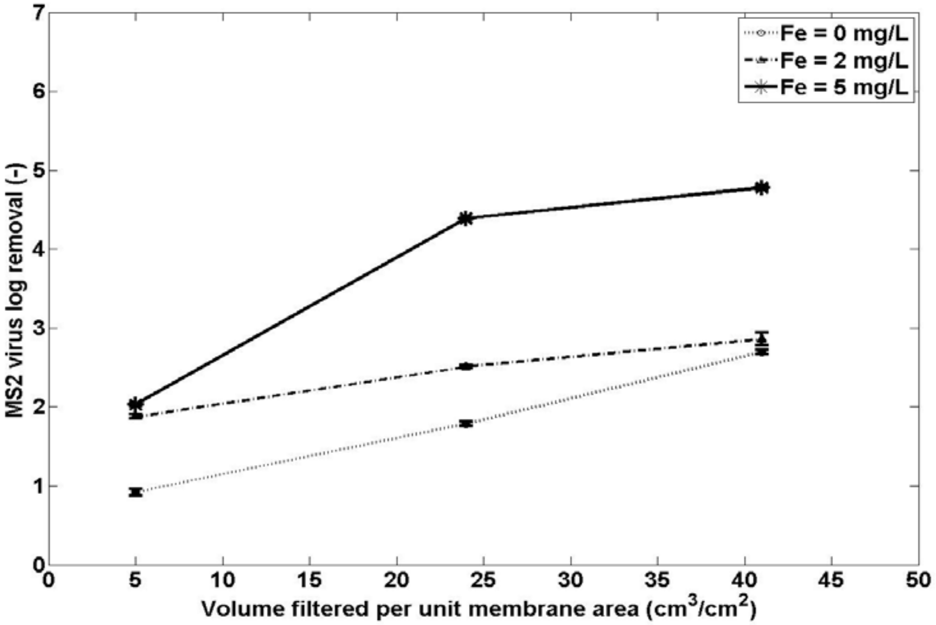


Figure 5-18.—Effect of Fe(III) dose and volume filtered on MS2 virus removal by CC-MF in lake water at pH 7.5, MS2 feed concentration = 3.2 x 106 PFU/mL.

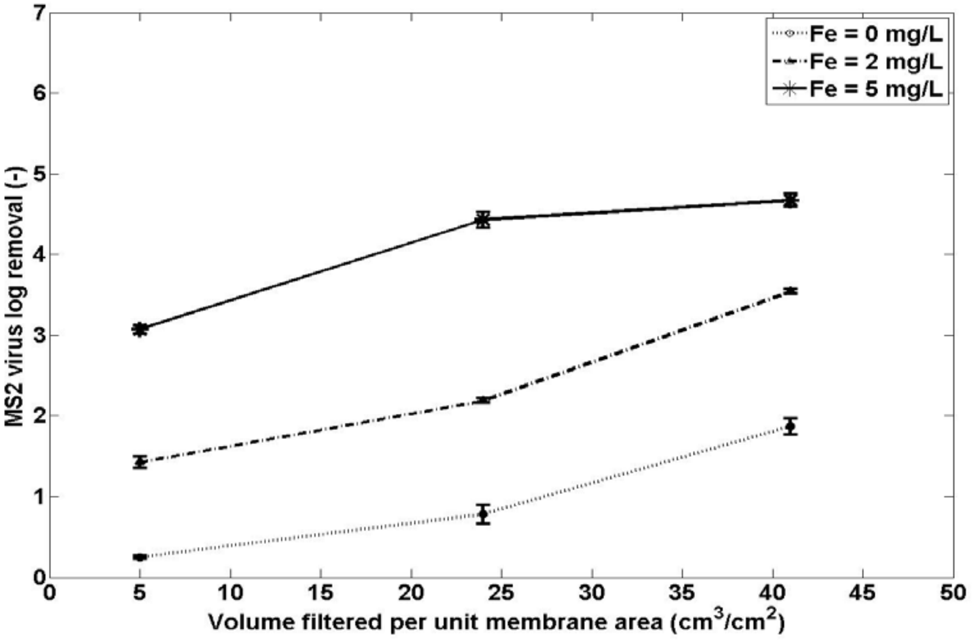


Figure 5-19.—Effect of Fe(III) dose and volume filtered on MS2 virus removal by CC-MF in lake water at pH 8.3, MS2 feed concentration = 1.5 x 106 PFU/mL.

4.4.1.5. *Effect of pH on MS2 removal by CC-MF using Fe(III) as a coagulant in lake water.*

The effect of pH on MS2 removal by CC-MF in lake water is presented here. . Figure 5-20, Figure 5-21, and Figure 5-22 depict the results for total volumes of 20, 100, and 170 mL filtered respectively. Similar to synthetic water, log removal of viruses in lake water generally increased as the pH decreased. As explained before, this is due to the increase in concentration of positively charged iron species at lower pHs, which will result in more virus removal. Better virus removal was achieved at pH 6.4 compared with pH 7.5 and 8.3. However, the log removals at pH 7.5 and 8.3 were not substantially different.

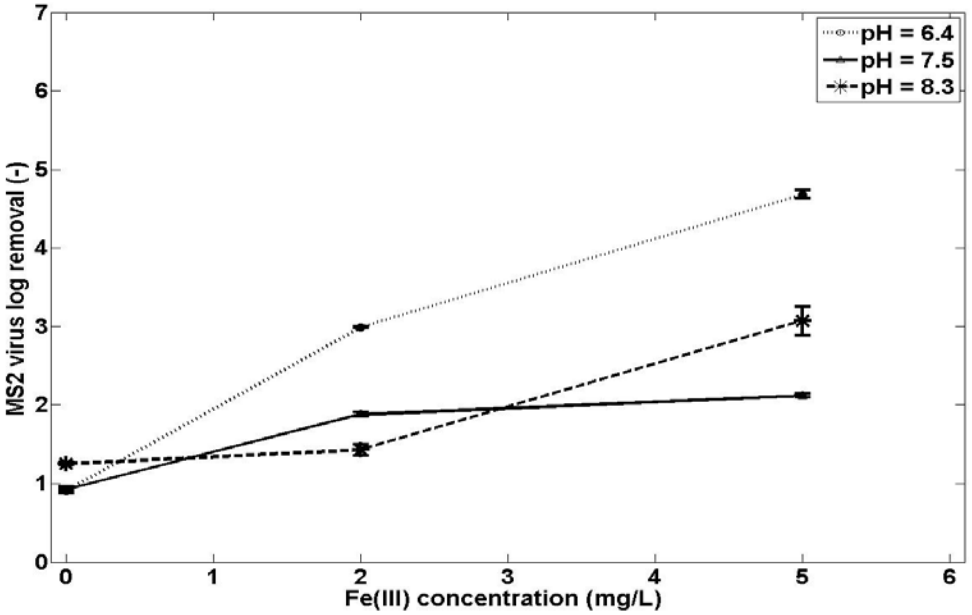


Figure 5-20.—Effect of pH and iron dose on MS2 virus removal by CC-MF using Fe(III) as a coagulant in lake water after 20 mL of volume was filtered.

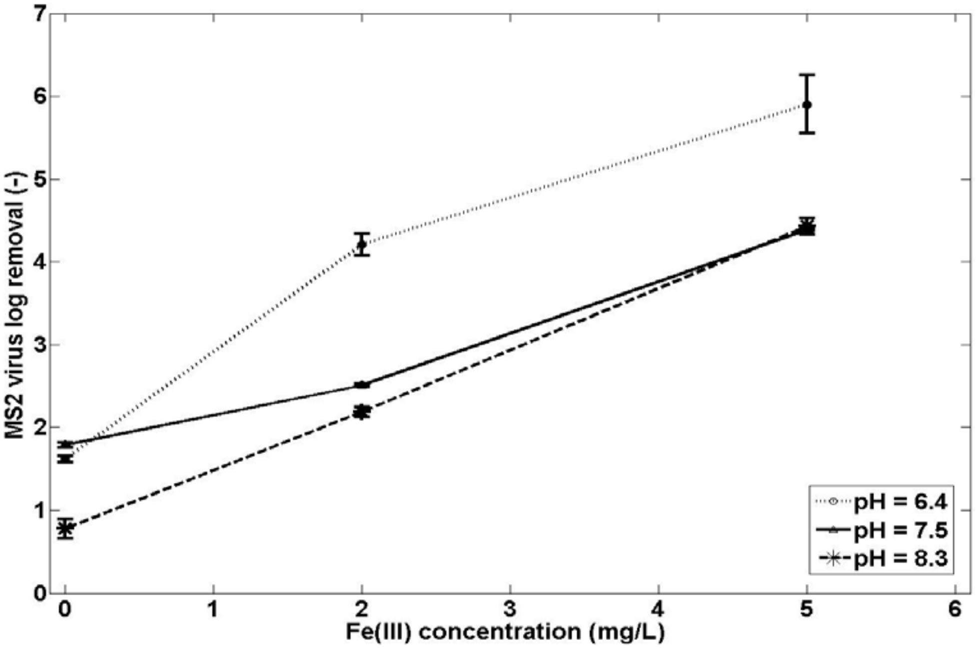


Figure 5-21.—Effect of pH and iron dose on MS2 virus removal by CC-MF using Fe(III) as a coagulant in lake water after 100 mL of volume was filtered.

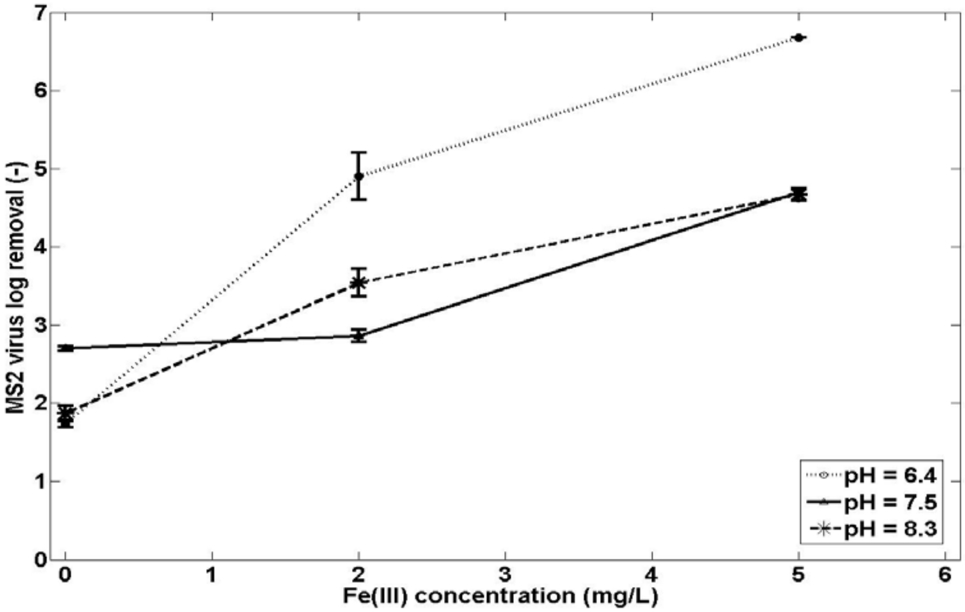


Figure 5-22.—Effect of pH and iron dose on MS2 virus removal by CC-MF using Fe(III) as a coagulant in lake water after 175 mL of volume was filtered.

4.4.1.6. *Mechanism of MS2 removal by CC-MF*

The average size of MS2 bacteriophage is 25 nm, which is about $^{1}/_{10}{}^{th}$ the nominal pore size (0.2 µm) of the MF membrane. Therefore, sieving during microfiltration without coagulant was not expected to remove significant amounts of MS2. Also, little MS2 removal by adsorption was expected because the viruses and the membrane surface are negatively charged at pHs above the isoelectric point (IEP)_ (pH_{IEP}) of 3.9 (Valegård et al. 1990). Accordingly, MS2 removal during CC-MF in both synthetic water and lake water was attributed to adsorption of negatively charged virus particles to the positively charged flocculated iron particles (pH_{IEP} = 8.2 for $Fe(OH)_3(s)$). The adsorption of the viruses increased as iron coagulant dose increased and hence more viruses were adsorbed onto the iron floc particles that were removed in the MF stage. Therefore, greater log virus removal was achieved with increasing iron coagulant dose. A similar explanation was given by (Zhu et al. 2005b) for virus removal by chemical coagulation-microfiltration in synthetic water.

4.4.2. Electrocoagulation – Microfiltration

4.4.2.1. *Operation of EC unit*

The operational efficiency of the EC unit was established in constant-current, variable-generation-time experiments using the water samples without virus spiking. As explained previously in §5.2, the theoretical amount of iron generated was calculated using Faraday's law as shown in Equation 1. A constant operating current of 300 mA was used in all the experiments, and the electrolysis time was varied to generate the required amount of iron.

4.4.2.2. *Fe(II) generation in the EC Unit*

As explained previously in §5.2, (Zhu et al. 2005a) had reported their studies on virus removal based on the assumption that Fe(III) was generated in the EC unit during electrocoagulation with iron anodes. However, recent research by (Lakshamanan 2007) has shown that EC actually generates Fe(II) in the pH range of 6.5-8.5. Due to the differences in these reports, it was necessary to independently determine the amount of Fe(II) generated in the EC unit in our experiments.

Figure 5-23 represents the % Fe(II) remaining after mixing for 2 minutes in the EC unit, as the mass of the iron generated in the EC unit increases with increasing generation time. The generation time for 1, 2, 5, 10, and 15 mg/L iron at constant current of 0.3 amps (A) was calculated using Faraday's law to be 4 seconds, 9 seconds, 22 seconds, 45 seconds, and 67 seconds respectively. As shown in the Figure 5-23, for 1.0 mg/L Fe dose about 70% of Fe(II) remained at pH 6.4, whereas only 11% remained at pH 7.5, i.e., the % Fe(II) remaining decreased with increasing mass of iron generated. These results are similar to those reported by (Lakshamanan 2007), who found that the decrease in % Fe(II) remaining with increasing iron generation was due to a local pH increase within EC unit with increasing iron generation and corresponding increase in the rate of Fe(II) oxidation. In these experiments, the reduction in the Fe(II) remaining at pH 7.5 (11%) as compared to pH 6.4 (70%) was also due to the increasing rate of Fe(II) oxidation to Fe(III) at increasing pH in aqueous solution (Stumm and Morgan 1996).

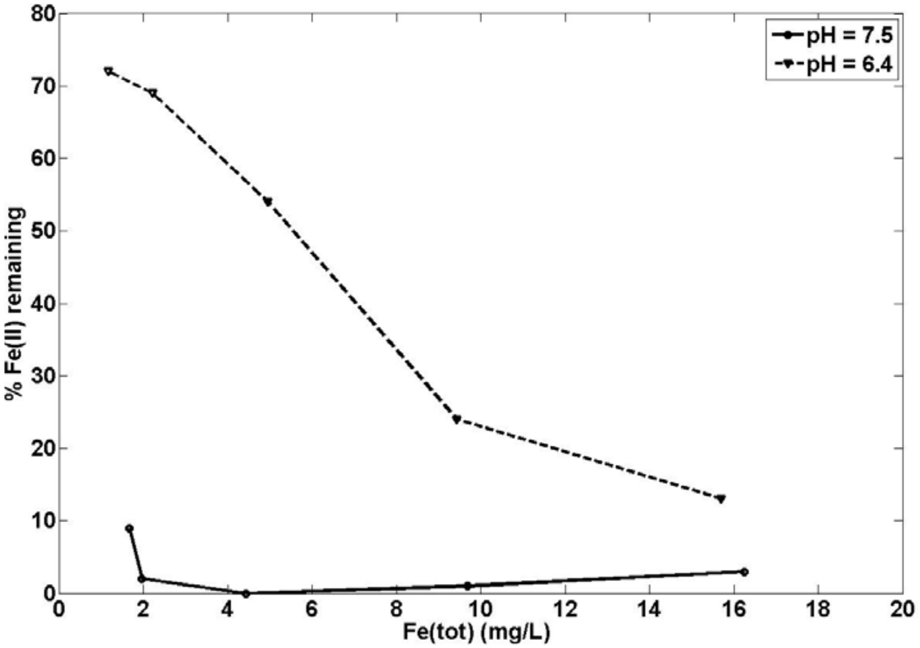


Figure 5-23.—Percent Fe(II) remaining in the EC unit as a function of Fe(Total) generated after mixing for 2 minutes at pH 6.4 and pH 7.5.

4.4.2.3. *Determination of Fe(II) or Fe(III) produced in the EC unit during deliberate rusting studies*

Since EC generated Fe(II) at all pHs studied, to further understand the iron generation mechanisms, a series of EC runs without any virus spiking and without cleaning and/or scrubbing the iron rods was conducted following each run.. These experiments were useful in determining if deliberate rusting of the rods helped in the oxidation of the soluble Fe(II) to insoluble Fe(III) that would be more effective in virus adsorption and removal. These experiments also closely simulated the previous work done on EC-MF using un-cleaned rods for virus removal from synthetic water (Zhu et al. 2005b). The EC unit was run at an operating current of 300 milliamps (mA) and generation time of 9 seconds, which was calculated to produce 2.0 mg/L Fe(III) in the effluent water. The samples were recirculated for 2 minutes, and the samples were analyzed for total iron and ferrous iron after this recirculation.

The results of iron speciation tests on the EC effluents after mixing for 2 minutes from 26 sequential runs at pH 6.4 are shown in Figure 5-24. It can be observed from the figure that even after 2 min of mixing in open atmospheric conditions, residual Fe(II) was very significant (32-64%) for all the runs. During the first seven runs, Fe(II) remaining decreased gradually from 62 to 39%, but then rose again to 53% by the ninth run. Considering all 26 runs, there was no clear trend in Fe(II) remaining after 2 min generation/mixing, which averaged about 50%.

Ten additional runs were performed at pH 7.5 and the speciation results are presented in Figure 5-25. Here again, there was no clear trend in Fe(II) remaining after 2 min of mixing, which averaged about 11% over the ten runs. As explained before, the decreasing trend in average Fe(II) production of 50% for pH 6.4 and 11% for pH 7.5 was expected based on the rate of Fe(II) oxidation to Fe(III), which increases as pH increases from 6.4-7.5 (Stumm and Morgan 1996).

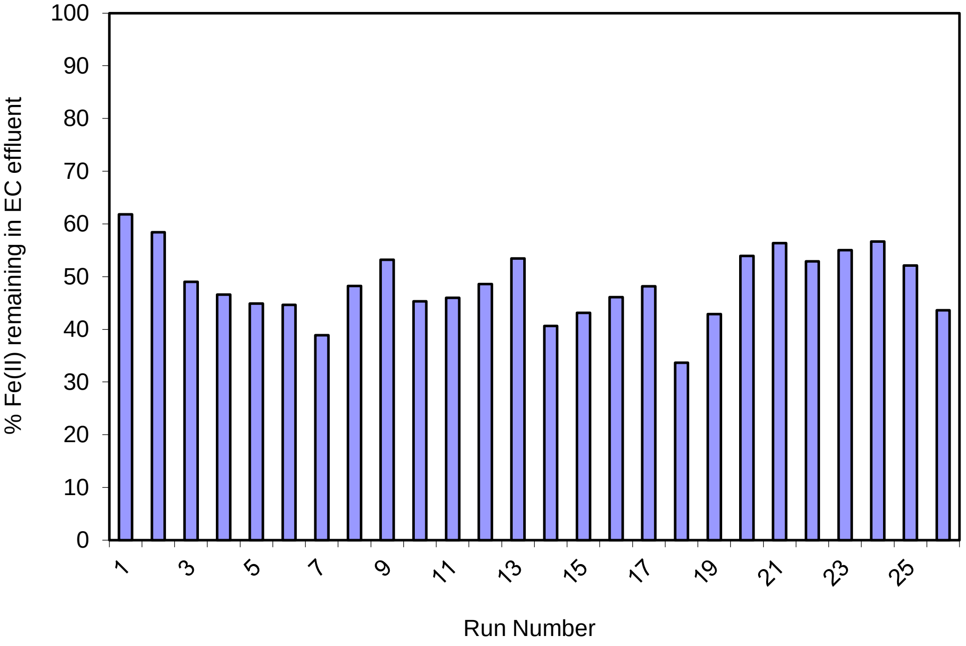


Figure 5-24.—Percentage of Fe(II) in the EC unit effluent after 2 minutes of generation/mixing at pH 6.4, operated in batch mode at current 0.3A with a generation time of 9 seconds.

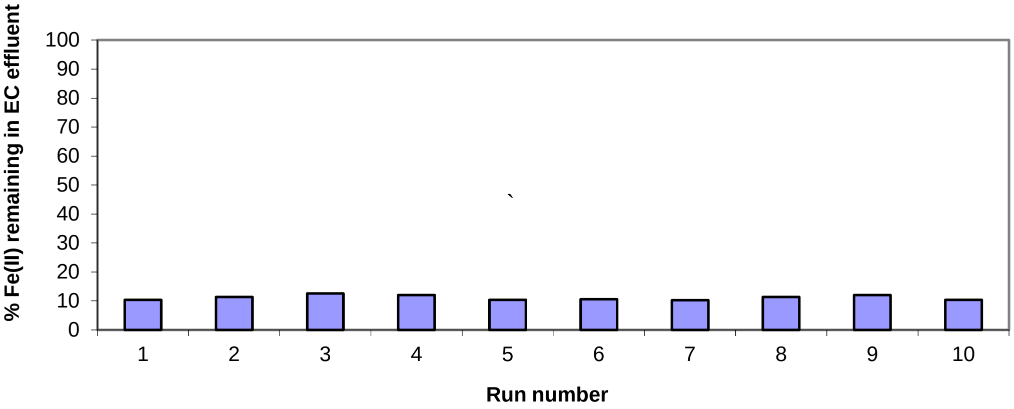


Figure 5-25.—Percentage of Fe(II) in the EC unit effluent after 2 minutes of generation/mixing at pH 7.5, operated in batch mode at current 0.3A with a generation time of 9 seconds.

4.4.2.4. *MS2 removal by electrocoagulation-syringe filtration (EC-SF) in lake water*

The results for the preliminary electrocoagulation-syringe microfiltration (EC-SF) experiments conducted on MS2-spiked lake water are presented here. They are considered screening experiments because the MF step was simplified by using 0.2 µm syringe filters instead of membrane filters to get a preliminary indication of virus removal by EC-MF in less time than it takes to filter large volumes of up to 175 mL of electrocoagulated water. The feed MS2 concentration for the spiked samples was 10^6 PFU/mL. The effect of the iron on the removal efficiency of MS2 by EC-SF at pH 7.5 is shown in Figure 5-26. As the results indicate, EC pretreatment (which generates ferrous, not ferric, iron) did not significantly improve virus removal at pH 7.5. A small increase in virus log removal was observed as iron dose increased from 1.0 to 15.0 mg/L. Less than 0.5 log removal was achieved for 15.0 mg/L, the highest dose of iron generated. In contrast, (Zhu et al. 2005b) had reported up to 5-log removal of MS2 at pH 7.3 and 10.0 mg/L iron dose by EC-MF in synthetic water without NOM.

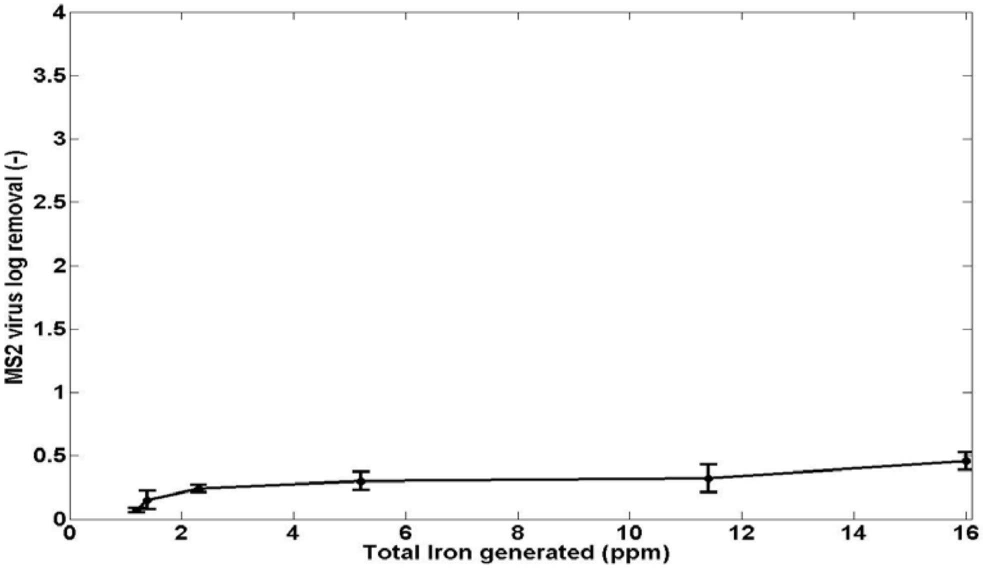


Figure 5-26.—Effect of iron generated on MS2 virus removal by EC-SF in lake water at pH 7.5, MS2 feed concentration = 4 x 106 PFU/mL.

4.4.2.5. *Mechanism of MS2 removal in lake water by EC-SF*

EC was expected to achieve effective virus removals in lake water, based on literature reports showing superior performance of EC compared with CC in removal of viruses from synthetic water (Zhu et al. 2005a). The reason for the differences in the results was probably due the significant differences in the background water and experimental conditions between their work and our work.

(Zhu et al. 2005a) had reported such high virus removals using EC when filtering the coagulated samples using MF for long durations (few hours to 1 day). Since EC primarily generates Fe(II), long hours of filtration provided sufficient time for the effective oxidation of Fe(II) to Fe(III) that could subsequently hydrolyze to form hydroxide precipitates to remove the viruses. In contrast, in the EC experiments conducted in this research (Figure 5-26), the samples were filtered immediately (within 5 minutes) after coagulation using 0.2 µm syringe microfilter. It is suggested that the drastic reduction in the filtration time and the difference in the background water resulted in ineffective oxidation of Fe(II) and its subsequent hydrolysis—thus causing poor virus removal.

Table 5-3 represents the total iron measured in the EC effluent before and after 0.2 µm syringe microfiltration at pH 6.4. The data shows that a significant amount of soluble iron, presumably Fe(II), was generated at the lower iron doses which did not cause any virus removal. However, less soluble iron (2-12%) was observed at higher iron dosages and hence should have resulted in good virus removal. Unexpectedly, low virus log removals were observed at high iron dosages also. Since the lake water contained a significant amount of NOM (~ 5 mg/L DOC), it was suspected that the presence of NOM had a detrimental effect on Fe(II) oxidation causing poor removal of viruses. Further experiments (Suwannee River) were conducted to study the effect of NOM on virus removal using EC, which is explained later in §5.4.2.13.

Table 5-3.—Iron in EC Effluent at pH 6.4 before and after 0.2 µm syringe filtration.

Fe(Tot) (ppm)		
Before filtration	**After filtration**	**% Fe in filtrate**
1.46	0.93	64%
2.70	0.85	31%
5.54	2.06	37%
10.19	1.23	12%
16.87	0.38	2%

Because of the poor performance of the EC in these preliminary experiments with lake water, it was necessary to verify the performance of EC-MF in synthetic water, which is devoid of NOM, to compare these results and those that Zhu et al. (2005a) reported. Therefore, we also conducted EC-MF experiments in synthetic water as a function of pH and iron dose.

4.4.2.6. *MS2 removal by EC-MF in synthetic water*

As can be observed from the following experimental results for EC-MF in synthetic water without NOM or turbidity, electrocoagulation produced a dramatic improvement in virus removal, which increased with increasing iron doses. The spiked MS2 concentration was 10^7 PFU/mL. Iron doses generated ranged from 0 to 13 mg/L as Fe, and sample pHs were pre-adjusted to 7.5 or 6.4. The effects of the coagulant dose and pH on EC-MF's removal efficiency of MS2 are shown in Figure 5-27 and Figure 5-28. The results show that EC pretreatment dramatically improved virus removal at both pHs. As observed with CC pretreatment, MF alone achieved less than 1 log removal, which appeared to increase slightly as more volume was filtered. As the iron concentration increased, the virus removal also increased, and > 6-log removal was achieved at 12.8 mg/L iron dose at pH 6.4. It should be noted that at high iron dosage of 12.8 mg/L, the virus log removals were close to 7-log removal for 100 mL and 175 mL volume filtered samples even when starting with an initial virus concentration of 10^7 PFU/mL. This is primarily because, the coagulated-microfiltered samples had very few countable plaques that was less than the allowable minimum detection limit (30 plaques per plate) i.e., a plaque assay of three samples of a single dilution resulted in one plaque in one plate, two plaques in another and one plaque in the third plate resulting in an average virus concentration of 1.3PFU/mL, which resulted in 7.9-log removal of viruses.

These results were similar to reported results on virus-removal using the same synthetic water, which showed up to five-log removal of MS2 at pH 6.3 and 10 mg/L iron dose (Zhu et al. 2005a). It should be noted that Zhu et al. (2005a) achieved such high virus log removal using EC in spite of Fe(II) being generated, because of the oxidation of Fe(II) to Fe(III) during the several hours of standing and/or continuous mixing prior to microfiltration. The Fe(III) formed could hydrolyze to form the hydroxide coagulant species that adsorbed the viruses and was removed effectively by the microfilter.

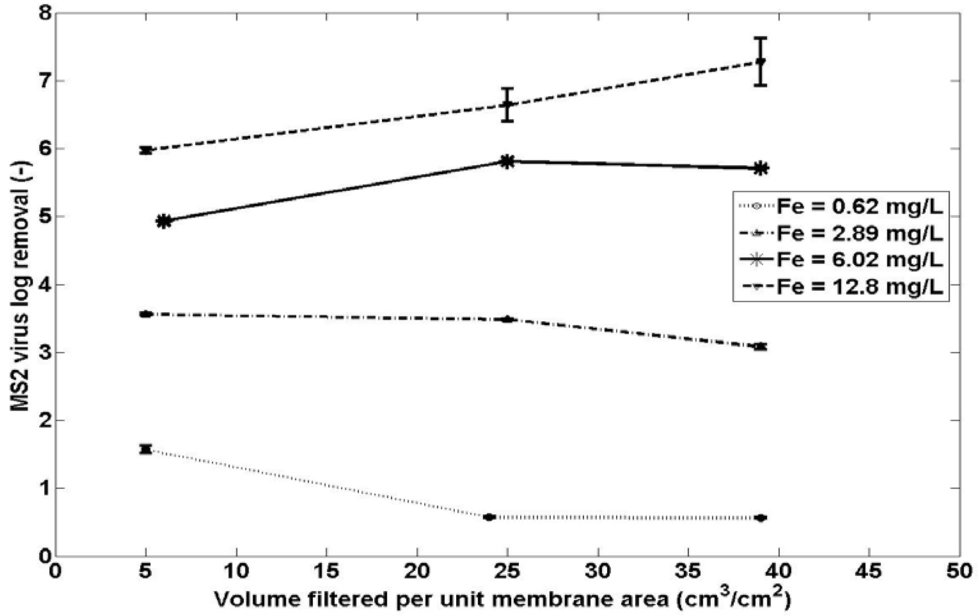


Figure 5-27.—Effect of iron generated and volume filtered on MS2 removal in synthetic water by EC-MF at pH 6.4. MS2 feed concentration = 4*107 PFU/mL.

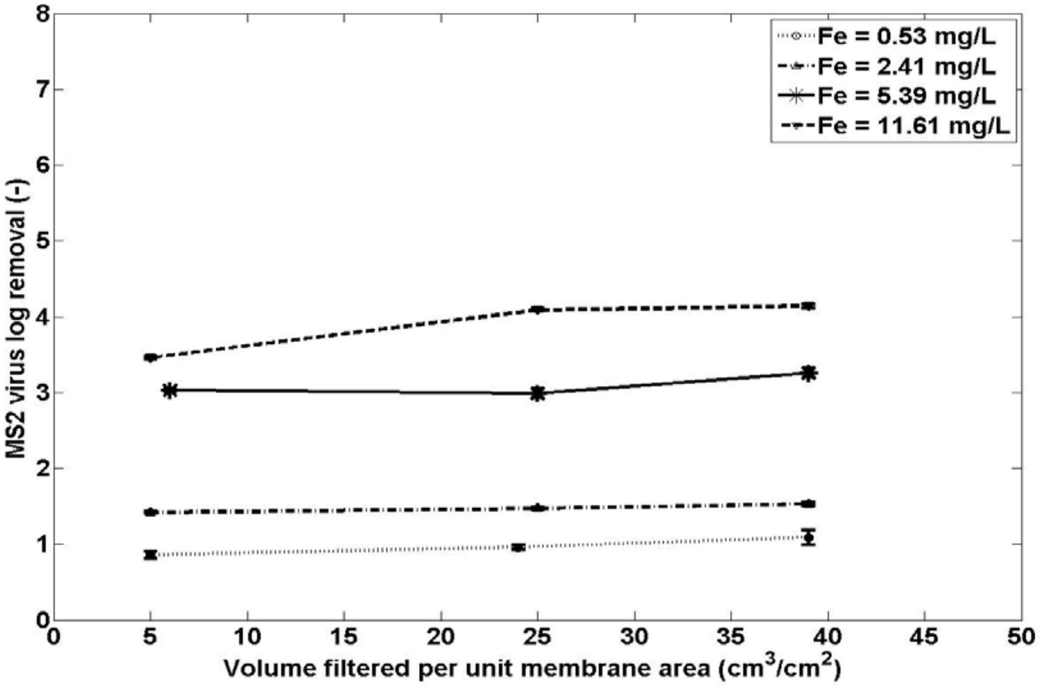


Figure 5-28.—Effect of iron generated and volume filtered on MS2 removal in synthetic water by EC-MF at pH 7.5. MS2 feed concentration = 3.93*107 PFU/mL.

4.4.2.7. *Effect of pH on MS2 removal from synthetic water using EC-MF*

Figure 5-29 shows the effect of pH on virus removal from synthetic water using EC-MF after 20 mL volume filtered. It is clear that virus log removal increased with increasing iron dose and decreasing pH. Similar observations were made for 100 mL and 175 mL volumes filtered as shown in Figure 5-30 and Figure 5-31 respectively. With no coagulant, virus log removal was not influenced by pH 6.4 or 7.5, which is expected based on the consistent negative charge on both the membrane and the MS2 virus in this pH range. However, for increasing iron doses, pH 6.4 produced nearly 2 log greater virus removal than pH 7.5. These results are not in agreement with reported results (Zhu et al. 2005a), which demonstrated that in the pH range of 6.3 to 8.3, pH had little effect on MS2 removal by EC-MF. The disagreement in the observed results must be due to the difference in standing and/or filtration time in these experiments, i.e. the time between iron generation stage during electrocoagulation and the filtration stage. As explained in §5.2, (Zhu et al. 2005a) had achieved high virus removal based on the assumption that Fe(III) is being generated in the EC unit. Based on their results, this research also started with the same assumption. However, as explained in §5.2, recent research by (Lakshamanan 2007) had reported the generation of Fe(II) in the EC unit, and its oxidation largely depended on the pH of the sample and time provided for oxidation. Therefore, providing long hours of standing and/or filtration will allow variable oxidation of Fe(II) to Fe(III) depending on the pH. However, both in this research and in studies reported by (Zhu et al. 2005a), the standing and filtration time of the electrocoagulated samples were not controlled, as it was not expected to affect the virus removal results. Therefore, it is believed that the varying concentrations of Fe(III) produced by oxidation of Fe(II) caused variable virus removal at pH 6.4-7.5, causing the differences in our results from those reported by (Zhu et al. 2005a).

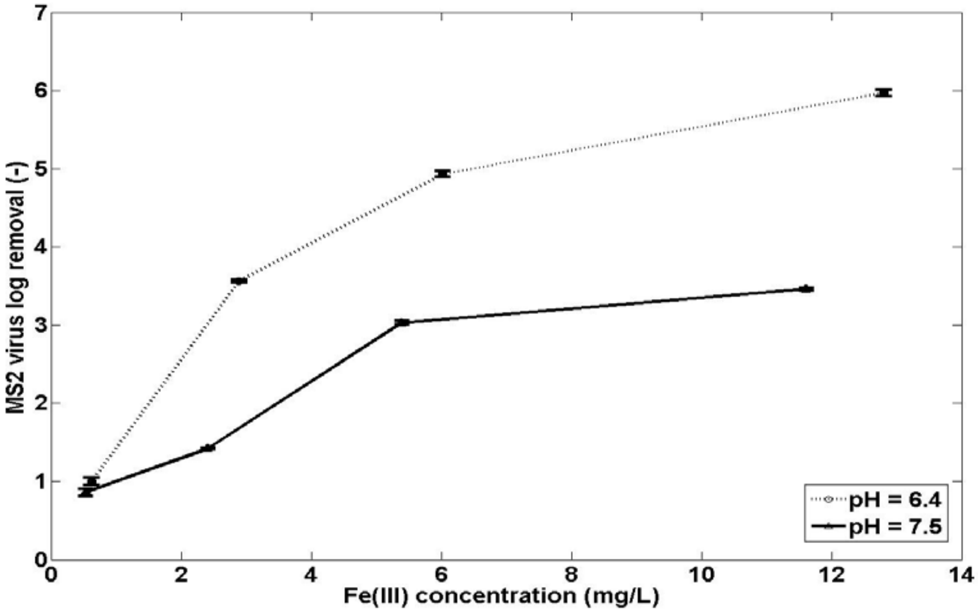


Figure 5-29.—Effect of iron dose and pH on MS2 removal from synthetic water by EC-MF after 20 mL of volume was filtered.

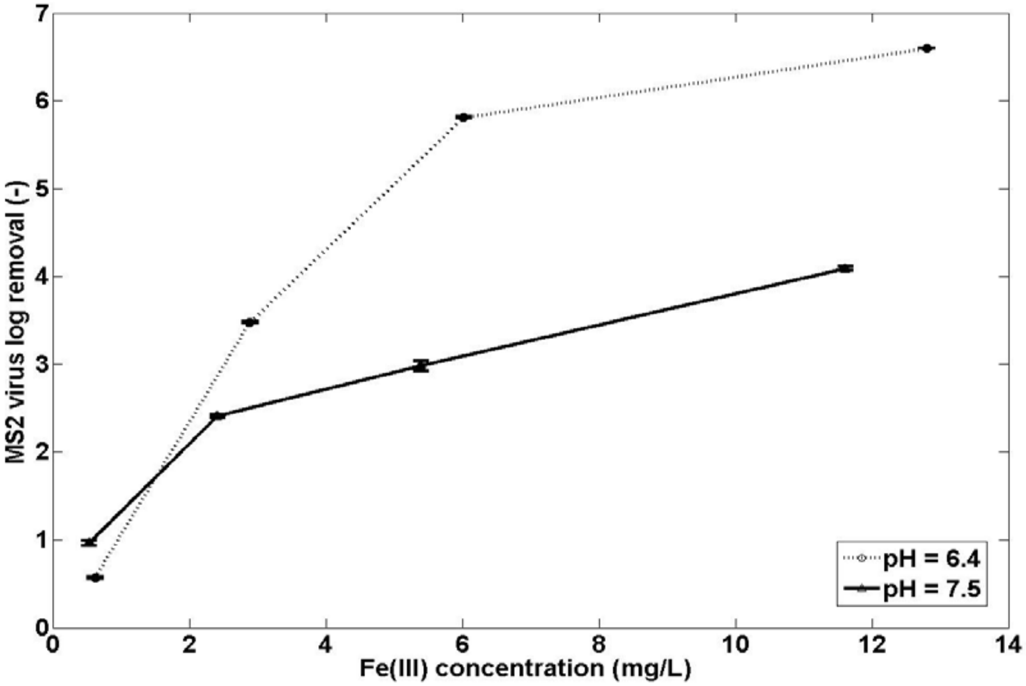


Figure 5-30.—Effect of iron dose and pH on MS2 removal from synthetic water by EC-MF after 100 mL of volume was filtered.

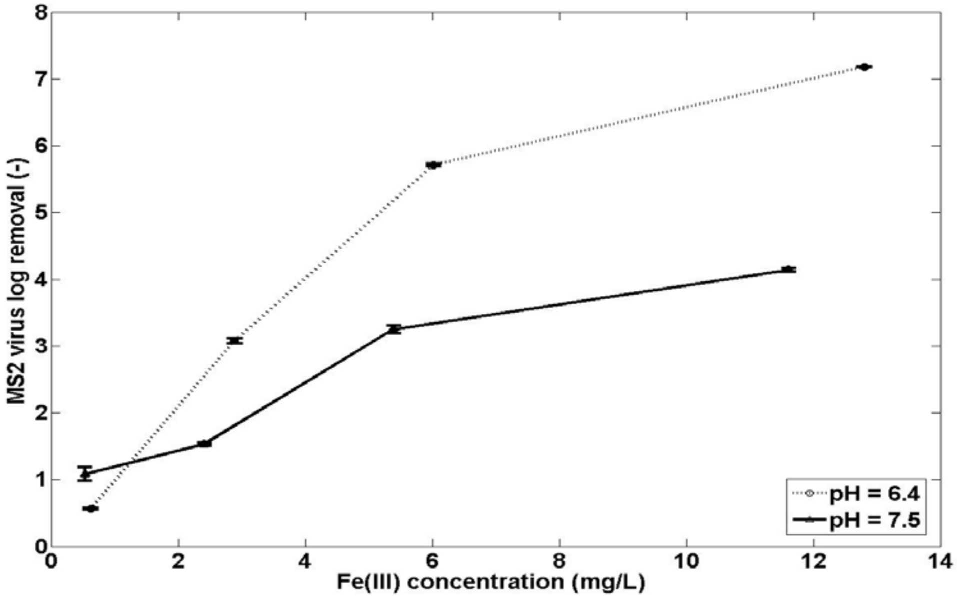


Figure 5-31.—Effect of iron dose and pH on MS2 removal from synthetic water by EC-MF after 175 mL of volume was filtered.

4.4.2.8. *MS2 removal by EC-MF in lake water with NOM*

As shown in Figure 5-32, Figure 5-33, and Figure 5-34, EC-MF did not improve virus removal in lake water. Less than 2-log removal was observed at all the pHs studied, even with iron concentrations as high as 13 mg/L. These results are quite different than the MS2 removals (typically 4-6 logs) observed for EC-MF for synthetic water. It is suspected that the poor performance of EC on lake water was due to the fact that EC generates Fe(II), which must be oxidized to Fe(III) before it can effectively act as a coagulant and adsorb the viruses prior to essentially complete removal by MF. The NOM present in lake water was also suspected to have a detrimental effect in EC's performance.

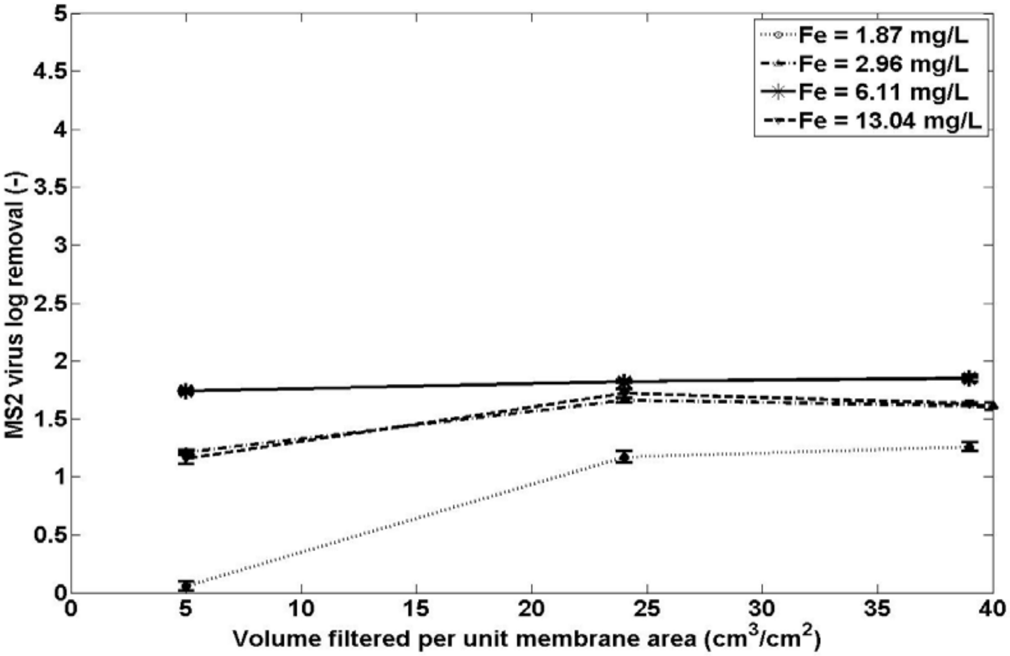


Figure 5-32.—Effect of iron generated and volume filtered on MS2 removal in lake water by EC-MF at pH 6.4, MS2 feed concentration = 5.73*106 PFU/mL.

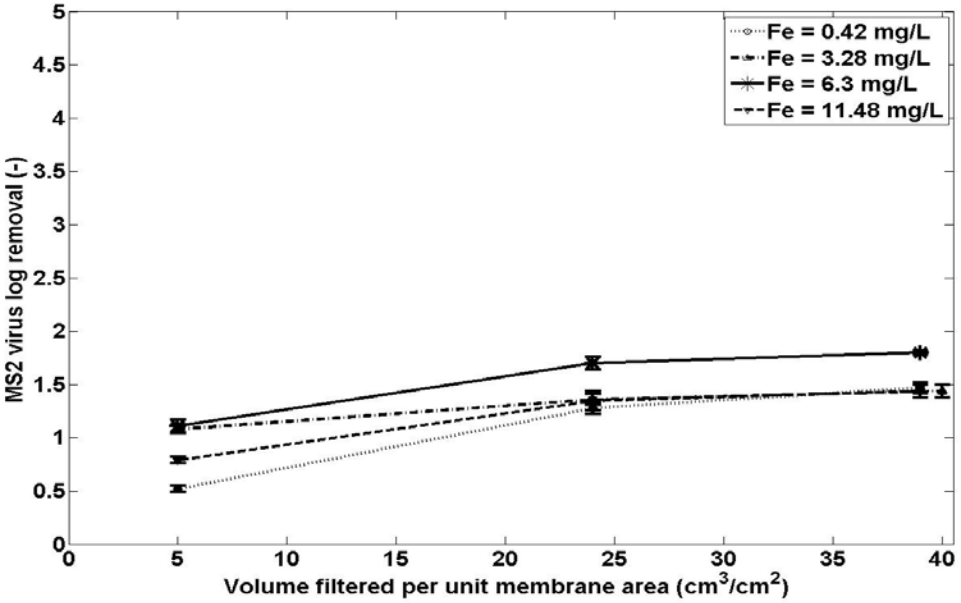


Figure 5-33.—Effect of iron generated and volume filtered on MS2 removal in lake water by EC-MF at pH 7.5, MS2 feed concentration = 8.4*106 PFU/mL.

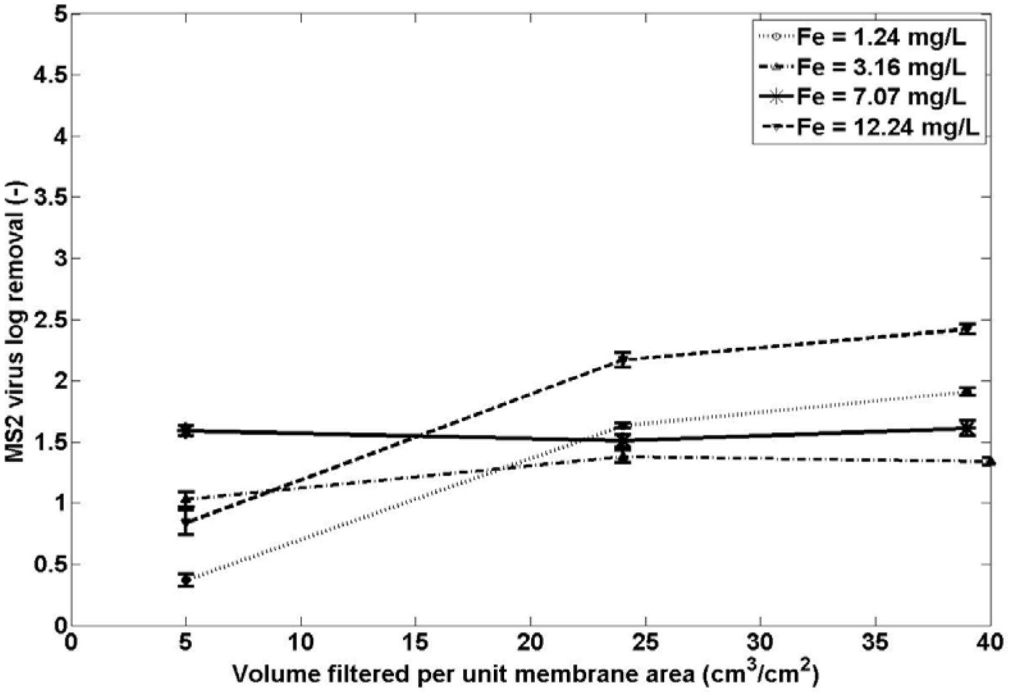


Figure 5-34.—Effect of iron generated and volume filtered on MS2 removal in lake water by EC-MF at pH 8.3, MS2 feed concentration = 2.6*106 PFU/mL.

4.4.2.9. *Comparison of MS2 removal by EC-MF in synthetic water without NOM and lake water with NOM*

To better understand EC-MF performance, the virus removal results were compared for synthetic water and lake water. The effects of the iron dose and pH on the removal efficiency of MS2 by EC-MF in synthetic and lake water are shown in Figure 5-35 and Figure 5-36 for pH 6.4 and 7.5, respectively. These results were shown in §5.4.2.8 in a different format, but are repeated here to clearly indicate the difference in the EC's performance for the two test waters. At pH 6.4, greater than 5-log removal was achieved at 12.8 mg/L iron generated in NOM-free synthetic water, whereas less than 2-log removal was achieved at 13.0 mg/L iron for lake water with 4 mg/L NOM. Similarly at pH 7.5, greater than 4-log removal was achieved at 11.6 mg/L iron for synthetic water, whereas less than 2-log removal was achieved at 11.5 mg/L iron in lake water.

Although the Fe(II) generated during electrocoagulation would pass through the microfilter without removing any viruses, EC-MF achieved 4-7 log virus removal in NOM-free synthetic water. This 4-7 log removal resulted from oxidation of Fe(II) to Fe(III) during the several hours of standing and/or continuous mixing prior to microfiltration. The Fe(II) oxidized and hydrolyzed to form $Fe(OH)_3(s)$, which adsorbed viruses and the iron floc particles were readily removed by the microfilter. However, similar high virus log removals were not observed in lake water—in spite of providing several hours of standing and mixing for Fe(II) oxidation.

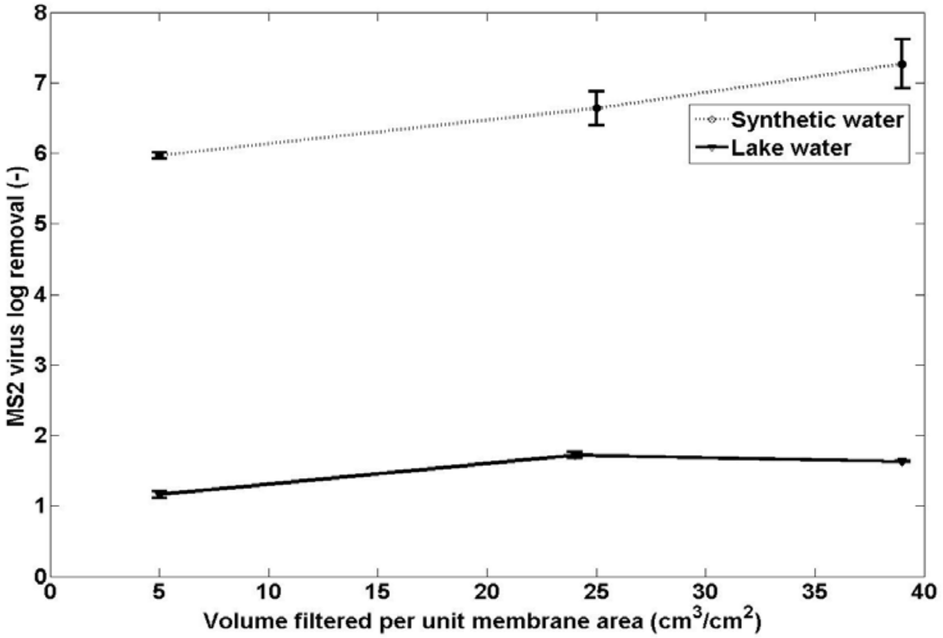


Figure 5-35.—Comparison of MS2 removal using EC-MF in synthetic water and lake water at pH 6.4 and nominal dose of 13 mg/L iron.

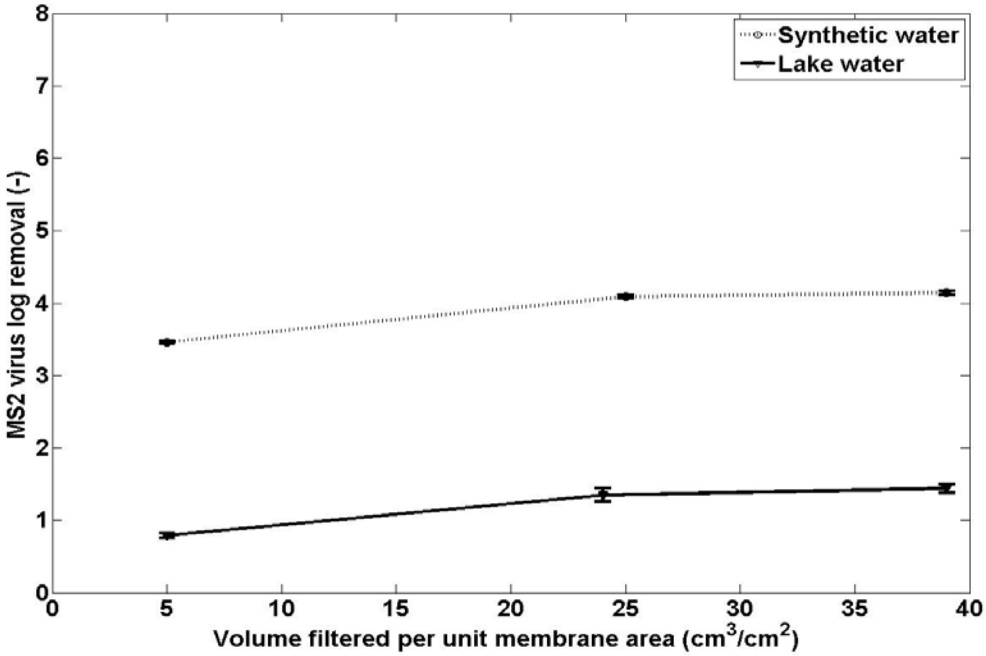


Figure 5-36.—Comparison of MS2 removal using EC-MF in synthetic water and lake water at pH 7.5 and nominal dose of 11.6 mg/L iron.

4.4.2.10. *Comparison of CC-MF and EC-MF for MS2 removal from synthetic water and lake water*

Figure 5-37 and Figure 5-38 depict the comparison of CC-MF and EC-MF for MS2 removal from synthetic water and lake water. As can be seen from Figure 5-37, both CC-MF and EC-MF were equally efficient in removing viruses from synthetic water at pH 6.4 and pH 7.5. For a nominal dose of 5.5 mg/L at pH 6.4, CC-MF achieved greater than 7-log removal and EC-MF achieved close to 6-log removal. Similarly, at pH 7.5 for an average iron concentration of 5.2 mg/L, CC-MF achieved 2-log removal whereas EC-MF achieved 3-log removal of viruses.

In contrast to the similar process performance in synthetic water, EC-MF did not achieve any significant virus removal for lake water whereas CC-MF achieved 4-6 log removal of viruses (Figure 5-38). At both pHs, CC-MF outperformed EC-MF for virus removal. CC-MF achieved > 5-log removal at pH 6.4 and > 4-log removal at pH 7.5 whereas < 2-log removal was observed at both the pHs by EC-MF. It should be noted that the primary difference in these experiments was the species of iron used for coagulation/flocculation. In CC, Fe(III) was used throughout the experiment in EC, Fe(II) was generated that got oxidized to Fe(III) over time that largely depended on pH and dissolved oxygen concentration. To better understand EC's performance, Fe(II) chemical coagulation experiments were conducted, the results are discussed in the next section.

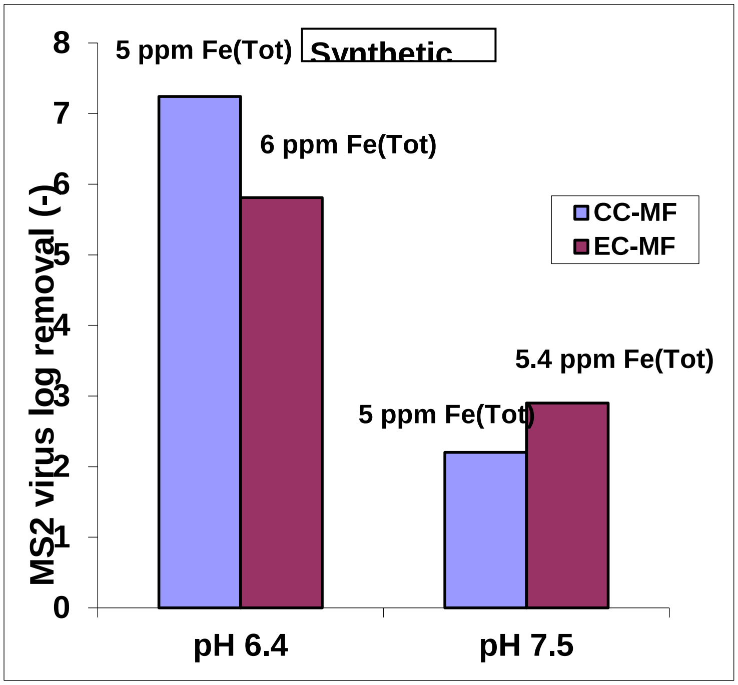


Figure 5-37.—Comparison of MS2 removal using CC-MF and EC-MF in synthetic water at pH 6.4 and pH 7.5.

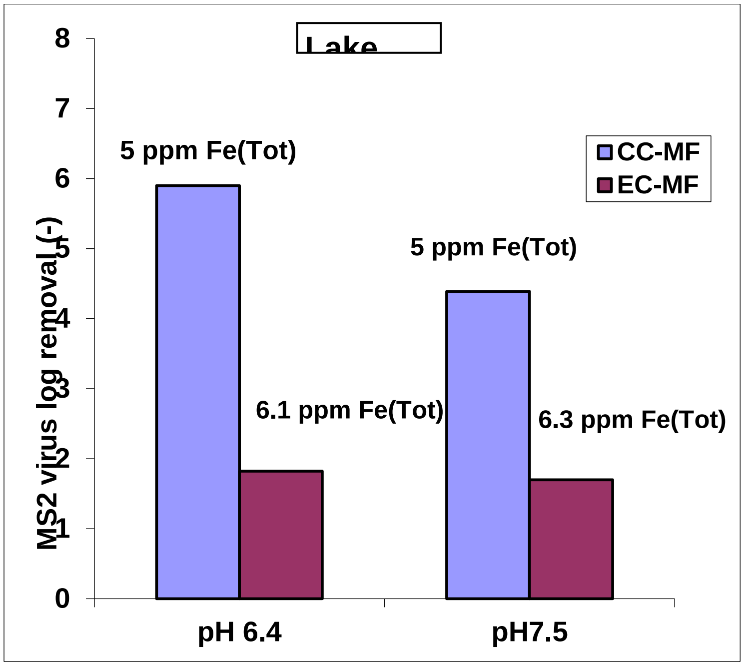


Figure 5-38.—Comparison of MS2 removal using CC-MF and EC-MF in lake water at pH 6.4 and 7.5.

4.4.2.11. *MS2 removal by chemical coagulation-syringe microfiltration (CC-SF) using Fe(II) as a coagulant*

As explained in the previous section, chemical coagulation experiments using $FeSO_4$ were conducted in synthetic water and lake water to chemically simulate the performance of electrocoagulation, because Fe(II) is the primary species being generated in the EC unit. The results representing the comparison of Fe(II) coagulation of synthetic water vs. lake water at pH 7.5 are presented in Figure 5-39. While up to 3.5 log removal was observed in NOM-free synthetic water at 10 mg/L Fe(II) dose, little improvement in the virus removal was observed even at such high Fe(II) doses in lake water with NOM. The 60-minute rapid flocculation period of the $FeSO_4$ coagulant during the experiment allowed sufficient time for Fe(II) to be oxidized to Fe(III) at pH 7.5 (Lakshamanan 2007) Thus, very good coagulation and virus removal were observed in synthetic water using Fe(II), because it gets effectively oxidized to Fe(III) and hydrolyzes to form $Fe(OH)_3(s)$ before microfiltration. However, a similar performance was not observed in lake water for all the Fe(II) doses considered.

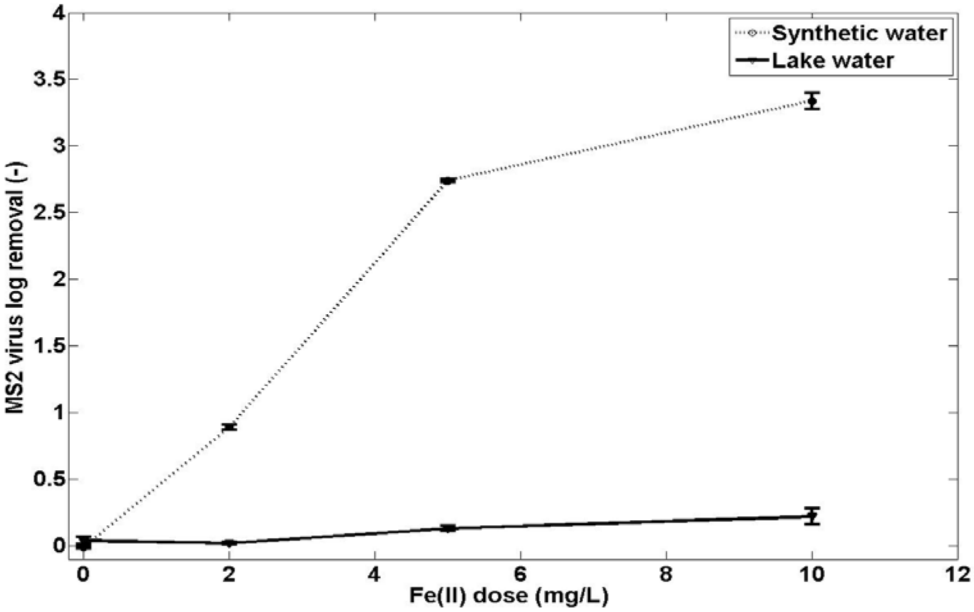


Figure 5-39.—Comparison of MS2 removal in synthetic water and lake water by CC-SF at pH 7.5 using Fe(II) as a coagulant, MS2 feed concentration = 7.9*106 PFU/mL.

4.4.2.12. *Proposed hypothesis for poor removal of viruses from lake water using EC-MF and Fe(II) CC-SF*

As explained above, both EC-MF and Fe(II) CC-SF did not improve virus removal in lake water although these processes did improve virus removal in the synthetic water. The reason for the dramatic difference observed in the EC's performance is hypothesized to be due to negatively charged NOM present in lake water, which binds quickly and strongly with the Fe(II) generated during the EC process, and hence hinders Fe(II) oxidation, hydrolysis, and the subsequent sorption of MS2 viruses. The interfering effect of NOM on Fe(II) oxidation has been reported in several studies (Liang et al. 1993, Ninh Pham et al. 2004, Rose and Waite 2003, Theis and Singer 1974). These researchers have studied the oxidation kinetics of Fe(II) in the presence of organic matter and have reported Fe(II) oxidation to be affected due to the Fe(II)-NOM complexes formed. Hence, it was assumed that the Fe(II)-NOM complex formed in the EC experiments affected the Fe(II) oxidation, thereby reducing the formation of hydroxide precipitates ($Fe(OH)_3(s)$) that can efficiently remove the viruses.

To confirm the hypothesis, Fe(II) chemical coagulation experiments were repeated in synthetic water with varying doses of a well characterized NOM (Suwannee River NOM) and the virus log removals were calculated.

4.4.2.13. *Effect of Suwannee River NOM on MS2 log removal in synthetic water by CC-SF using Fe(II) as the coagulant*

As explained in the previous section, to test the hypothesis that NOM—and not some other constituent of lake water—caused poor virus removal, varying concentrations of a "standard" aquatic NOM from the Suwannee River NOM (0-2.7 mg/L DOC) were dosed into the synthetic water, and virus removal was determined for a Fe(II) dose of 9-10 mg/L.

The experimental results for the effect of NOM on virus removal are presented in Figure 5-40. As the DOC concentration increased, the MS2 virus log removal decreased significantly from 3.5 to 0.01. This clearly indicates that the presence of NOM interferes with virus removal during chemical coagulation with Fe(II). These results support our hypothesis that the decrease in log virus removal results from Fe(II)-NOM complexation that is affecting Fe(II) oxidation and hydrolysis. Hence, it can also be concluded that the presence of NOM in lake water is responsible for poor performance of virus removal by EC-MF due to generation of Fe(II) and subsequent formation of Fe(II)-NOM complex.

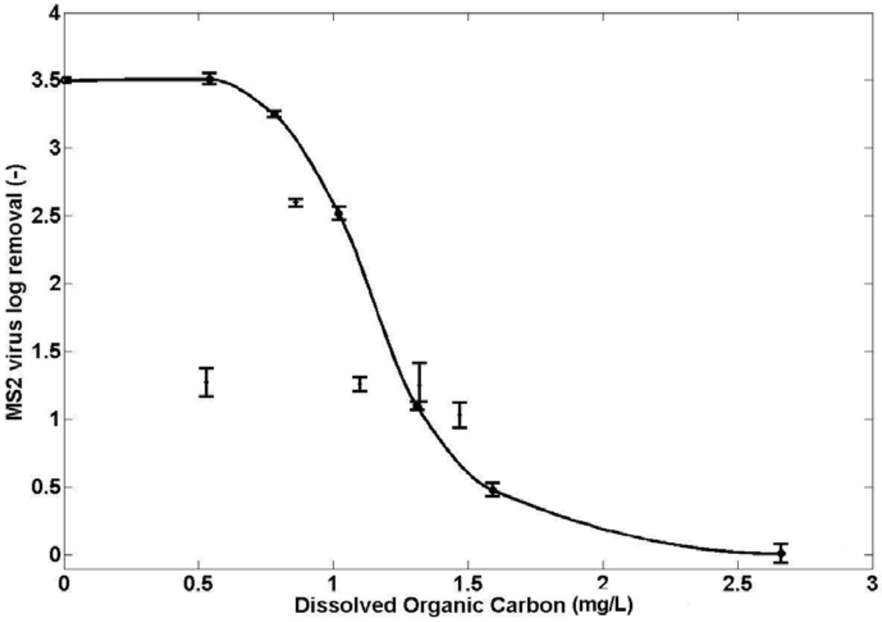


Figure 5-40.—Effect of Suwannee River NOM on MS2 log removal in synthetic water by CC-SF at pH 7.5 using Fe(II) as the coagulant with dose 9-10 mg/L.

4.4.2.14. *Fe(II)-NOM interactions*

To better understand the mechanism of the Fe(II)-NOM complex being formed in the EC unit, the total iron was measured in the microfiltered water from the experiments shown in Figure 5-40. It was expected that the filtered synthetic water would contain iron because of the presence of the un-oxidized Fe(II) that is soluble and would have passed through the filter. The analytical results represented in Table 5-4 show that after Fe(II) coagulation of the synthetic water, the percentage of total iron passing through the filter ranged from 0 to 94% as the iron dose decreased. As explained previously, either uncombined soluble Fe(II) ion or a soluble NOM-Fe(II) complex was expected to pass through the filter, and result in a large percentage of iron in the filtrate. Thus, the presence of ~0 % Fe in the filtrate for large Fe(II) doses was not expected.

Table 5-4.—Iron in CC effluent before and after 0.2 µm syringe microfiltration

DOC in feed (ppm)	**Fe(Tot) (ppm)**		**% Fe in filtered effluent**
	Feed	**Filtered effluent**	
3.78	**0.43**	**0.41**	**94%**
3.46	**1.13**	**1.05**	**93%**
3.98	**2.31**	**0.02**	**1%**
3.99	**5.95**	**0.05**	**1%**
5.03	**12.87**	**0.09**	**1%**
3.23	**18.03**	**0.10**	**1%**

To better understand why samples coagulated with high iron dosages resulted in ~0 % Fe in the filtrate, the total DOC was compared with the total iron in the microfiltered effluent samples. Figure 5-41 shows the % Fe and % DOC in the filtrate as a function of the DOC/Fe ratio in the CC feed. At DOC/Fe ratios ≥ 3.1, there was > 90% Fe and > 90% DOC present in the effluent after filtration. However, at DOC/Fe ratio ≤ 1.7, there was 57 to 83% DOC and ~ 0% Fe in the filtered effluent.

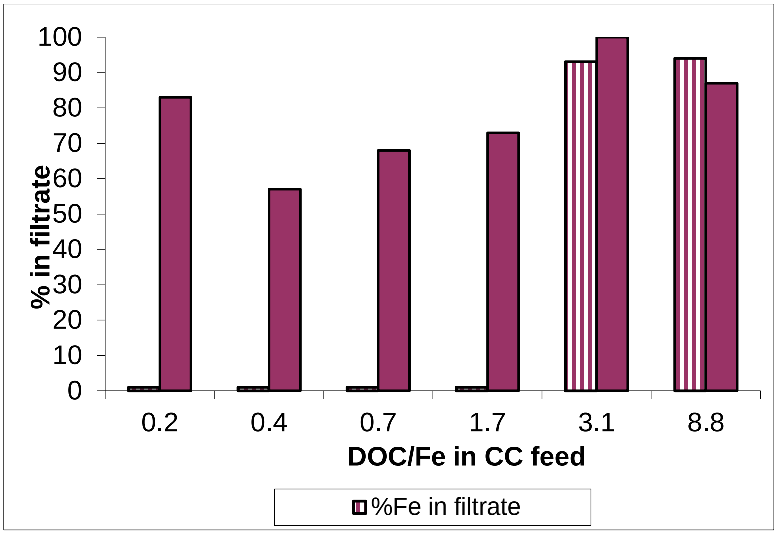


Figure 5-41.—Percentages of Fe and DOC in the filtered effluent as a function of DOC/Fe ratio in feed synthetic water by CC-SF at pH 7.5 using Fe(II) as the coagulant with dose 9-10 mg/L.

These differences in the Fe and DOC concentrations will be explained using the proposed theory discussed below. Figure 5-42 and Figure 5-43 are schematic diagrams of the relative concentrations of NOM and Fe present in the feed at high (≥ 3.1) and low (≤ 1.7) DOC/Fe ratios respectively. As shown in Figure 5-42, at high DOC/Fe ratios, there was at least three times the mass of DOC as compared with mass of Fe present in the feed sample. This resulted in (a) Fe(II) binding to a single NOM molecule and (b) less number of Fe(II)-NOM complexes being formed that would be large enough to be filtered out by the 0.2 µm microfilter. Hence, it was expected that large fraction of Fe(II) and DOC would pass through the filter and appear in the filtrate.

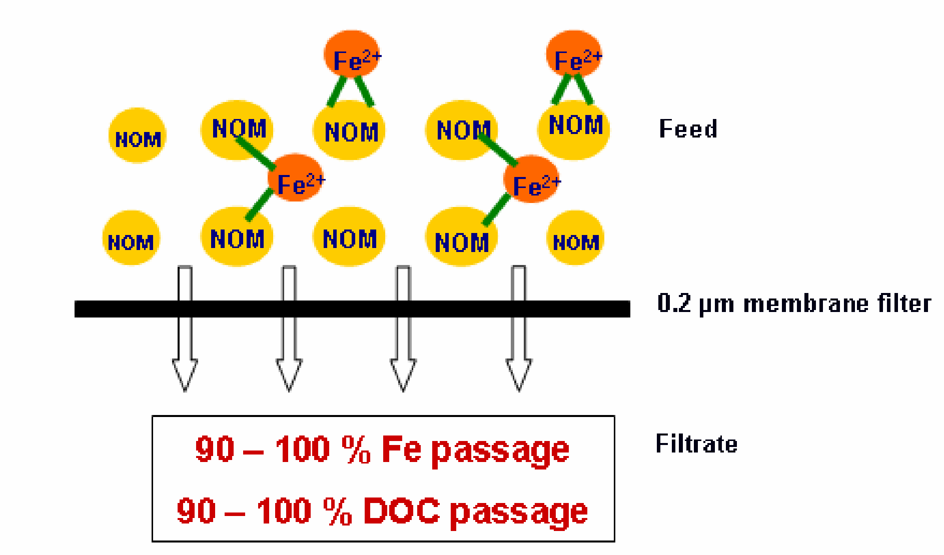


Figure 5-42.—Schematic representation of NOM and Fe molecules present at high DOC to Fe ratios (DOC/Fe ≥ 3.1).

However, when DOC/Fe ratios were low (≤ 2), it is hypothesized that the Fe(II) ions linked the NOM molecules together to form NOM-Fe polymers that were large enough to be filtered out by the 0.2 µm microfilter (as observed in Figure 5-43). This resulted in effective (nearly 100%) removal of all the iron molecules by the microfilter. However not all the DOC was removed because many NOM molecules do not bind with Fe(II) and are smaller in size. Therefore, these NOM molecules effectively passed through the filter and appeared in the filtrate as 58 to 83% DOC.

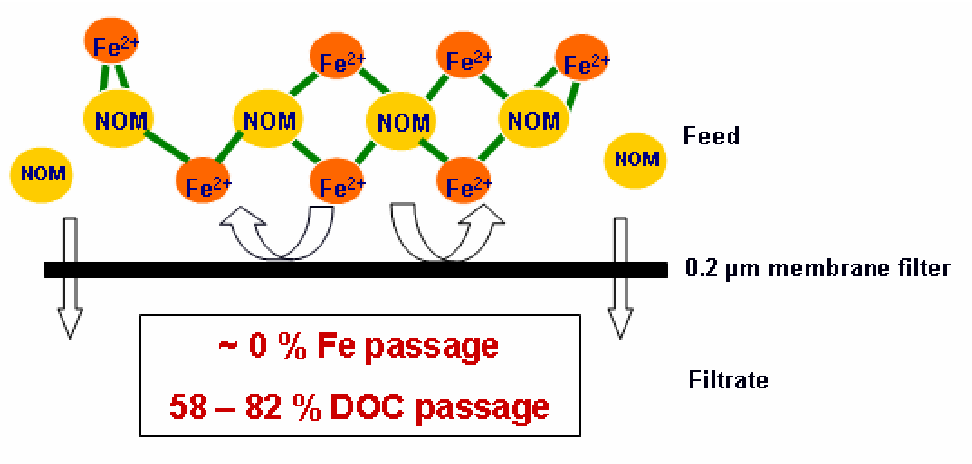


Figure 5-43.—Schematic representation of NOM and Fe molecules present at low DOC to Fe ratios (DOC/Fe ≤ 1.7).

The above simple models describing NOM (or) DOC binding with Fe(II) qualitatively explain the following observations:

1. In samples containing significant concentrations of DOC, the NOM binds with Fe(II) and prevents its oxidation to Fe(III) and subsequent hydrolysis to $Fe(OH)_3(s)$ that can adsorb the viruses and remove them effectively in EC and Fe(II) CC experiments. This resulted in poor virus removal in these experiments.
2. Although Fe(II) ions are soluble, Fe(II) is not present in the microfiltered effluent when the DOC/Fe ratios are low (≤ 1.7).
3. Fe(II) passes through the microfilter when the DOC/Fe ratios are relatively high (≥ 3.1).

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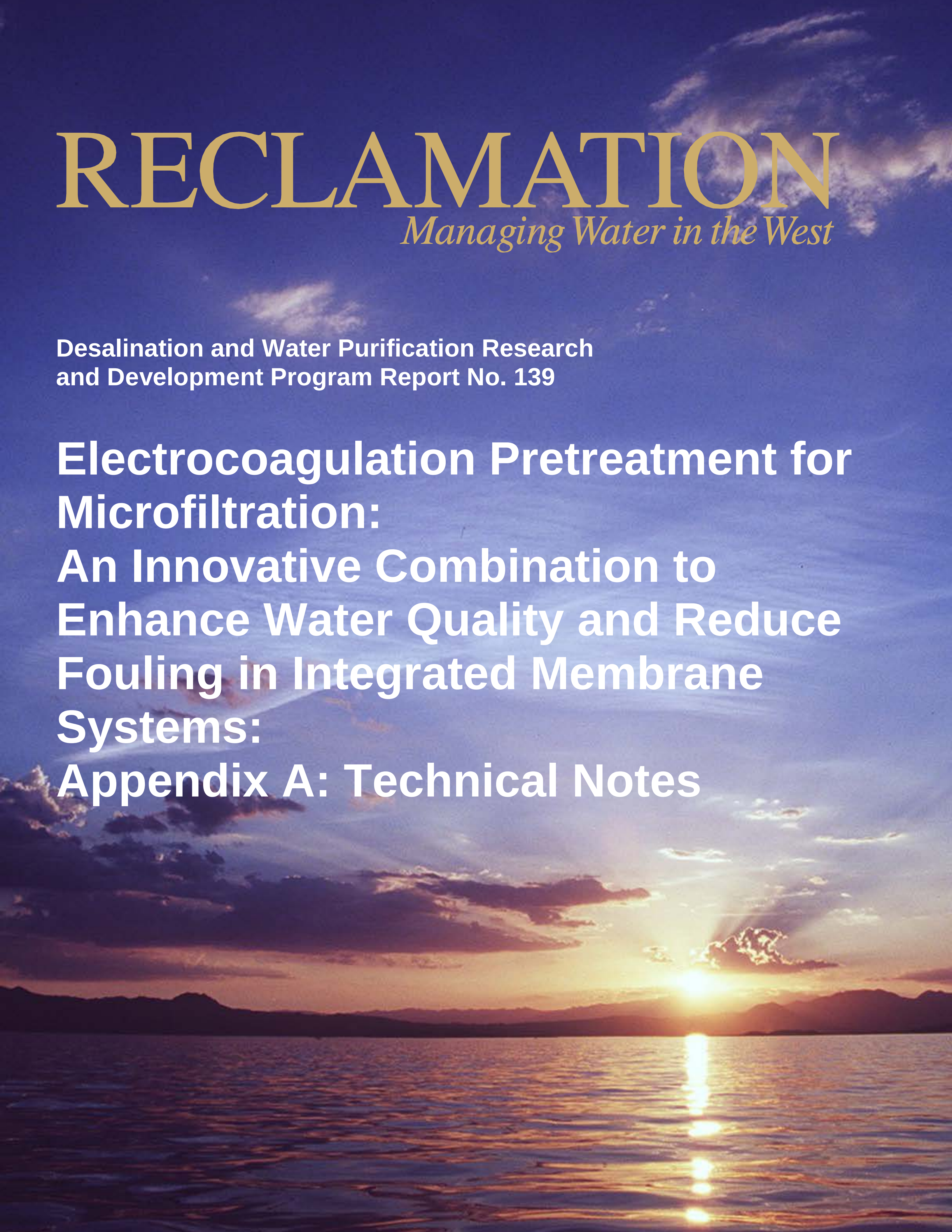

RECLAMATION

Managing Water in the West

Desalination and Water Purification Research
and Development Program Report No. 139

Electrocoagulation Pretreatment for Microfiltration: An Innovative Combination to Enhance Water Quality and Reduce Fouling in Integrated Membrane Systems:

Appendix A: Technical Notes

Appendix A. Technical Notes

A.1. Effects of transmembrane pressure on duration of intermediate blocking

Intermediate blocking law has been derived based on the assumption that when the particles are bigger membrane pores, the incoming foulant layer has an equal probability to deposit on an already deposited foulant layer as on the clear membrane surface. The general form of the intermediate blocking law is given as

$$\frac{d^2t}{dV^2} = k_i \frac{dt}{dV}\,. \qquad \text{(A.1)}$$

Upon integration, equation (A.1) yields

$$\frac{dt}{dV} = \frac{1}{Q_0} e^{k_i V}\,. \qquad \text{(A.2)}$$

The value of k_i is given as

$$k_i = \frac{\sigma \Delta P}{\mu R_m Q_0} \qquad \text{(A.3)}$$

Where:

Q_0 is the initial flow rate

ΔP is the transmembrane pressure

μ is the absolute viscosity

R_m is the clean membrane resistance

σ is the membrane surface area blocked per unit volume of suspension filtered.

The value of σ for spherical particles has been derived as Equation A.4 from Hermia (1982)

$$\sigma = 1.5 \frac{\gamma_s s}{\gamma_0 d \psi} \qquad \text{(A.4)}$$

Where:

γ_s and γ_0 are the densities of suspension and particles respectively

s is the mass fraction of particles in the suspension

d is the particle diameter

ψ is the effective deposition factor that gives an idea about the shape and structure of the aggregates deposited on the membrane surface.

Substituting the value of σ in equation A.3, the value of k_i becomes

$$k_i = 1.5 \frac{\gamma_s s \Delta P}{\mu R_m Q_0 \gamma_0 d \psi} \qquad \text{(A.5)}$$

where ψ is the only unknown parameter and was evaluated by using it as a fitting parameter in equation A.2. The variation of ψ as a function of trans-membrane pressure is depicted in Figure A1. The deposition factors decreased with increasing pressure, suggesting the strong dependence of colloidal morphology on initial permeate flux.

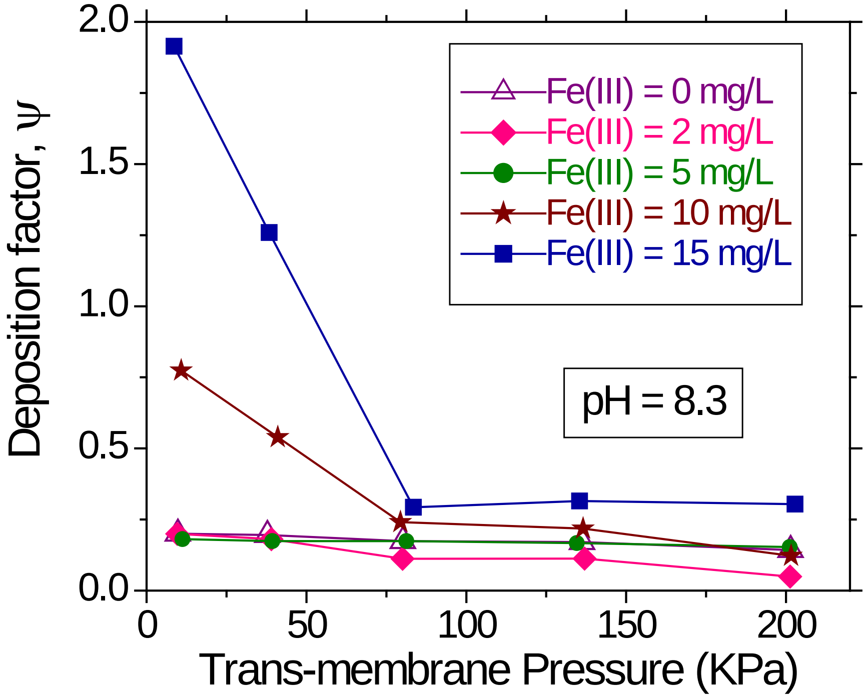

Figure A1.—Effects of transmembrane pressure on deposition factors of natural colloids.

Also, decreasing values of ψ resulted in an increase in k_i value with an increase in the applied pressure. Thus, an increase in the value of k_i should result in an increase in inverse instantaneous flux based on Equation A.2. Importantly, the initial flow rate also increased with pressure. However, Equation A.2 was more sensitive to changes in flow rate than the exponential of k_iV term. Therefore, an increase in trans-membrane pressure resulted in a decrease in the inverse flux, or in other words, an increase in the permeate flux, This explains the reduction in the intermediate blocking stage due to increase in transmembrane pressure.

A.2. Coagulation-microfiltration pretreatment to nanofiltration (NF) in integrated membrane systems

Another important aspect of this research was to quantify the possible reductions in NF membrane fouling as a result of coagulation-microfiltration pretreatment. Minimizing fouling not only decreases the operational costs by reducing the energy demand, but minimizing fouling may also reduce the additional cost associated with frequent cleaning operations and membrane replacement

A.2.1. Experimental work

A commercially available, polyamide thin film composite nanofiltration membrane, DL (Osmonics, Minnetonka, Minnesota) was employed. Cross-flow experiments were performed using the system shown schematically in Figure A2. Results are shown in Figure A3.These experiments were conducted using a stainless steel pressurized cell (SEPA-CF Cell, Osmonics, Minnetonka, Minnesota) that accommodates a 19 × 14 cm flat membrane sheet (with an effective filtration area of 155 cm^2). Initially, the fresh membrane coupon was soaked in ultrapure water over a period of 24 hours to rinse any preservative chemicals. The coupon was then placed in the steel holder, and ultrapure water was passed through the entire system for 24 hours at a constant pressure of 470 kilopascal (kPa) . Following this setting period, pure water permeability measurements were made by measuring steady state fluxes in the pressure range of 130-500 kPa. The system was operated in a closed loop mode where both the retentate and permeate streams were recycled to a 6L feed tank (containing microfiltered lake water) to keep the feed concentration constant and to limit the volume of feed water required. The fouling tests were conducted at a constant feed water recovery of ~ 70% and a constant trans-membrane pressure (470 KPa). A positive displacement gear pump (model 74011-11, Cole Parmer, Chicago, Illinois) was employed to pump magnetically stirred feed water to the membrane cells. Inert materials such as Teflon or stainless steel were used for all the wetted components like tubing, connections and the membrane cell. The retentate and permeate flow rates were monitored manually using a stopwatch and a measuring cylinder.

Filtration pressure and temperature were monitored using analog transducers (PX603 and TJ120 CPSS 116G, Omega Engineering Company, Stamford, Connecticut). The temperature of the feed water was recorded using a 12 in rugged probe (TJ120 CPSS 116G, Omega Engineering Co., Stamford, Connecticut) and was maintained at 23 ± 1°C. Data acquisition was accomplished using a program written in LabVIEW at a sampling rate of 0.002 Hz. Instantaneous flux (and specific flux) values were plotted with respect to time to evaluate the fouling rate over the experimental duration.

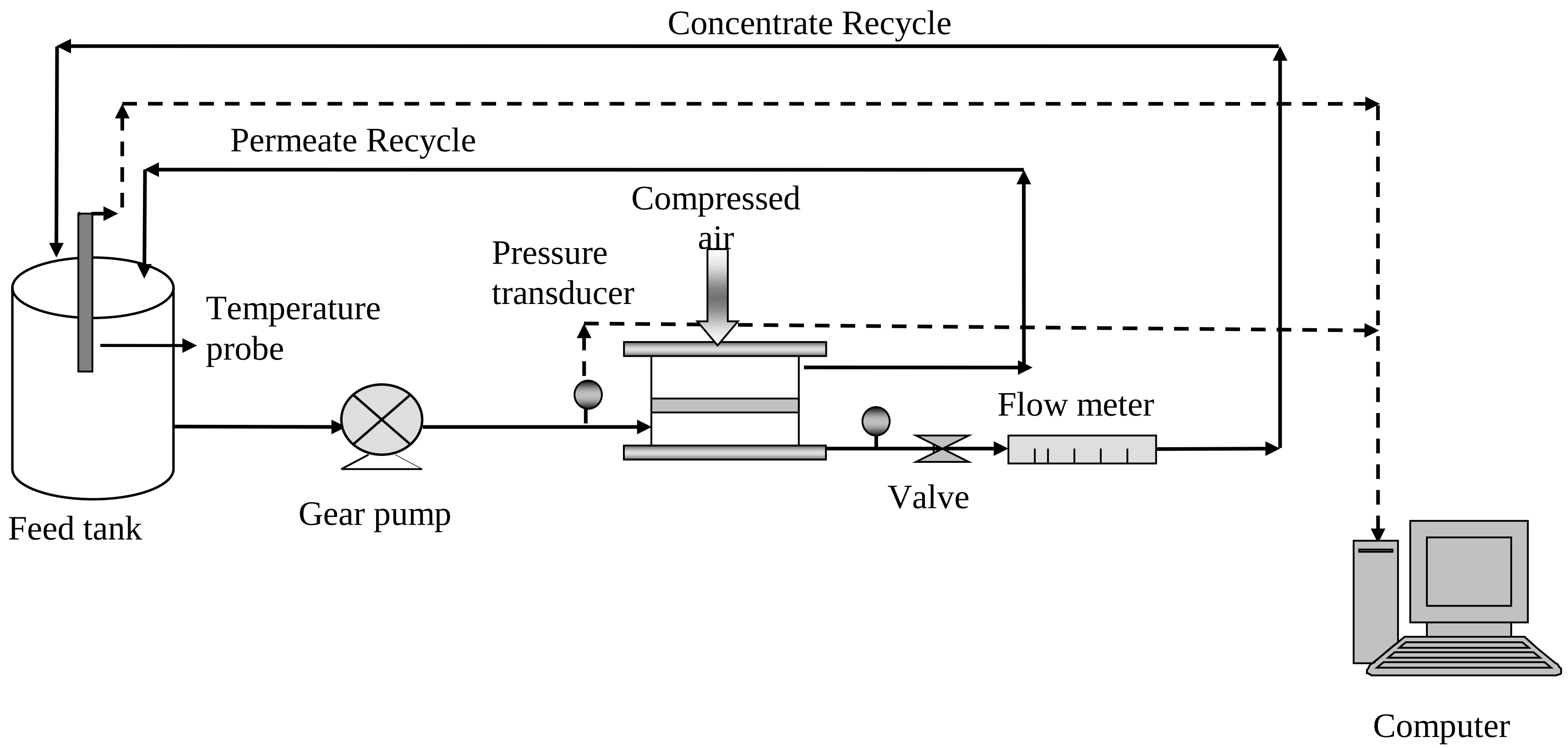

Figure A2.—Schematic of the closed loop bench scale nanofiltration apparatus.

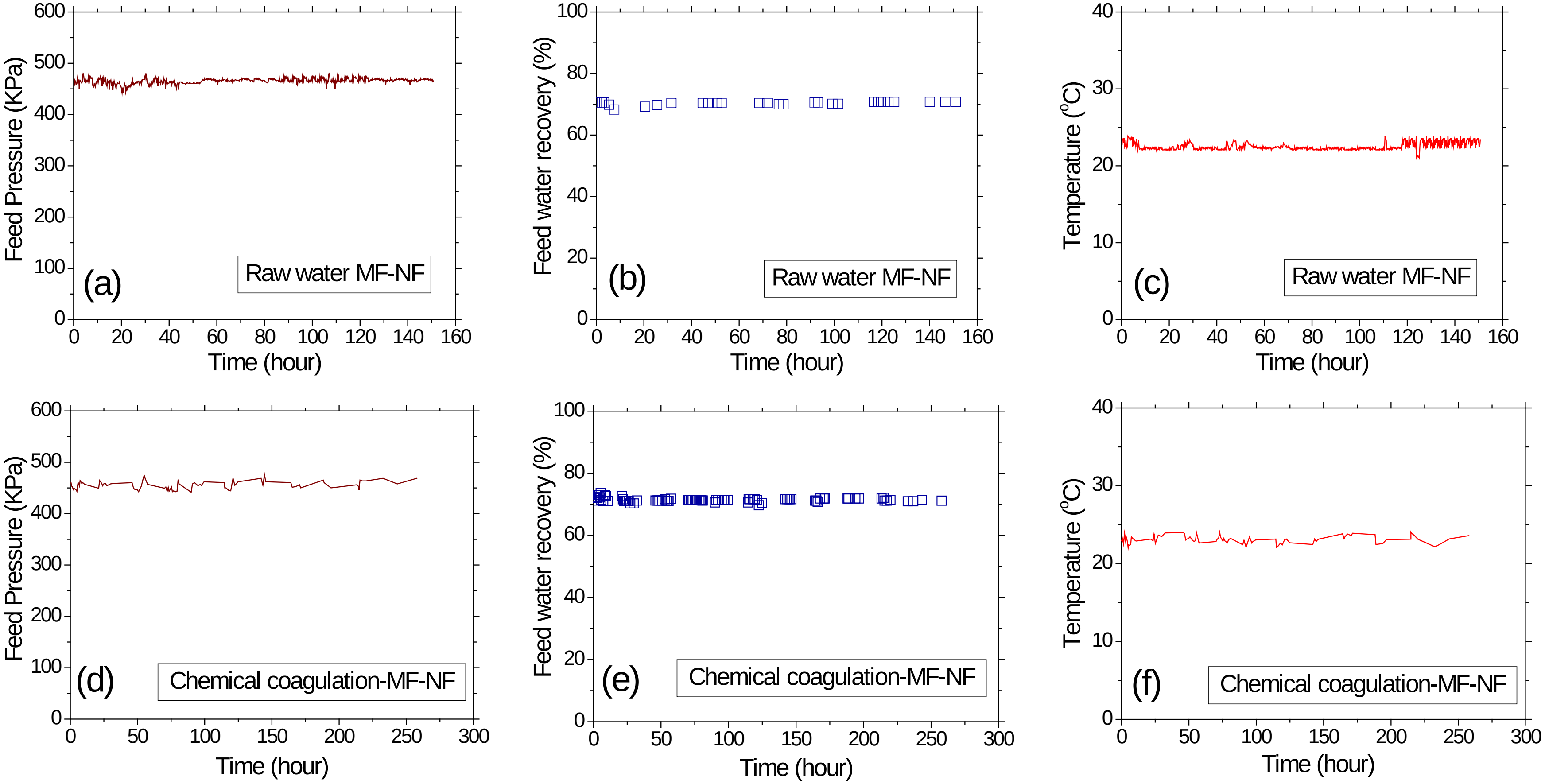

Figure A3.—Continuous monitoring of permeate recovery, trans-membrane pressure and temperature of the feed water throughout the NF fouling test as a measure of quality control.

A.2.2 Results and discussion

A.2.2.1. Quality control and quality assurance

As depicted in Figure A3, for both the experiments, the transmembrane pressure, recovery, and temperature were all maintained constant over a period of 7 to 11 days at ~ 466 KPa (68 psi), 70% and 23°C respectively, allowing us to have a quantitative comparison of the results obtained over the entire duration of the experiment.

A.2.2.2. Effect of pretreatment on nanofiltration membrane fouling

We used the instantaneous specific permeate flux to compare differences in fouling of the nanofiltration membrane with varying pretreatments. The specific permeate flux declined by ~24-30% over a period of ~150-250 hours for the microfiltration only as well as for electrocoagulated-microfiltration pretreatment scenario, whereas the specific permeate flux decline was only ~7.5% over the same duration when Lake Houston water was pretreated using chemical coagulation and microfiltration (Figure A4).

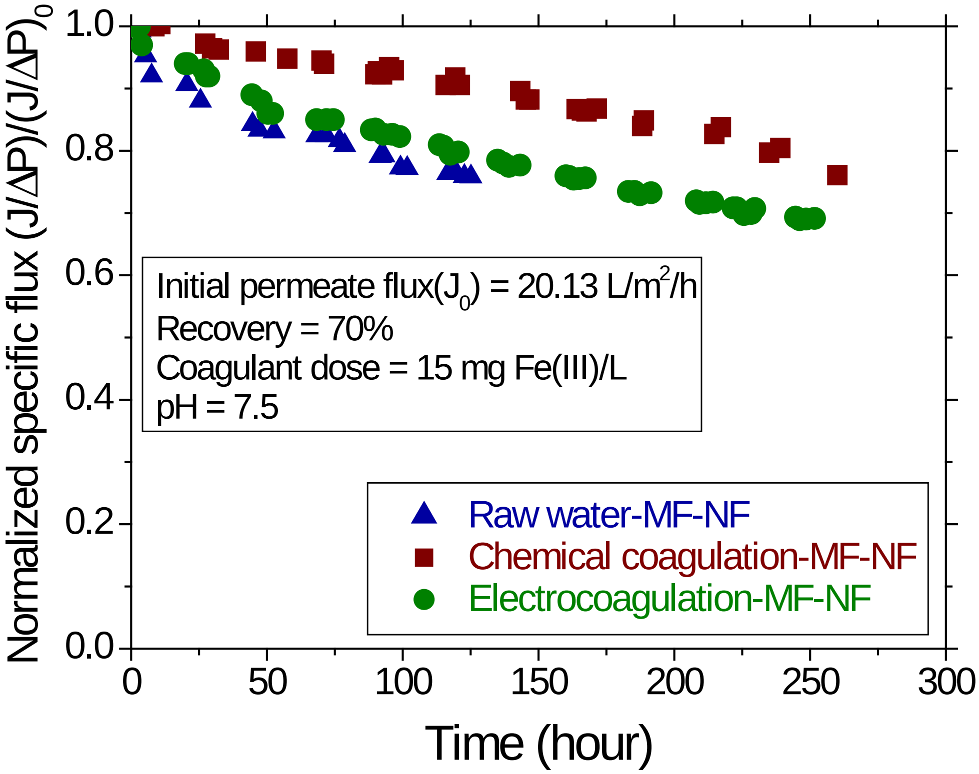

Figure A4.—Decreased NF membrane fouling for Lake Houston water pretreated using chemical coagulation and microfiltration, compared with microfiltration alone.

In addition to differences in the total amount of specific permeate flux decline, the two types of pretreatment also caused variations in the kinetics of fouling or shape of the fouling profiles. For example, results shown in Figure A4 also indicate that

NF membrane fouling rate did not remain constant throughout the duration of the experiment using Lake Houston water pretreated using MF alone. The specific permeate flux declined rapidly over the first 25 hours of operation at a rate of 0.0215 liters per meter square hour bar (L/m^2-bar-h). However, during the latter stages of the experiment, the fouling rate decreased to 0.0054 L/m^2-bar-h. The average fouling rate for the entire duration of the experiment was 0.0084 L/m^2-bar-h, which is approximately twice the fouling rate (0.0038 L/m^2-bar-h) observed for Lake Houston water that was pretreated using chemical coagulation and microfiltration. These results demonstrate that using a combined chemical coagulation – microfiltration pretreatment for nanofiltration reduced fouling of the nanofiltration membrane employed in our experiments. Additionally, the average fouling rate for the Lake Houston water pretreated using electrocoagulation and microfiltration pretreatment (0.0071 L/m^2-h-bar-h) was found to be almost similar to that of Lake Houston water pretreated using MF alone. These results clearly show that electocoagulation pretreatment did not prove to be beneficial for reducing fouling of NF membranes.

At the end of the fouling run, the membranes were carefully removed from the cells and photographs of the membranes were taken by a digital camera. Typical photographs of the fouled as well as the fresh membrane coupon are shown in Figure A5. A brown-colored fouling layer can be observed in Figure A5b that potentially caused the decline in permeate flux.

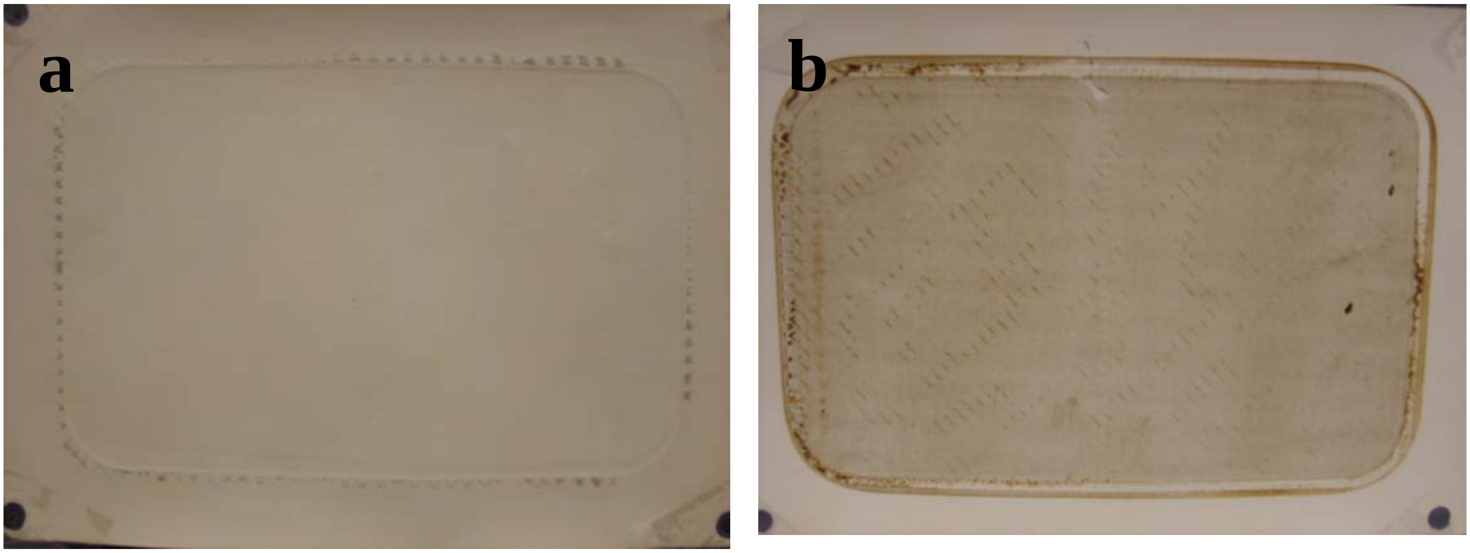

Figure A5.—Snapshots of the fresh (a) and the fouled (b) membranes at the beginning and at the end of the fouling run.

A scanning electron micrograph of the foulant layer is shown in Figure A6. As seen, the foulant layer was rough potentially consisting of NOM, colloids, and salt precipitates.

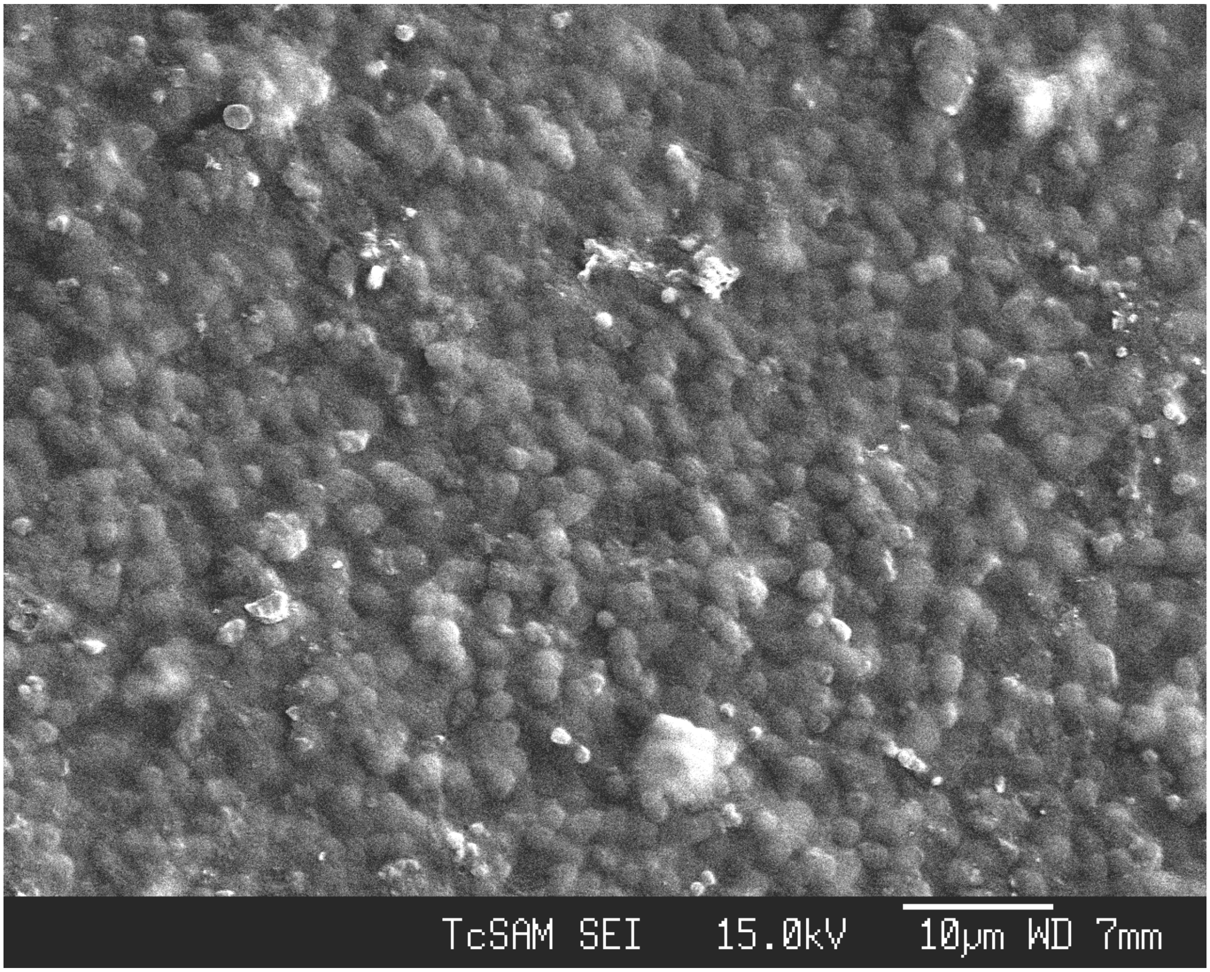

Figure A6.—SEM image of the foulant layer on the nanofiltration membrane formed by CC-MF pretreated Lake Houston water at pH 7.5.

A.3. Batch adsorption tests using Lake Houston water and nanofiltration membranes

Organic fouling of nanofilters could be caused by a combination of NOM adsorption on the membrane and permeation-drag induced deposition on the membrane. The second mechanism is essentially caused by the convective flow of water across the membrane due to the transmembrane pressure along with clean water permeation. The propensity of NOM adsorption on the DL membrane was determined in batch tests as described below.

A.3.1. Materials and methods

To estimate the adsorption capacity of the DL membrane, different areas of membrane, ranging from 1 cm^2 to 45 cm^2 (cut into small square pieces of ~ 1 cm^2) were suspended in 125 mL of water sample (microfiltered-Lake Houston water) in different amber-colored bottles wrapped in aluminum foil to prevent the possibility of light-mediated changes in aquatic NOM content. All tests were set up in duplicate to verify the reproducibility of the measurements.

Two different sets of controls were also set up in addition to the test samples: one with ultrapure water (125 mL) and a membrane area of 45 cm^2 and the second control bottle had microfiltered-Lake Houston water with no membrane. The controls were also set up in duplicate.

A.3.2. Results and discussion

As depicted in Figure A7a, the UV_{254} value of the aqueous phase did not change in the sample bottle subjected to maximum area (45 cm^2) of membrane over a period of 7 days. Similar results were obtained for the rest of the bottles as well that contained lesser amounts of the membrane. Control experiments revealed:

(1) No NOM adsorption on the sample bottles and caps as evidenced by constant UV_{254} measurements in tests with no membranes (Figure A7b)

(2) No leaching of organics from the membrane as evidenced by the constant and near zero UV_{254} measurements in tests with the largest membrane area employed (45 cm^2) and suspended in ultrapure water (Figure A7c).

Note that the error bars corresponding to the standard deviation are within the size of the symbols used in Figures A7a, b, and c demonstrating the precision of UV_{254} measurements and reproducibility of our experimental protocols.

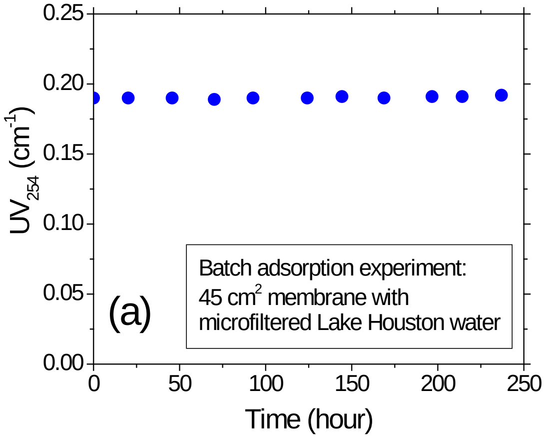

Figure A7a.—Constant UV254 concentration in water sample subjected to membrane area of 45cm2.

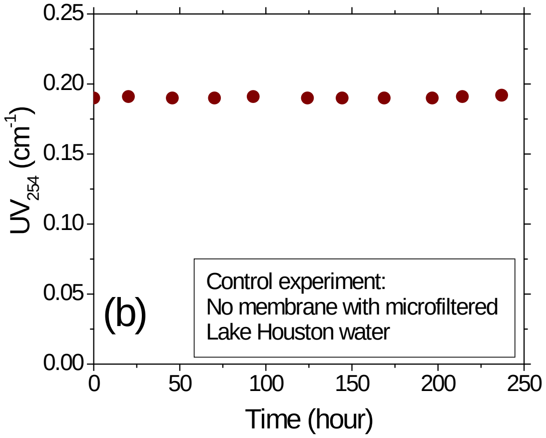

Figure A7b.—Constant aqueous phase NOM concentration over ~ 250 hours during the batch adsorption tests in the absence of any membrane.

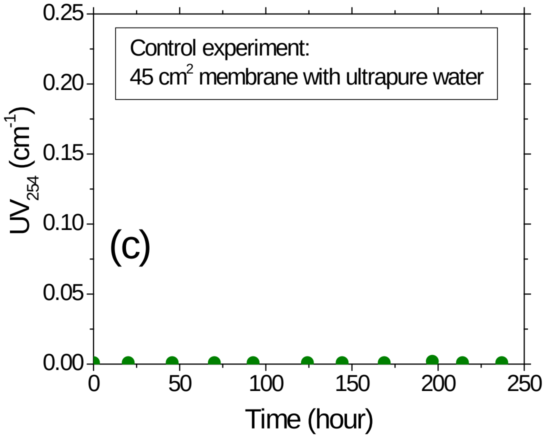

Figure A7c.—No leaching of dissolved organic carbon from the membrane during the ~ 250 hours of the batch adsorption tests.

Nearly constant UV_{254} measurements reported in Figure A7a along with the results from the controls reported in Figures A7b and c demonstrate that NOM adsorption was not the predominant fouling mechanism. Therefore, convective deposition of NOM arising from permeation of water across the membrane caused by the trans-membrane pressure is expected to be the major cause of fouling under our experimental conditions. Further, these results suggest that the reduction in feed water NOM concentration is caused by the formation of a foulant layer on the membrane surface (as shown in Figures A5 and A6).

A.4. Disinfection byproduct precursor removal achieved by chemical coagulation

As reported previously, NOM was characterized in terms of DOC, ultraviolet absorbance at 254 nm and 1 cm path length (UV_{254}), and DBP precursors. For this reason, simple linear correlations were developed between DOC, UV_{254}, trihalomethanes (THM) and haloacetic acids (HAA) that are summarized next.

Chlorination of drinking waters containing organic carbon and bromide ion results in the formation of potentially carcinogenic, teratogenic, and mutagenic by-products, including THMs and HAAs. To date, only five HAAs are currently regulated under the Disinfectants/Disinfection By-Products (D/DBP) Rule: monochloro-, dichloro-, trichloro-, monobromo-, and dibromoacetic acid (ClAA, Cl_2AA, Cl_3AA, BrAA, and Br_2AA, respectively). A sixth HAA (viz. bromochloroacetic acid, BrClAA) was included in the Information Collection Rule (ICR). For this reason, all the four trihalomethanes and six haloacetic acids were estimated. Note that DBPs were enumerated under “uniform formation conditions” corresponding to an incubation time of 24 hours, a 24-hour chlorine residual ~ 1 mg/L, and 8.3 pH.

A.4.1. Experimental work

A.4.1.1 Chlorination experiments

The chlorine doses for raw as well as pretreated waters were determined by conducting preliminary chlorine demand tests to achieve a free-chlorine residual of 1.0 ± 0.4 mg/L as Cl_2 after headspace-free storage for 24 ± 1 hour at 20 ± 1ºC in the dark (Summers et al. 1996). Chlorination was performed in 250 mL amber colored glass bottles with polytetrafluoroethylene (PTFE)-lined caps and zero chlorine demand on samples buffered to pH 8.0 with borate buffer. Sufficient quantities of sodium hypochlorite were added to well-stirred samples to assure a free-chlorine residual of 0.6-1.4 mg/L. After chlorination, the samples were stored headspace-free at 20 ± 1ºC in dark for 24 hours. After incubation, free chlorine residual was measured by adding DPD free chlorine reagent (Hach Company, Loveland, Colorado) in 10 mL of chlorinated sample. The absorbance of the resulting complex was measured at 530 nm wavelength using a glass cell with one inch path length and a spectrophotometer (DR/4000 Spectrophotometer, Hach

Company, Loveland, Colorado). The chlorine demand was calculated as the difference between the free-chlorine dosage and the free-chlorine residual after 24 hours of incubation at 20ºC.

A.4.1.2. DBP formation tests

After 24 hours of incubation, the samples were collected head-space free in 40 mL glass vials with polypropylene screw caps and Teflon-lined septa for subsequent THM and HAA analysis. 100 mg/L of ammonium sulfate was added in the vials to quench the residual free chlorine. All the samples were stored at 4ºC for not more than two weeks prior to analysis.

The THM4 calibration solutions were prepared from THM Calibration Mix standard solution (Supelco Inc., Bellefonte, Pennsylvania). Fluorobenzene and 1,4- dichlorobenzene were used as internal standards. Method performance was monitored by adding 1-bromo-2-fluorobenzene as the surrogate. Individual THM species were identified and quantified using a gas chromatography system (CP-3800, Varian Instruments, Walnut Creek, California) equipped with a mass spectrometer (Saturn 2000, Varian Instruments, Walnut Creek, California) and a purge and trap concentrator (3100, tekmar Control Systems, Inc., Spokane, Washington).

HAA6 compounds were analyzed by liquid/liquid extraction in accordance with EPA analytical method 552.2. 40mL of the sample was adjusted to $pH < 0.5$ by adding 4 mL of conc. H_2SO_4 and was extracted with 4 mL of methyl *tert*-butyl ether (MTBE). The HAAs partitioned in the organic phase were converted to methyl esters by adding acidic methanol followed by slight heating. The acidic extract was then neutralized by addition of 4 mL of saturated sodium bicarbonate solution. We extracted 0.5 mL of the upper organic phase and identified and quantified the target analytes using a capillary column gas chromatograph fitted with an electron capture detector (5890 GC, Agilent Technologies, Foster City, California). We used 1,2,3- trichloropropane (Restek Corporation, Bellefonte, Pennsylvania) as an internal standard and 2-bromopropionic acid (Restek Corporation, Bellefonte, Pennsylvania) as a surrogate to monitor the HAA extraction efficiency. The extracts were stored at -10ºC for not more than two weeks prior to analysis. The HAA6 stock solution was prepared in MTBE from Haloacetic Acid Mix #2 solution (Restek Corporation, Bellefonte, Pennsylvania). Standard solutions with known concentrations of 6 HAAs were prepared from this stock and were extracted using the method described above. The retention times observed for various HAAs while analyzing standard solutions were used to identify the individual HAA peaks generated during the gas chromatographic analysis of actual samples.

The sample preparation for DBP analyses was conducted in the University of Houston labs, whereas the analyses using gas chromatography – electron capture detection (GC/MS) and gas chromatography – mass spectrometry (GC/ECD) were conducted at City of Houston’s East Water Purification Plant. Laboratory manager, Dr. Ying Wei and other staff personnel at the East Water Purification

Plant assisted in these experiments by providing consumables, equipment, and labor in sample analysis and interpretation of the analytical results. Their support is highly appreciated.

A.4.2 Results and discussion

A.4.2.1. Changes in DBP precursor removal induced by chemical coagulation

As reported in §4.4.4, in addition to effective DOC removal, chemical coagulation was also beneficial in reducing its reactivity with chlorine to form DBPs. As depicted in Figures A8a, b, and c, chemical coagulation was found to be highly effective in the removal of 24-hour chlorine demand and trihalomethanes and haloacetic acid precursors at both pH 6.4 and 7.5. The DBP precursor removal increased monotonically with increasing Fe(III) dosages. However, higher DOC and UV_{254} removals at lower pH value of 6.4 (Figure 4.7) were responsible for higher DBP precursor removals achieved at pH 6.4 as compared to pH 7.5 (Figure A8).

A.4.2.2 Correlations between natural organic matter removal and disinfection byproduct precursors.

Linear regression analyses were performed to identify possible surrogates for natural organic matter (Figure A9) and disinfection by-product precursors (Figure A10) in pretreated waters. Note that separate correlations were obtained for chemically coagulated waters and electrocoagulated waters because the mechanisms of NOM (and turbidity) removal by these two pretreatment processes are potentially different.

As seen in Figure A9, DOC concentrations correlated very well with ultraviolet absorbance at 254 nm and 1 cm path length. Additionally, as depicted in Figure A10, in general, good correlations were obtained for the two main disinfection by-products analyzed, viz. total trihalomethanes ($CHCl_3$ + $CHCl_2Br$ + $CHClBr_2$ + $CHBr_3$) and the sum of six haloacetic acids ($ClCH_2COOH$ + $Cl_2CHCOOH$ + Cl_3CCOOH + $BrCH_2COOH$ + $Br_2CHCOOH$ + $ClBrCHCOOH$). Importantly, different DOC concentrations and UV_{254} values were not obtained by sample dilution but were generated by changing iron dosages and coagulation pH.

Results shown in Figure A10 demonstrate that in accordance with several previous studies on conventionally treated waters (Jacangelo et al. 1995, Kalscheur et al. 2006, and Reckhow et al. 1990), the occurrence and formation of various classes of DBPs were well correlated with DOC. In other words, our results identified that DOC and UV_{254} are excellent surrogates for DBPs in chemically- and electrically-coagulated Lake Houston water. Therefore, as with several other feed waters, the need for expensive and time-consuming DBP analyses could potentially be limited to regulatory compliance and more frequent monitoring of these surrogates undertaken. This will allow better process control thereby reducing potentially adverse health effects of various DBPs.

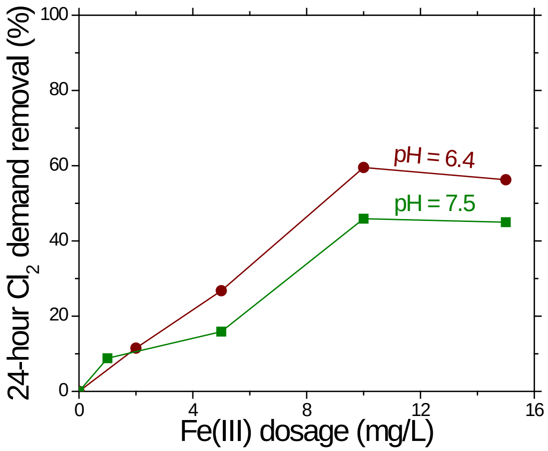

Figure A8a.—Reduction in chlorine demand with increasing coagulant dose at pH 6.4 and 7.5.

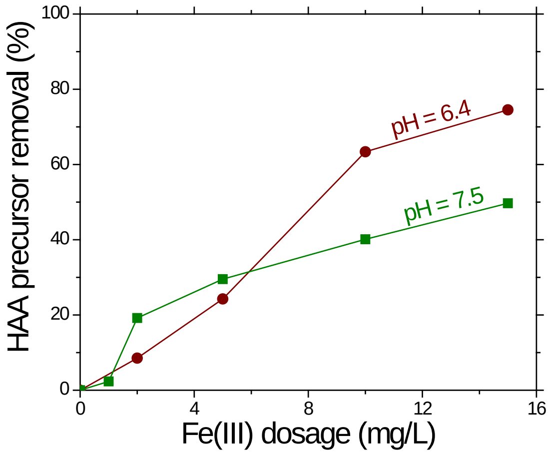

Figure A8b.—Effect of coagulant dose and coagulation pH on HAA6 precursor removal.

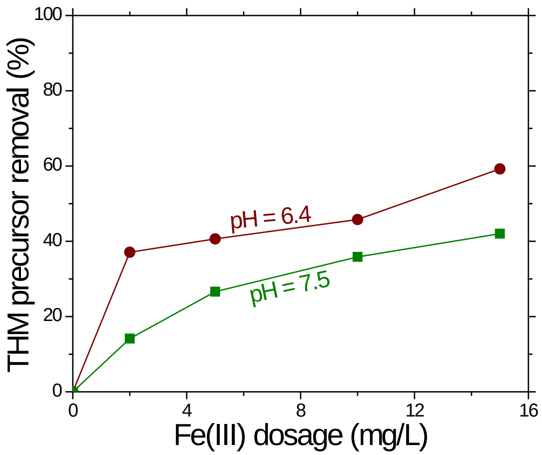

Figure A8c.—Effect of coagulant dose and coagulation pH on THM4 precursor removal.

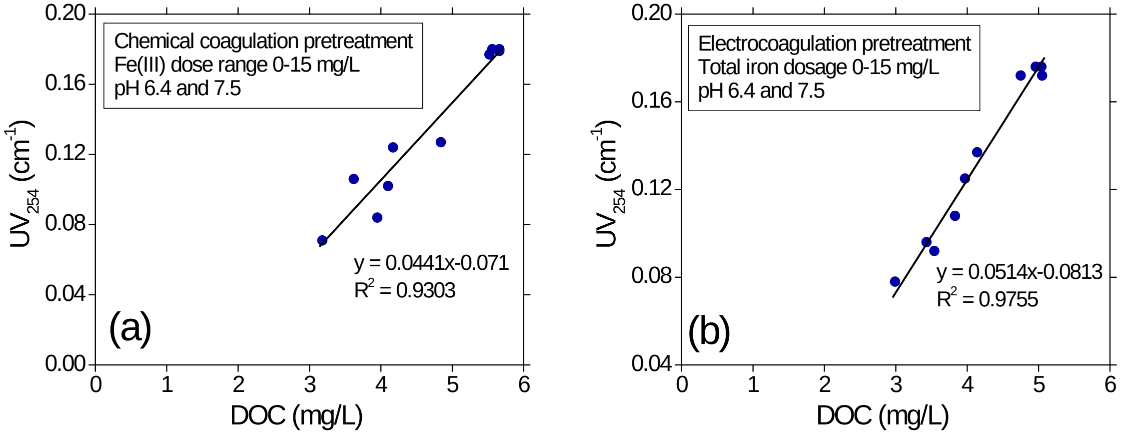

Figure A9.—Correlations between DOC concentrations and UV$_{254}$ in pretreated waters.

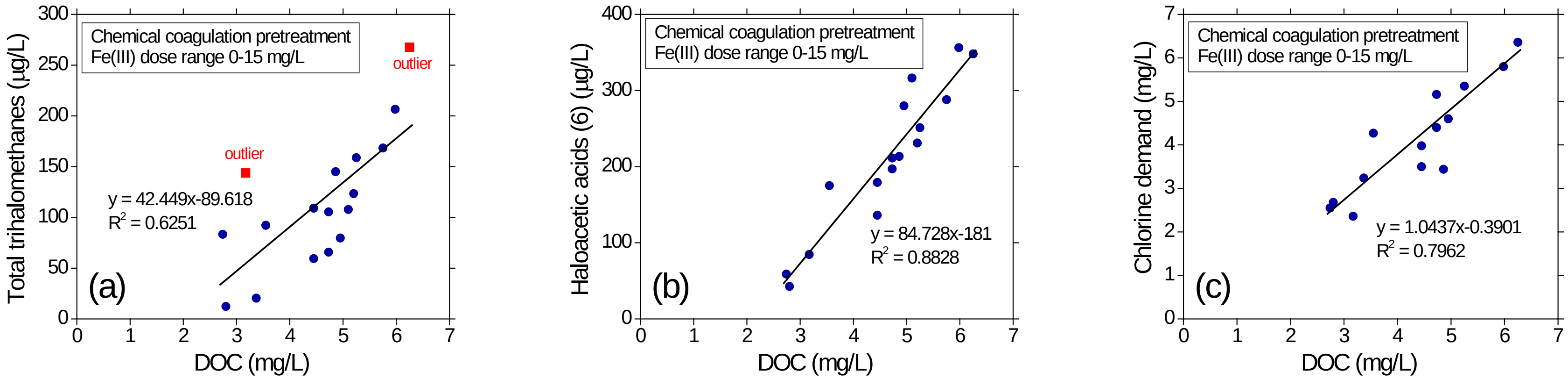

Figure A10.—Correlations of total THMs (a), HAA6 (b), and chlorine demand (c) measured during simulated distribution system tests (under uniform formation conditions) with DOC.

A.5. Compressibility of cakes formed by natural and pretreated Lake Houston water using chemical coagulation

Figure A11 shows compressible cakes formed by raw and chemically coagulated Lake Houston water at pH 6.4 and pH 8.3.

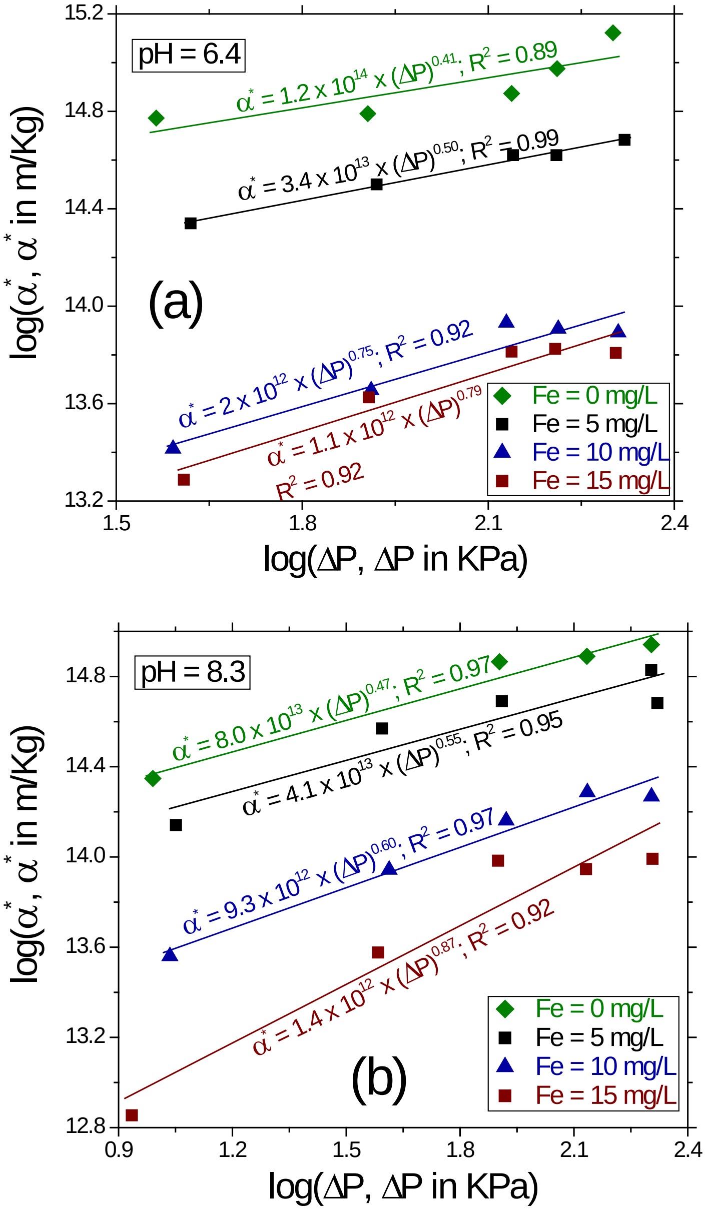

Figure A11.—Power-law compressible cakes formed by raw and chemically coagulated Lake Houston water at (a) pH 6.4 and (b) pH 8.3.

A.6. Chemical composition of Lake Houston water

The chemical composition of Lake Houston water was analyzed using flame atomic absorption spectroscopy (Varian AA220FS Fast Sequential AAS, Varian Instruments, Walnut Creek, California) and ion chromatography (LC20, Dionex Corporation, Sunnyvale, California). These analyses were also conducted at the City of Houston's East Water Purification Plant. Table A1 shows these results.

Table A1.—Chemical composition of Lake Houston water

Species	Concentration (mg/L)	Molecular Weight (mg/mmole)	Equivalent Weight (mg/meq)	Concentration (meq/L)
Cations				
Na^+	35.00	22.9898	22.9898	1.522
Mn^{2+}	0.010	54.9380	27.4690	0.0004
Fe^{3+}	0.042	55.8470	18.6160	0.0023
Mg^{2+}	2.60	24.3050	12.1525	0.214
K^+	3.32	39.0983	39.0983	0.085
Ca^{2+}	17.90	40.0780	20.0390	0.893
			Total =	**2.717 meq/L**
Anions				
F^-	0.2683	18.9884	18.9884	0.0141
Cl^-	34.9110	35.4530	35.4530	0.9847
NO_2^-	0.0020	46.0055	46.0055	0.00004
Br^-	0.0701	79.9040	79.0940	0.0009
NO_3^-	0.4704	62.0049	62.0049	0.0076
o-PO_4^{3-}	0.0655	94.9354	31.6571	0.0020
SO_4^{2-}	10.1276	96.0636	48.0318	0.2108
HCO_3^-	92.72	61.0171	61.0171	1.512
			Total =	**2.739meq/L**

mg/mmole = milligrams per millimole
mg/meq = milligrams per milliequivalent
meq/L = milliequivalent per liter

A.7. Bench scale dead-end microfiltration apparatus

Figure A12 shows the microfiltration apparatus described in §4.3.5.

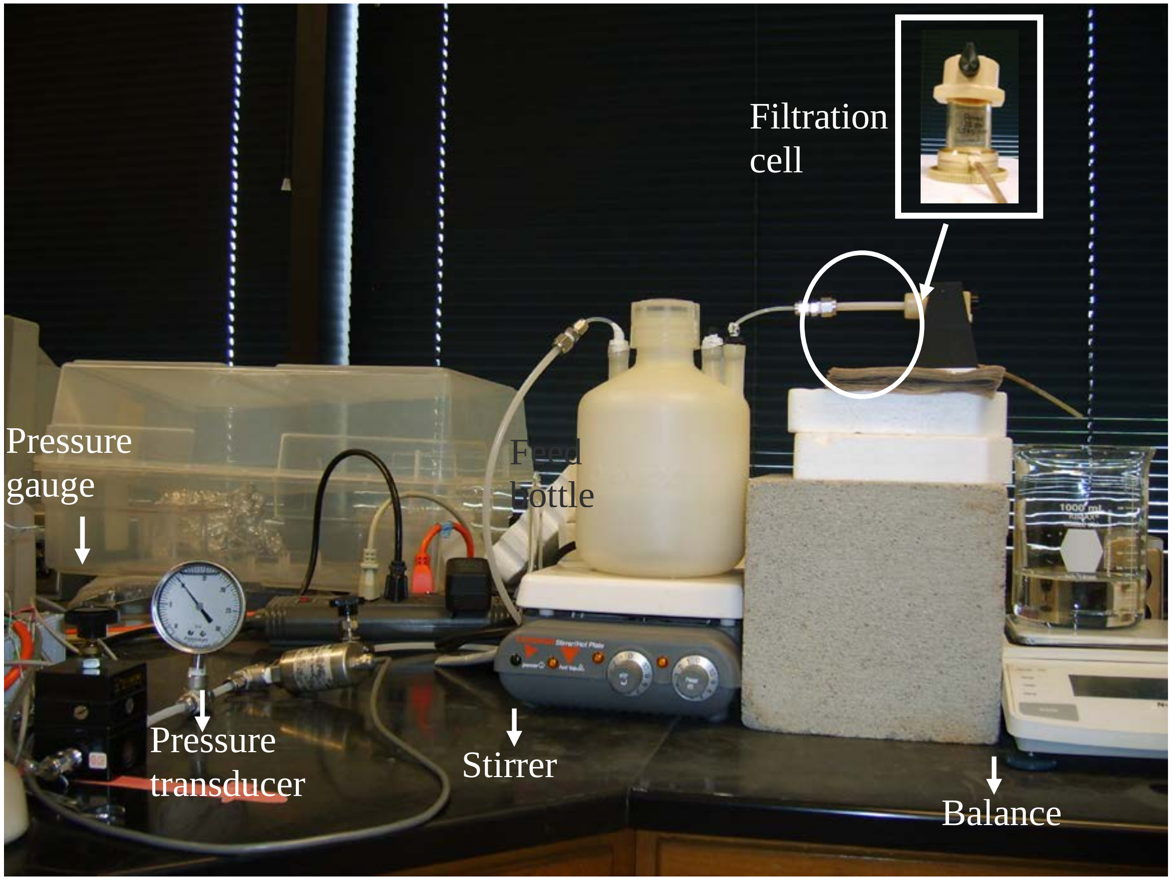

Figure A12.—Photograph of the dead-end constant pressure microfiltration apparatus described in §4.3.5.

A.8. Bench scale electrocoagulation apparatus

Figure A13 shows the schematic of the bench-scale electrocoagulation unit.

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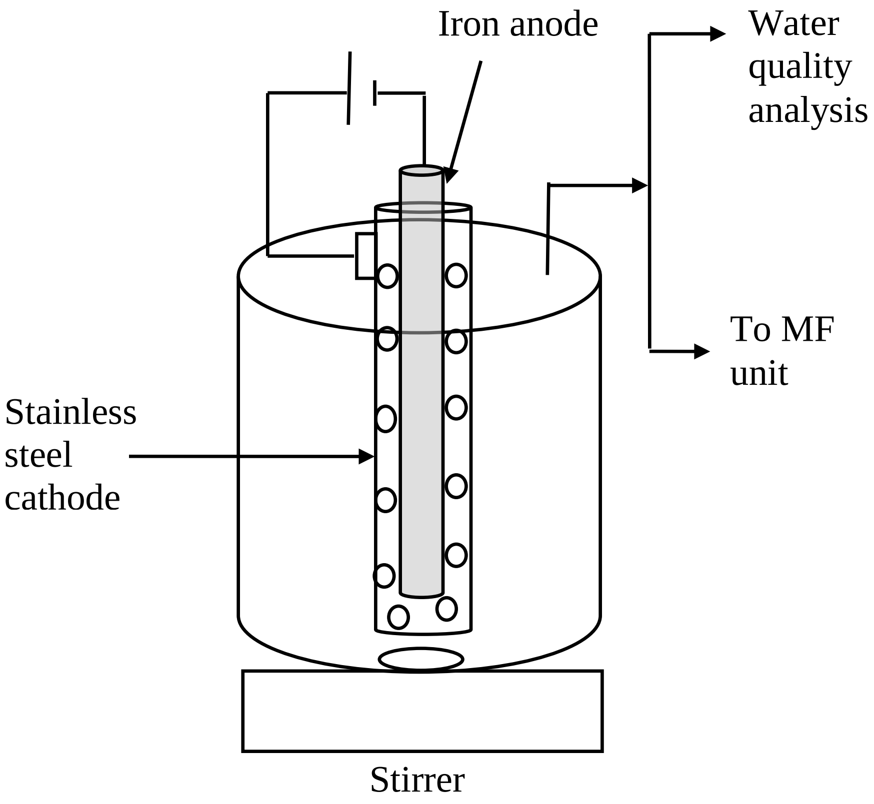


Figure A13.—Schematic of the bench scale electrocoagulation unit. The configuration and working of the unit is described in detail in §4.3.3.

A.9. Dissolved organic carbon measurement

As mentioned in §4.3.8, DOC concentration in raw and coagulated Lake Houston waters were estimated using high-temperature combustion method (Standard Method 5310B (Clesceri et al. 1998)), with the help of a total organic carbon (TOC) analyzer (TOC5050A, Shimadzu Scientific Instruments, Columbia, Maryland). Samples were acidified to pH<2 with 2N HCl and purged with ultra-zero air for 2 minutes to remove inorganic carbon. The carbon component in the sample is converted to carbon dioxide, which is detected by a non-dispersive infrared gas analyzer (NDIR). The NDIR outputs a detection signal that generates a peak whose area is calculated by a data processor. The peak area is proportional to the carbon concentration of the sample. The instrument was calibrated with standards prepared using 4.8 mm stock solution of potassium hydrogen phthalate (Kanto Chemical Co., Inc., Japan). A typical calibration curve generated with these standards is shown in Figure A14.

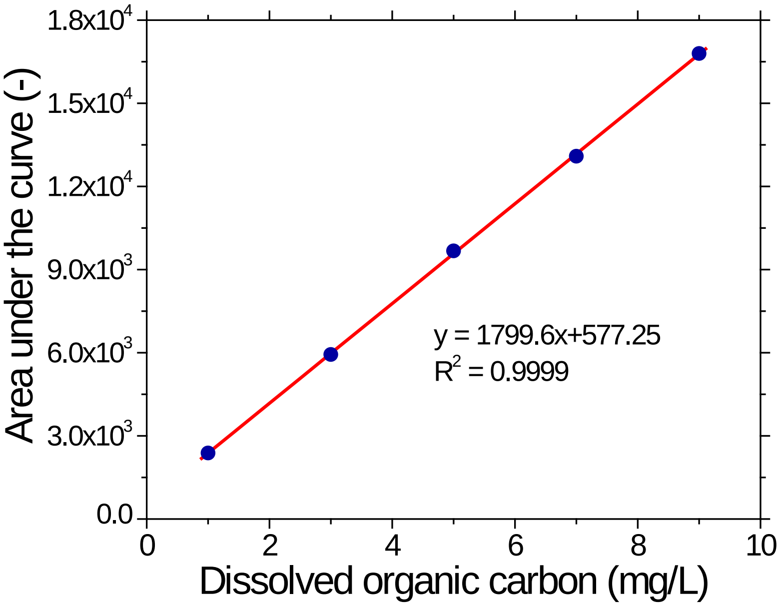


Figure A14.—Calibration curve of dissolved organic carbon measurement.

A.10. Pressure measurement and typical pressure profiles for dead-end microfiltration experiments

As reported in §4.3.5, the pressure on the feed suspension was continuously monitored using an analog pressure transducer (0.5-5.5 V, PX303-050G5V, Omega Engineering Company, Stamford, Connecticut). The pressure transducer was excited by a 24 volt (V) unregulated external power supply (U24Y101, Omega Engineering Company, Stamford, Connecticut). Prior to filtering Lake water samples, the pressure transducer was calibrated using a glycerin filled pressure gauge (Duralife® Pressure Gauge Type 1009, Ashcroft, Stratford, Connecticut), having a range of 0-30 psi and an accuracy of ±1%. Figure A15 depicts the pressure calibration curve for the pressure transducer. Typical pressure profiles obtained during constant pressure dead-end microfiltration experiment are depicted in Figure A16. Straight lines with zero slope were fitted to the experimental data points. As shown in Figure A16, the pressure did not change during each of the experiment, allowing us to have a quantitative comparison of the results obtained over the entire duration of the experiment.

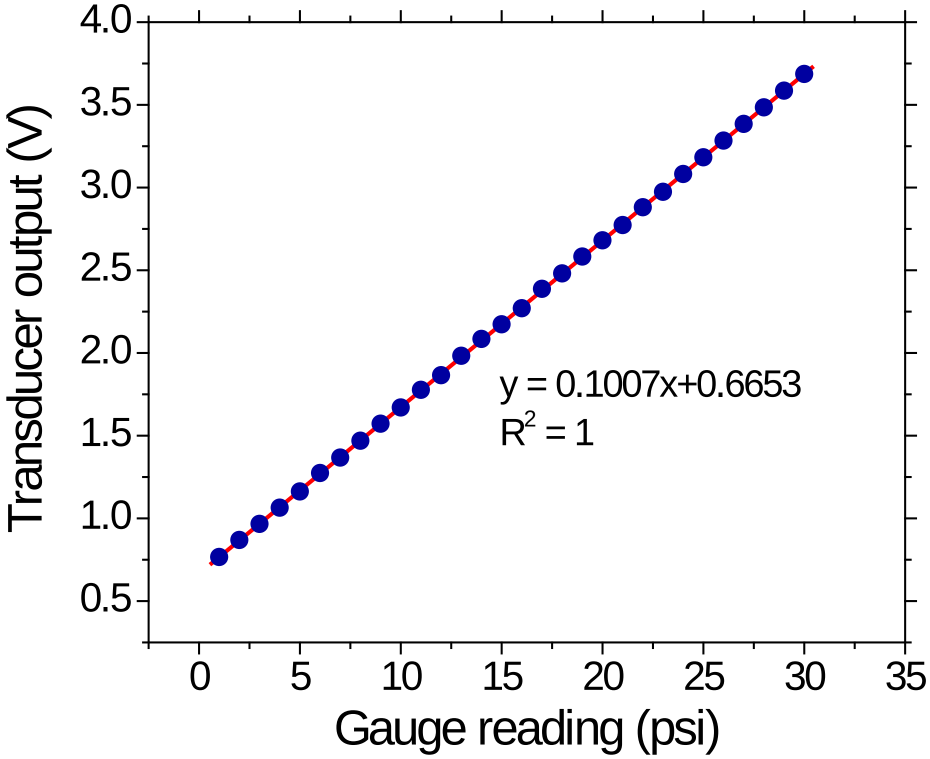


Figure A15.—Calibration curve of the pressure transducer.

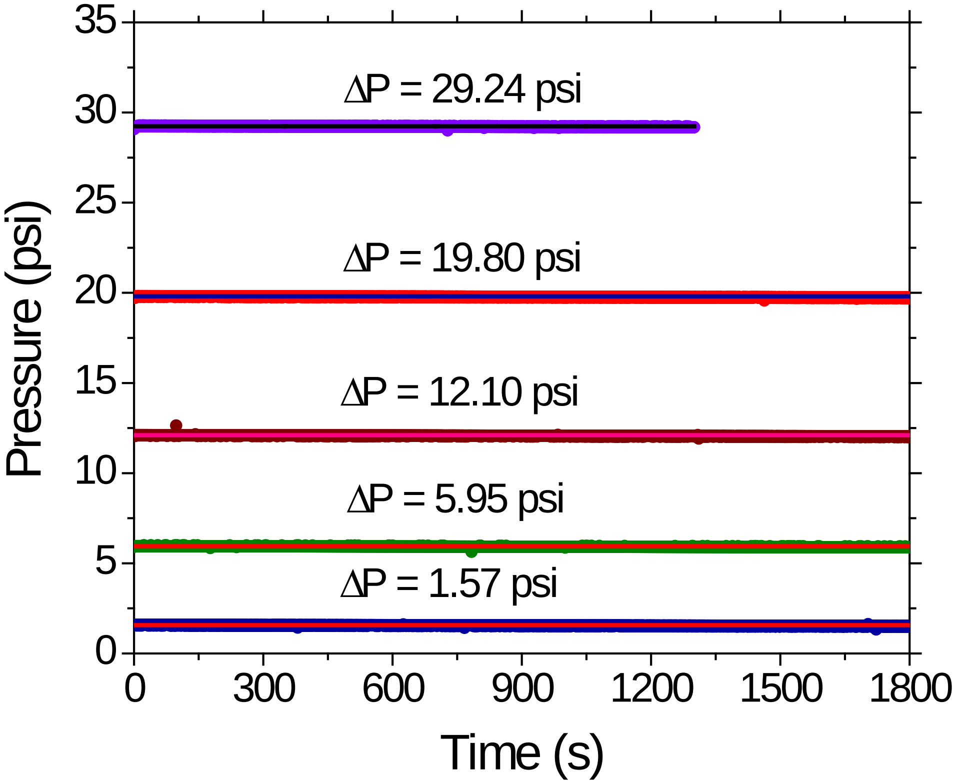
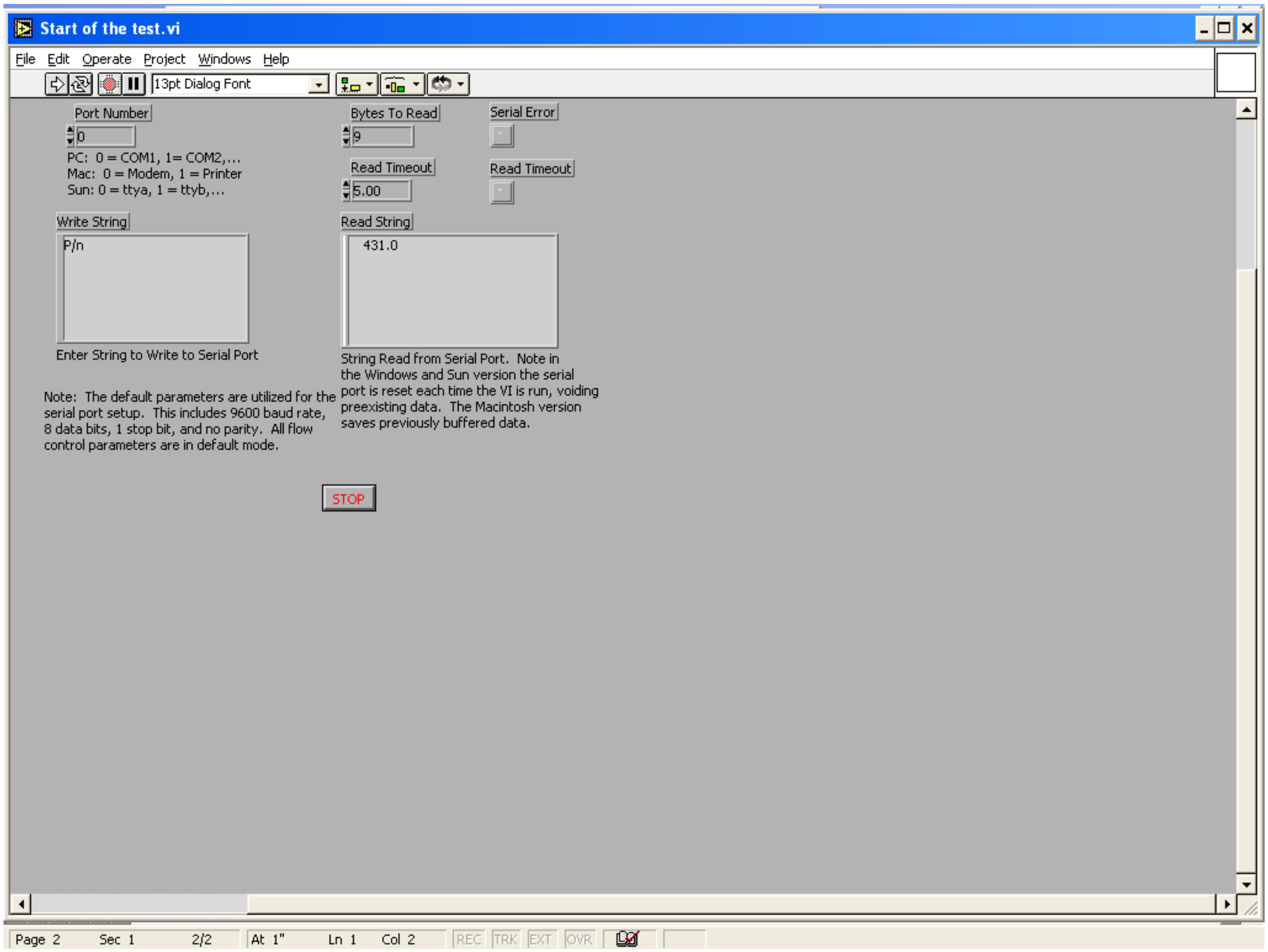


Figure A16.—Typical pressure profiles obtained during five different constant pressure, dead-end microfiltration experiments.

A.11. LabVIEW programming

Instrument control and data acquisition, during dead-end constant pressure microfiltration experiments, were achieved using a program written in LabVIEW (version 5.1, National Instruments, Austin, Texas). A screen picture of the operational windows and the circuit diagram of the program for this constant pressure experiment are shown in Figures A17 and A18.



Port Number: 0

PC: 0 = COM1, 1= COM2,...
Mac: 0 = Modem, 1 = Printer
Sun: 0 = ttya, 1 = ttyb,...

Bytes To Read: 9

Read Timeout: 5.00

Write String: P/n

Enter String to Write to Serial Port

Read String: 431.0

String Read from Serial Port. Note in the Windows and Sun version the serial port is reset each time the VI is run, voiding preexisting data. The Macintosh version saves previously buffered data.

Note: The default parameters are utilized for the serial port setup. This includes 9600 baud rate, 8 data bits, 1 stop bit, and no parity. All flow control parameters are in default mode.

Figure A17.—Screen picture of the operational windows of LabVIEW program.

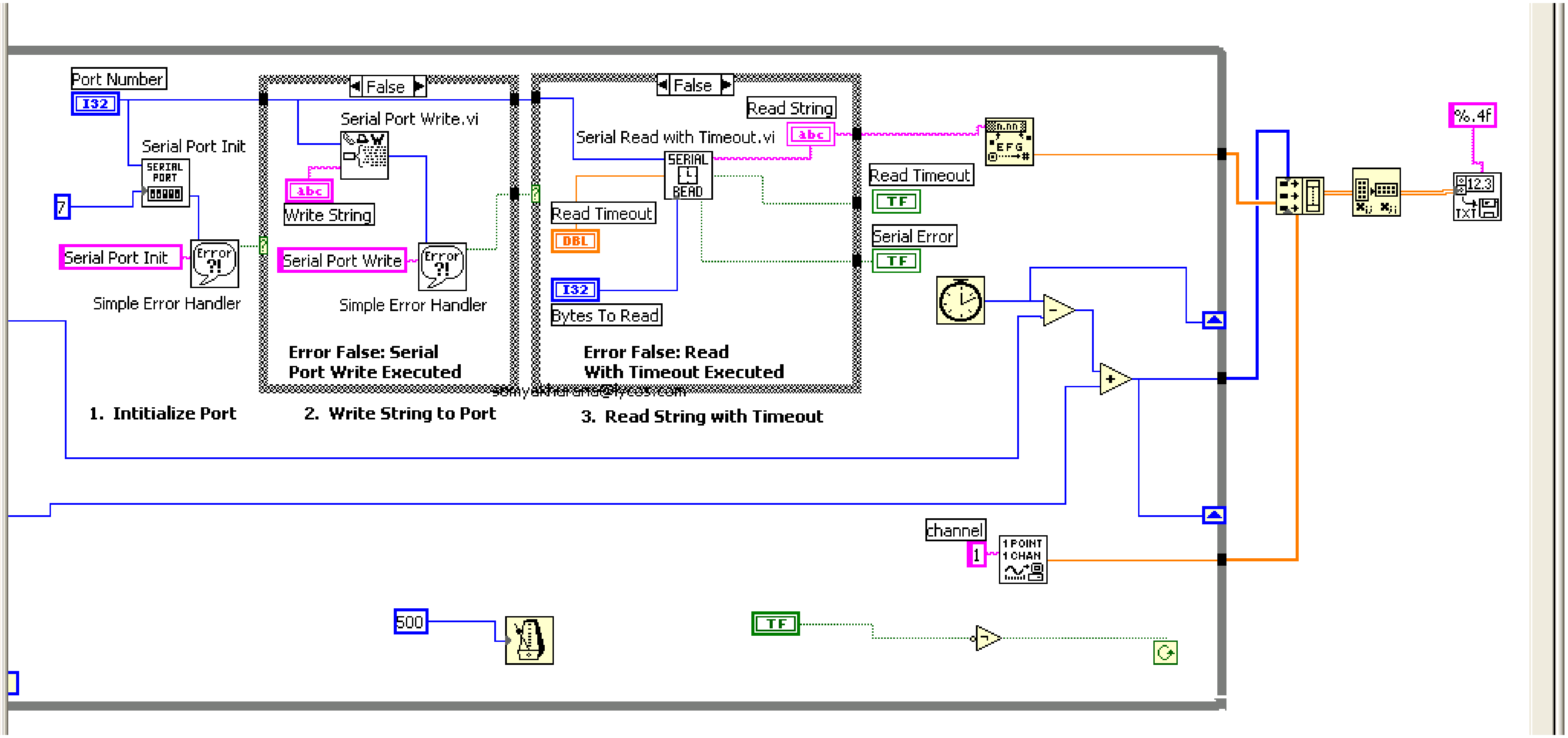


Figure A18.—Screen picture of the diagram view of LabVIEW program.

A.12. Record of Numerical Data

Figure A19.

Date	Turbidity (NTU)	Alkalinity (mg/L as $CaCO_3$)	pH (-)	DOC (mg/L)	UV_{254} (cm^{-1})	TDS (mg/L)	Ca Hardness (mg/L as $CaCO_3$)	Total Hardness (mg/L as $CaCO_3$)
1/12/2006	41	76	6.59	5.37	0.133	153.12	44	--
1/12/2006	41	75.8	6.54	5.3	0.132	156.83	44	--
1/12/2006	42	76	6.6	5.35	0.133	151.73	42.4	55.2
1/19/2006	35	76	6.64	5.35	0.139	154.98	44	55.2
1/19/2006	35	75.2	6.62	5.32	0.139	155.9	44.6	55.2
1/25/2006	40	78.4	6.66	4.95	0.128	151.26	43.2	56
2/3/2006	43	81.2	6.54	5.05	0.142	151.26	43.6	54.7
2/10/2006	40	76.8	6.6	5.43	0.145	155.9	42.4	57.6
2/15/2006	30	76	6.71	5.07	0.145	150.8	41.6	56
2/17/2006	37	82.4	6.57	5.17	0.137	157.3	44	57.2
2/24/2006	27	72	7.61	5.13	0.137	154.98	42	55.2
3/3/2006	--	72.8	7.35	5.15	0.134	158.22	42	55.2
3/16/2006	28	78	7.5	5.34	--	152.19	46	58.8
3/23/2006	46	68	7.77	5.47	0.117	155.9	43.6	55.2
3/31/2006	39	64.8	7.83	5.25	0.117	154.51	41.6	56
4/7/2006	35	77	7.65	5.3	0.115	153.58	44	57.5
4/14/2006	38	64	7.76	4.7	0.124	155.44	41.6	50.4
4/15/2006	34	69	7.65	4.89	0.121	155.44	46	61.2
4/19/2006	36	66.4	7.85	4.86	0.123	155.9	41	57.6
5/1/2006	35	69.2	7.65	4.85	0.126	153.12	42	57.2
5/18/2006	37	68	7.57	4.87	0.128	148.94	43.2	56
5/27/2006	45	73.2	7.85	4.83	0.114	154.51	44	54.4
6/8/2006	42	71.2	7.53	4.79	0.13	154.05	41.6	52.8
6/18/2006	47	75.6	7.49	4.86	0.121	154.05	42.4	57.2
6/23/2006	33	67.2	7.42	4.56	0.122	153.12	44	52.8
6/30/2006	31	76	7.59	4.53	0.121	147.55	48.4	58
7/6/2006	36	68	7.34	4.79	0.122	148.48	45.6	52
7/7/2006	12.6	58.8	7.41	5.46	0.183	111.82	32.4	38.8
7/13/2006	11	56.8	7.41	5.52	0.17	108.58	33.2	38.8
7/21/2006	12.6	56.8	7.5	5.56	0.182	100.69	34.4	40
8/4/2006	16.5	56	7.48	5.49	0.184	100.22	30.4	39.2
8/11/2006	18	54	7.29	5.53	0.184	101.62	31.8	38
8/20/2006	15	58.4	7.34	4.95	0.176	99.76	28	40.8
8/25/2006	13	54	7.29	5.26	0.185	101.62	31.6	38
9/1/2006	19	55.6	7.37		0.183	102.54	29.2	38.4
9/8/2006	15	53.8	7.32		0.181	99.76	30.8	39.2
9/15/2006	16	54.9	7.25	5.06	0.182	99.296	32.4	37.5
9/22/2006	14.7	60	7.31	4.96	0.18	97.904	31.2	40
9/29/2006	16	56.8	7.32	4.94	0.177	99.76	29.6	41.6
10/6/2006	12	56	7.35	5.02	0.178	98.368	31.8	40
10/13/2006	13	56.4	7.36	4.98	0.174	99.296	32.6	41.2
10/22/2006	10	58.2	7.45	5.22	0.182	98.368	31	37.8
10/27/2006	12.8	56	7.36	5.4	0.18	100.69	32	43.2
11/4/2006	12	61	7.29	5.31	0.18	100.22	29.8	45.2
11/10/2006	12	56.2	7.44	5.12	0.175	97.904	31.6	42.6
11/18/2006	11	59.2	7.35		0.181	100.69	31	44.8
11/24/2006	12	57.2	7.4	5.25	0.179	99.296	31	43.8
12/2/2006	11.2	57.9	7.41		0.179	99.76	32.5	43.5
12/8/2006	10.8	56	7.47	5.32	0.179	98.832	32	44
12/15/2007	13	56.8	7.35	5.15	0.178	101.15	31.2	43.2
12/22/2007	12.8	58	7.42	5.28	0.18	100.69	31.6	42
12/29/2007	12	59.2	7.45	5.3	0.182	99.76	31.2	41.6
1/5/2007	13.2	57.6	7.55	5.2	0.179	100.69	31.2	42.4
1/12/2007	13.5	56.8	7.45	5.2	0.183	98.832	32	44
1/19/2007	12.8	58	7.5	5.3	0.182	99.76	32.5	44
1/26/2007	13.1	59.6	7.44	5.41	0.173	99.296	33.2	41.6
2/3/2007	14.1	60.4	7.59	5.6	0.171	97.904	32.8	40.8
2/10/2007	14.5	59.2	7.51	5.54	0.175	99.76	31.6	43.8
2/17/2007	13.7	56.4	7.43	5.35	0.177	100.69	34	39.2
2/25/2007	12.5	58.8	7.49	5.9	0.176	103.47	34.8	42.8
3/3/2007	13.9	57.3	7.41	5.48	0.179	100.69	33.4	41.8
3/11/2007	12.5	56.8	7.55	5.45	0.173	102.54	33.6	44
3/17/2007	12.7	60	7.46	5.53	0.177	99.76	32.5	43.7
3/30/2007	12.6	58.6	7.44	5.43	0.18	101.15	32.9	43.2
4/6/2007	13	56	7.29	5.5	0.185	101.15	32.8	44
4/13/2007	13.1	60	7.36	5.4	0.178	102.08	33.6	44
4/20/2007	13	58	7.38	5.46	0.18	102.54	33	44
4/27/2007	13.2	58.4	7.42	5.51	0.182	103.01	32.2	43.8
5/4/2007	12.8	56	7.5	5.68	0.184	103.01	31.8	44
5/11/2007	13.1	57	7.43	5.6	0.184	101.62	33.2	43.2
5/18/2007	12.9	60	7.58	5.58	0.187	100.22	32.6	43.6
5/25/2007	13.3	59	7.5	5.52	0.185	100.69	33	44
6/1/2007	13	57.4	7.54	5.6	0.183	103.01	33.2	44
6/8/2007	13.2	57.4	7.48	5.5	0.184	102.08	32.9	43.6
6/15/2007	13	58	7.5	5.56	0.186	101.62	33.1	44
6/22/2007	12.9	57	7.45	5.48	0.181	102.08	32	43
6/29/2007	13.1	57.6	7.48	5.6	0.186	103.01	33.6	43.2

Figure A20.

Fe^{3+} = 0 mg/L		Fe^{3+} = 2 mg/L		Fe^{3+} = 5 mg/L		Fe^{3+} = 10 mg/L		Fe^{3+} = 15 mg/L	
V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0
0.00611	0.81484	0.00574	0.72412	0.00122	0.74675	0.01955	0.84879	0.0143	0.85537
0.00697	0.81226	0.011	0.65237	0.01588	0.59432	0.0281	0.81464	0.01955	0.84947
0.00722	0.81392	0.01552	0.59446	0.02688	0.48878	0.03533	0.7694	0.02688	0.79963
0.0073	0.81442	0.02041	0.56394	0.03665	0.43128	0.04277	0.74799	0.03299	0.81308
0.00732	0.75121	0.02529	0.50507	0.04521	0.39858	0.04888	0.68931	0.04032	0.80031
0.011	0.65177	0.03177	0.47916	0.05253	0.34534	0.05498	0.67206	0.04643	0.77305
0.02041	0.59306	0.04031	0.41508	0.05865	0.31619	0.06109	0.63577	0.05254	0.77324
0.03018	0.45961	0.0501	0.36206	0.06476	0.27772	0.0672	0.60129	0.05865	0.76404
0.04021	0.42681	0.05584	0.32094	0.07087	0.2733	0.0721	0.57395	0.06475	0.7487
0.05009	0.35844	0.06073	0.30018	0.07576	0.25333	0.07698	0.54358	0.07087	0.7358
0.06073	0.31085	0.06843	0.26564	0.08187	0.25233	0.08306	0.54368	0.07576	0.72553
0.07051	0.28335	0.08309	0.22912	0.08639	0.2322	0.08676	0.51998	0.08187	0.71335
0.08028	0.24296	0.09653	0.20366	0.09042	0.22217	0.09165	0.49876	0.08676	0.69151
0.09006	0.21886	0.10509	0.18545	0.09531	0.21034	0.09652	0.47253	0.09286	0.68089
0.10753	0.17882	0.11609	0.17507	0.09898	0.21002	0.10106	0.46319	0.09776	0.67088
0.13319	0.14574	0.12342	0.16371	0.1035	0.20062	0.10509	0.44006	0.10264	0.65289
0.15397	0.12551	0.12953	0.15086	0.10631	0.18753	0.10875	0.43038	0.10753	0.65142
0.1723	0.10763	0.13686	0.14418	0.11083	0.18569	0.11242	0.42954	0.11353	0.64151
0.1894	0.09861	0.14297	0.13462	0.11486	0.1764	0.11609	0.42187	0.11731	0.62229
0.20407	0.09238	0.14908	0.13409	0.11853	0.17295	0.11975	0.40017	0.1222	0.61377
0.21873	0.08764	0.15519	0.12949	0.12219	0.16407	0.12342	0.38729	0.12953	0.6016
0.23217	0.08276	0.16127	0.12427	0.12464	0.16392	0.1283	0.38196	0.13441	0.58375
0.24561	0.07993	0.16619	0.11993	0.12831	0.1613	0.1316	0.37633	0.13808	0.58625
0.25783	0.07629	0.17107	0.11638	0.13186	0.15395	0.13442	0.37692	0.14382	0.56101
0.26883	0.07345	0.17596	0.11414	0.13442	0.15568	0.13808	0.36778	0.14786	0.55667
0.28105	0.06973	0.18207	0.10966	0.13686	0.14981	0.14171	0.35489	0.15152	0.55684
0.28338	0.06945	0.18696	0.10519	0.14052	0.14687	0.14297	0.34925	0.15638	0.53997
0.28349	0.07043	0.19184	0.10505	0.14297	0.14393	0.14663	0.348	0.16093	0.54016
0.28349	0.06935	0.19551	0.10076	0.14663	0.14032	0.1503	0.34444	0.16496	0.52735
0.28349	0.06926	0.2004	0.10078	0.14908	0.13897	0.15274	0.34137	0.16863	0.51693
0.28349	0.06926	0.20529	0.09658	0.15152	0.13704	0.15638	0.32299	0.1723	0.51621
0.28468	0.06917	0.20895	0.09448	0.15397	0.13577	0.15885	0.32738	0.17718	0.50555
0.28472	0.06884	0.21262	0.09328	0.15641	0.13173	0.1613	0.32453	0.18085	0.49849
0.28472	0.06907	0.21751	0.09142	0.15885	0.1282	0.16496	0.31791	0.18452	0.48799
0.28472	0.06907	0.22117	0.08963	0.16252	0.12706	0.16741	0.30878	0.18695	0.48565
0.28472	0.06907	0.22484	0.08978	0.16496	0.1254	0.16985	0.31223	0.18818	0.4817
0.28472	0.06898	0.22606	0.08811	0.16741	0.12378	0.17352	0.30789	0.19026	0.48102
0.28594	0.06789	0.22728	0.08925	0.16985	0.12168	0.17596	0.30549	0.19185	0.47634
0.28594	0.06889	0.22962	0.08778	0.1723	0.12015	0.17841	0.30157	0.19392	0.47896
0.28594	0.06889	0.23095	0.08763	0.17474	0.12023	0.18085	0.29755	0.19551	0.47209
0.28594	0.0688	0.23217	0.08601	0.17718	0.11764	0.18452	0.291	0.19673	0.46766
0.28594	0.0688	0.23339	0.08601	0.17963	0.11806	0.18696	0.28982	0.19918	0.46766
0.28594	0.0678	0.23462	0.08601	0.18207	0.11545	0.1894	0.28591	0.2004	0.4642
0.28716	0.06761	0.23584	0.08439	0.18329	0.11478	0.19185	0.28317	0.20284	0.46361
0.28716	0.0687	0.23706	0.08626	0.18574	0.11347	0.19429	0.28063	0.20407	0.45097
0.28716	0.06861	0.23828	0.08306	0.18818	0.11113	0.19673	0.27426	0.20651	0.45037
0.28716	0.06861	0.2395	0.08306	0.19063	0.10925	0.19918	0.27026	0.20773	0.45325
0.28716	0.06861	0.24195	0.08242	0.19307	0.10984	0.20162	0.26772	0.21018	0.44674
0.28827	0.06743	0.24317	0.0829	0.19429	0.10752	0.20407	0.26523	0.2114	0.44316
0.28838	0.06743	0.24439	0.08257	0.19673	0.10692	0.20651	0.264	0.21381	0.44258
0.28838	0.06843	0.24561	0.08225	0.19918	0.10633	0.20895	0.25904	0.21506	0.43965
0.28838	0.06843	0.24684	0.08146	0.2004	0.10407	0.2114	0.25784	0.21629	0.43318
0.28838	0.06834	0.24806	0.08146	0.20284	0.10239	0.21384	0.25821	0.21872	0.43618
0.28838	0.06735	0.24928	0.08041	0.20529	0.10071	0.21629	0.25697	0.21995	0.42978
0.2896	0.06725	0.2505	0.07998	0.20651	0.10071	0.21873	0.25232	0.22117	0.43473
0.2896	0.06716	0.25172	0.0785	0.20895	0.09903	0.21995	0.25011	0.2224	0.43438
0.2896	0.06825	0.25294	0.08001	0.2114	0.09735	0.2224	0.24902	0.22484	0.43082
0.2896	0.06816	0.25417	0.0786	0.21384	0.09735	0.22362	0.24902	0.22606	0.43082
0.2896	0.06816	0.25539	0.07866	0.21506	0.09568	0.22606	0.2458	0.22728	0.4269
0.2896	0.06699	0.25661	0.07866	0.21751	0.09567	0.22851	0.2437	0.22973	0.42309
0.29083	0.06699	0.25783	0.07739	0.21873	0.09567	0.23092	0.24162	0.23095	0.42655
0.29083	0.06798	0.25905	0.07746	0.22117	0.094	0.23217	0.23618	0.23338	0.42023
0.29083	0.06798	0.25991	0.07752	0.2224	0.09362	0.23462	0.23747	0.23462	0.41907
0.29083	0.06713	0.26028	0.07759	0.22484	0.09326	0.23706	0.23556	0.23584	0.41516
0.29083	0.06681	0.2615	0.07637	0.22606	0.09311	0.23828	0.23368	0.23817	0.41515
0.29205	0.06681	0.26272	0.07645	0.22851	0.09165	0.24073	0.23528	0.2395	0.41124
0.29205	0.06672	0.26394	0.07653	0.22973	0.09004	0.24317	0.23241	0.24073	0.40849
0.29205	0.0678	0.26516	0.07626	0.23217	0.09004	0.24558	0.23241	0.24195	0.40732
0.29205	0.06771	0.26639	0.07581	0.23339	0.08976	0.24684	0.22867	0.24317	0.40341
0.29205	0.06663	0.26761	0.07608	0.23584	0.08976	0.24928	0.22736	0.24561	0.39949
0.29205	0.06655	0.26883	0.075	0.23706	0.08818	0.25172	0.22603	0.24684	0.39949
0.29327	0.06655	0.27005	0.07509	0.2395	0.08818	0.25417	0.22537	0.24806	0.39761
0.29327	0.06655	0.27127	0.07406	0.24073	0.08661	0.25539	0.22344	0.24928	0.39757
0.29327	0.06637	0.27213	0.07365	0.24195	0.08614	0.25783	0.22049	0.25136	0.39568
0.29327	0.06637	0.2725	0.07325	0.24439	0.08661	0.25905	0.21986	0.25294	0.3918
0.29449	0.06628	0.27372	0.07406	0.24561	0.08503	0.26028	0.21993	0.25417	0.39346
0.29449	0.06628	0.27494	0.07355	0.24684	0.08346	0.26272	0.21862	0.25539	0.38994
0.29449	0.06735	0.27616	0.07344	0.24928	0.08346	0.26516	0.21739	0.25661	0.38663
0.29449	0.06621	0.27738	0.07324	0.2505	0.08303	0.26639	0.2145	0.25894	0.38976
0.29449	0.0662	0.27861	0.07304	0.25283	0.08261	0.26883	0.2139	0.26027	0.38828
0.29449	0.06611	0.27983	0.07171	0.25417	0.08309	0.27005	0.2133	0.2615	0.38448
0.29571	0.06611	0.28105	0.0705	0.25539	0.08116	0.2725	0.21212	0.26272	0.38269
0.29571	0.06611	0.28105	0.07132	0.25772	0.08075	0.27372	0.20929	0.26516	0.38269
0.29571	0.06709	0.28227	0.07113	0.25905	0.07888	0.27616	0.20938	0.26639	0.38099
0.29571	0.06594	0.28349	0.07093	0.2615	0.07976	0.2786	0.2059	0.26761	0.38058
0.29571	0.06594	0.28472	0.07074	0.26272	0.07937	0.27983	0.20368	0.26883	0.37916
0.29694	0.06585	0.28594	0.07055	0.26394	0.0786	0.28227	0.2059	0.27005	0.37542
0.29694	0.0648	0.28716	0.06917	0.26516	0.07984	0.28349	0.20366	0.27127	0.37369
0.29694	0.06577	0.28838	0.06898	0.26761	0.0798	0.28594	0.20142	0.27335	0.37221
0.29694	0.06577	0.2896	0.0688	0.26883	0.07855	0.28713	0.20142	0.27493	0.37227
0.29815	0.06568	0.29083	0.06861	0.27005	0.07911	0.28838	0.19918	0.27616	0.36732
0.29816	0.06568	0.29083	0.06952	0.27213	0.07789	0.2896	0.1992	0.27738	0.36731
0.29816	0.0659	0.29205	0.06933	0.27372	0.07789	0.29205	0.19918	0.27861	0.36929
0.29816	0.06559	0.29327	0.06915	0.27494	0.07668	0.29327	0.19695	0.27983	0.36568
0.29816	0.06551	0.29449	0.06896	0.27616	0.07668	0.29571	0.19695	0.28105	0.36407
0.29927	0.06542	0.29571	0.06878	0.27738	0.07546	0.29693	0.19471	0.28338	0.36406
0.29938	0.06542	0.29694	0.06753	0.27983	0.07546	0.29694	0.19491	0.28468	0.36444
0.29938	0.06525	0.29694	0.06735	0.28105	0.07547	0.29816	0.19695	0.28594	0.36444
0.3006	0.06517	0.29816	0.06718	0.28227	0.07547	0.29938	0.19695	0.28716	0.36285
0.3006	0.06517	0.29938	0.06594	0.28349	0.07428	0.3006	0.19471	0.28838	0.36285

Figure A21.

pH = 6.4		pH = 7.5		pH = 8.3	
V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0
0.01466	0.80649	0.01164	0.89827	0.01466	0.80443
0.01674	0.72326	0.02082	0.76089	0.02441	0.73982
0.02066	0.74557	0.03024	0.68799	0.02807	0.67308
0.02555	0.63419	0.03693	0.57419	0.03044	0.64404
0.03055	0.54382	0.04002	0.54712	0.03288	0.58127
0.03543	0.47922	0.04543	0.52358	0.0391	0.52819
0.04021	0.45258	0.05564	0.45633	0.04521	0.47212
0.04277	0.40732	0.0614	0.44099	0.05096	0.42356
0.04765	0.36035	0.06545	0.40349	0.05621	0.40141
0.05254	0.34382	0.07156	0.3598	0.06099	0.36662
0.05742	0.33115	0.07487	0.35208	0.06562	0.36276
0.06221	0.31741	0.07889	0.34716	0.06962	0.3251
0.06598	0.29005	0.08171	0.3234	0.07328	0.31088
0.07076	0.29231	0.08606	0.32083	0.07698	0.30562
0.07417	0.28531	0.0899	0.3059	0.08065	0.28853
0.07817	0.28528	0.0939	0.30133	0.08431	0.28076
0.08187	0.27154	0.09724	0.29885	0.08798	0.2764
0.08553	0.25967	0.1009	0.27976	0.09161	0.26269
0.0892	0.25887	0.10345	0.26021	0.09409	0.25236
0.09287	0.24791	0.10735	0.26064	0.09765	0.25236
0.09617	0.24793	0.11068	0.25969	0.1002	0.24436
0.09898	0.23905	0.11289	0.2555	0.10386	0.23327
0.10264	0.23222	0.11592	0.23496	0.10717	0.2376
0.10631	0.22428	0.11924	0.23344	0.10987	0.23147
0.10875	0.2248	0.12178	0.24313	0.11241	0.22028
0.11242	0.22473	0.12534	0.2188	0.11486	0.22217
0.11486	0.22217	0.12781	0.22146	0.11731	0.20935
0.11852	0.21614	0.13026	0.21635	0.11975	0.20668
0.12216	0.21563	0.1327	0.21558	0.1222	0.20832
0.12464	0.20073	0.13318	0.2165	0.12464	0.20098
0.12708	0.20366	0.13762	0.20835	0.12708	0.19857
0.12953	0.19687	0.14011	0.20813	0.12953	0.1959
0.13308	0.19869	0.14281	0.2018	0.13197	0.18851
0.13563	0.19869	0.14527	0.20163	0.13442	0.19118
0.13808	0.19576	0.14745	0.19696	0.13686	0.17991
0.14052	0.19304	0.14989	0.19555	0.14175	0.18703
0.14297	0.18301	0.15226	0.19513	0.14419	0.1808
0.14541	0.18837	0.15273	0.18956	0.14627	0.17718
0.14908	0.18464	0.15623	0.19018	0.15029	0.16972
0.15152	0.18451	0.15871	0.18776	0.15397	0.16663
0.15397	0.1809	0.16089	0.17914	0.15641	0.16566
0.15641	0.176	0.16325	0.17762	0.15885	0.16039
0.15885	0.1728	0.16577	0.17719	0.16093	0.16026
0.16127	0.17193	0.16729	0.1759	0.16252	0.15767
0.16338	0.1715	0.16945	0.17172	0.16495	0.16216
0.16619	0.17361	0.17103	0.16919	0.16863	0.15967
0.1686	0.16604	0.17415	0.173	0.16985	0.15509
0.17071	0.16643	0.17558	0.17667	0.1723	0.15502
0.1723	0.16547	0.178	0.16825	0.17352	0.15274
0.17474	0.16123	0.17951	0.16803	0.17596	0.15274
0.17718	0.15875	0.18167	0.16413	0.17718	0.14821
0.17962	0.15806	0.18317	0.16328	0.17962	0.14546
0.18085	0.15806	0.18533	0.15798	0.18085	0.14821
0.18451	0.15574	0.18684	0.16173	0.18329	0.14678
0.18574	0.15178	0.18867	0.15794	0.18696	0.14237
0.18818	0.15325	0.1905	0.15752	0.18818	0.141
0.19062	0.15325	0.19378	0.15352	0.19062	0.141
0.19185	0.15056	0.19513	0.15384	0.19307	0.13834
0.19429	0.14909	0.19661	0.15277	0.19551	0.13632
0.19673	0.15027	0.19903	0.15168	0.19673	0.13703
0.19915	0.14764	0.20029	0.15182	0.19796	0.13504
0.2004	0.14439	0.20244	0.14939	0.2004	0.13295
0.20283	0.14622	0.20369	0.14868	0.20162	0.13412
0.20407	0.14455	0.2061	0.14839	0.20529	0.13091
0.20651	0.14319	0.20735	0.14838	0.20651	0.13163
0.20773	0.14312	0.20883	0.1481	0.20773	0.13057
0.21018	0.13934	0.211	0.14773	0.21017	0.12782
0.21262	0.14178	0.2125	0.14412	0.2114	0.12708
0.21495	0.13803	0.21466	0.13977	0.21384	0.1285
0.21629	0.13886	0.21591	0.13966	0.21506	0.12749
0.21862	0.13666	0.21628	0.13965	0.21629	0.12749
0.21995	0.13646	0.21978	0.13846	0.21751	0.12635
0.22229	0.13555	0.22106	0.13629	0.21994	0.12631
0.22362	0.13119	0.22322	0.13531	0.22117	0.12484
0.22603	0.1323	0.22446	0.13251	0.2224	0.12484
0.22728	0.13013	0.22594	0.13311	0.22484	0.12234
0.22851	0.13061	0.22725	0.13431	0.22973	0.12043
0.23095	0.12809	0.22933	0.13138	0.23095	0.12043
0.23217	0.12809	0.23083	0.13131	0.23217	0.11999
0.23462	0.12912	0.23336	0.13053	0.23584	0.1175
0.23706	0.12783	0.23544	0.12994	0.23706	0.11603
0.23828	0.12782	0.23694	0.12975	0.23828	0.11547
0.2395	0.12651	0.23817	0.1278	0.2395	0.1165
0.24195	0.12521	0.23947	0.12717	0.24073	0.11637
0.24317	0.12522	0.24155	0.12503	0.2428	0.11479
0.24558	0.12429	0.24194	0.12439	0.24428	0.11438
0.24684	0.1226	0.24417	0.12478	0.24561	0.1133
0.24806	0.12169	0.2455	0.12479	0.24684	0.1133
0.2505	0.1213	0.24888	0.12221	0.24806	0.11277
0.25172	0.11999	0.25038	0.12161	0.24928	0.11135
0.25294	0.11872	0.25291	0.11947	0.25171	0.11083
0.25539	0.11869	0.25499	0.11821	0.25294	0.11032
0.25661	0.11738	0.25771	0.11586	0.25417	0.11032
0.25783	0.11812	0.2578	0.11571	0.25539	0.10981
0.25991	0.11682	0.25902	0.11625	0.25905	0.10811
0.2615	0.11627	0.26024	0.11567	0.26028	0.10644
0.26272	0.11497	0.26232	0.11551	0.2615	0.10774
0.26394	0.1156	0.26357	0.11398	0.26272	0.1059
0.26516	0.11425	0.26479	0.11348	0.26516	0.10543
0.26516	0.11452	0.26601	0.11347	0.26761	0.10451

Figure A22.

ΔP = 13.79 KPa		ΔP = 41.37 KPa		ΔP = 82.74 KPa		ΔP = 206.84 KPa	
V/A (m^3/m^2)	J/J$_0$	V/A (m^3/m^2)	J/J$_0$	V/A (m^3/m^2)	J/J$_0$	V/A (m^3/m^2)	J/J$_0$
0.00243	0.9584	0.01222	0.8791	0.00121	0.85668	0.00232	0.70245
0.00611	0.93275	0.01577	0.82755	0.00355	0.79963	0.00692	0.63849
0.00855	0.88868	0.02041	0.80058	0.00597	0.74735	0.01059	0.57205
0.01466	0.86839	0.02929	0.74421	0.01062	0.69814	0.01295	0.50188
0.01833	0.85989	0.03665	0.66765	0.01577	0.64965	0.01425	0.41179
0.02199	0.85348	0.04388	0.63082	0.02041	0.5908	0.01551	0.37982
0.02677	0.81462	0.05009	0.58422	0.02554	0.52953	0.01944	0.32824
0.03055	0.81461	0.0561	0.55205	0.03018	0.44787	0.02921	0.26005
0.03421	0.80246	0.06106	0.53089	0.03407	0.41325	0.03507	0.21016
0.03909	0.79609	0.06684	0.5049	0.03996	0.36186	0.03776	0.19366
0.04276	0.78606	0.07209	0.4806	0.04632	0.31469	0.04485	0.17684
0.04643	0.77605	0.07662	0.47231	0.05131	0.28578	0.05121	0.15773
0.05095	0.77322	0.0815	0.46663	0.05621	0.27799	0.05729	0.14067
0.05498	0.7607	0.08554	0.44756	0.0611	0.25634	0.06221	0.13603
0.05829	0.76044	0.09031	0.43083	0.06598	0.24786	0.06684	0.12534
0.06195	0.76119	0.09409	0.40888	0.07087	0.23622	0.07206	0.1195
0.06588	0.74779	0.09861	0.40919	0.07454	0.22932	0.07576	0.11786
0.06965	0.7358	0.10142	0.38977	0.07821	0.21841	0.08064	0.11558
0.07321	0.72412	0.1062	0.38973	0.08187	0.21556	0.08543	0.10799
0.07576	0.71395	0.10961	0.39118	0.08673	0.20543	0.08919	0.10878
0.08064	0.71281	0.11242	0.37253	0.09039	0.19404	0.09287	0.10064
0.08395	0.71434	0.11727	0.37142	0.09287	0.18429	0.09653	0.09822
0.08676	0.69459	0.11975	0.36699	0.09653	0.19117	0.10106	0.09956
0.09042	0.67896	0.12341	0.36368	0.1002	0.17706	0.10387	0.0931
0.09398	0.68056	0.12697	0.36207	0.10376	0.17476	0.10753	0.09194
0.09772	0.67195	0.13072	0.3623	0.10631	0.17391	0.1112	0.08919
0.1002	0.65998	0.13319	0.34904	0.10997	0.16704	0.11486	0.08517
0.10386	0.66115	0.13649	0.3407	0.11242	0.16703	0.11842	0.085
0.10631	0.64007	0.1393	0.33855	0.11486	0.16757	0.12097	0.08501
0.10997	0.64039	0.14294	0.33852	0.11853	0.16059	0.12464	0.08155
0.11363	0.64244	0.14541	0.33794	0.12097	0.15558	0.1282	0.08138
0.11609	0.62303	0.14897	0.32767	0.12342	0.15512	0.13075	0.07869
0.11972	0.61899	0.15152	0.32162	0.12708	0.15065	0.13441	0.07845
0.1222	0.61377	0.15518	0.31513	0.12953	0.15049	0.13797	0.07592
0.12464	0.6027	0.15763	0.31427	0.13197	0.14297	0.14052	0.07863
0.12831	0.61376	0.16008	0.31421	0.13442	0.14283	0.14297	0.07406
0.13075	0.59447	0.16252	0.30731	0.13686	0.1433	0.14541	0.0721
0.13441	0.58375	0.16496	0.30656	0.14052	0.13919	0.14907	0.07188
0.13686	0.5757	0.16863	0.3061	0.14297	0.13794	0.15152	0.07289
0.14041	0.57854	0.17107	0.298	0.14541	0.13537	0.15482	0.0723
0.14297	0.57879	0.17352	0.2958	0.14785	0.13096	0.15752	0.06926
0.14541	0.56101	0.17596	0.29532	0.15027	0.13323	0.16007	0.06924
0.14786	0.55448	0.17841	0.29213	0.15263	0.12901	0.16252	0.06954
0.1503	0.55354	0.18085	0.28502	0.15518	0.12994	0.16496	0.06953
0.15274	0.55592	0.18452	0.28482	0.1576	0.12994	0.16741	0.06837
0.15519	0.54988	0.18696	0.2839	0.16007	0.12903	0.16985	0.0682
0.15763	0.54061	0.1894	0.27801	0.16215	0.12523	0.17352	0.06714
0.1613	0.53304	0.19185	0.27925	0.16374	0.12469	0.17596	0.06677
0.16374	0.52656	0.19429	0.27873	0.16619	0.12469	0.17841	0.0665
0.16619	0.52653	0.19673	0.2695	0.16863	0.12251	0.18085	0.06485
0.16863	0.51623	0.19918	0.2695	0.17106	0.12568	0.18329	0.06403
0.17107	0.51621	0.20162	0.26889	0.17352	0.12284	0.18574	0.06358
0.17352	0.51621	0.20406	0.26328	0.1756	0.1219	0.18818	0.06336
0.17352	0.51056	0.20651	0.25954	0.17718	0.12061	0.19063	0.06486
0.17718	0.51114	0.20895	0.25954	0.17963	0.12061	0.19307	0.06369
0.17962	0.50316	0.2114	0.2617	0.18207	0.11646	0.19551	0.06368
0.18207	0.49806	0.21384	0.25803	0.18415	0.11638	0.19795	0.06288
0.18451	0.48565	0.21629	0.25782	0.18574	0.11447	0.2004	0.06145
0.18696	0.48565	0.21862	0.25778	0.18815	0.11447	0.20284	0.06143
0.1894	0.48102	0.21995	0.25533	0.19062	0.11544	0.20529	0.06143
0.19148	0.47649	0.2224	0.25533	0.19296	0.11544	0.20773	0.06044
0.19307	0.47642	0.22484	0.25159	0.19429	0.11456	0.21017	0.0611
0.19551	0.47203	0.22728	0.25113	0.19673	0.11287	0.21259	0.06098
0.19796	0.46772	0.22973	0.24889	0.19796	0.11011	0.21506	0.06016
0.2004	0.4654	0.23217	0.24879	0.2004	0.10943	0.21748	0.05978
0.20284	0.4636	0.23451	0.24543	0.20281	0.1095	0.21873	0.0584
0.20526	0.45037	0.23584	0.24543	0.20407	0.11038	0.22117	0.05847
0.20772	0.45037	0.23828	0.24305	0.20651	0.11032	0.22362	0.0587
0.20895	0.44674	0.24072	0.24305	0.20773	0.10889	0.22606	0.0587
0.2114	0.44316	0.24317	0.24404	0.21103	0.10889	0.22847	0.05881
0.21384	0.44316	0.24561	0.24168	0.21262	0.10716	0.23058	0.05807
0.21628	0.43947	0.24769	0.23835	0.2147	0.10717	0.23339	0.05807
0.21751	0.43426	0.24928	0.2394	0.21629	0.10611	0.23573	0.05805
0.21995	0.42977	0.25172	0.23607	0.21862	0.10611	0.23706	0.05726
0.22236	0.43473	0.25417	0.23385	0.21995	0.10677	0.2395	0.05671
0.22362	0.43082	0.25539	0.23385	0.22362	0.10575	0.24195	0.05639
0.22606	0.43081	0.25783	0.23175	0.2257	0.10524	0.24436	0.05695
0.2284	0.4269	0.26024	0.23146	0.22728	0.10474	0.24561	0.05639
0.23094	0.4269	0.26272	0.23275	0.22969	0.10424	0.24806	0.0564
0.23217	0.41907	0.26513	0.22952	0.23181	0.10212	0.2505	0.05581
0.23462	0.41906	0.26639	0.22793	0.23339	0.10374	0.25294	0.05532
0.23669	0.41515	0.26882	0.22992	0.23573	0.10115	0.25528	0.05534
0.23828	0.41512	0.27005	0.22906	0.23706	0.10163	0.25661	0.05534
0.24073	0.41124	0.2725	0.22708	0.23828	0.10023	0.25905	0.05558
0.24195	0.40732	0.2725	0.22708	0.24073	0.10182	0.2615	0.05459
0.24439	0.40729	0.2725	0.22708	0.24195	0.10206	0.26272	0.05431
0.24561	0.39949	0.2725	0.22708	0.24439	0.10077	0.26516	0.05497
0.24806	0.40144	0.2725	0.22708	0.24561	0.10076	0.2676	0.05366
0.24928	0.39757	0.2725	0.22708	0.24805	0.09996	0.27005	0.05431
0.25172	0.39533	0.2725	0.22708	0.24928	0.09995	0.27246	0.05425
0.25294	0.38995	0.2725	0.22708	0.25161	0.09919	0.27249	0.05429
0.25539	0.38992	0.2725	0.22509	0.25294	0.09915	0.27249	0.0543
0.25747	0.38628	0.2725	0.22648	0.25539	0.09975	0.2725	0.05384
0.25905	0.38562	0.27335	0.2269	0.25661	0.09837	0.2725	0.05371
0.26147	0.38828	0.27361	0.22702	0.25905	0.09756	0.2725	0.05365
0.26272	0.38269	0.27369	0.22806	0.26028	0.09759	0.2725	0.05365
0.26516	0.38205	0.27371	0.22609	0.26261	0.09819	0.2725	0.05365
0.2675	0.38092	0.27372	0.2255	0.26394	0.09842	0.2725	0.05365
0.26883	0.37913	0.27372	0.22532	0.26516	0.09738	0.27335	0.05365
0.27005	0.37402	0.27372	0.22527	0.26761	0.09819	0.27361	0.05365

Figure A23.

pH = 6.4	
Fe^{3+} dosage (mg/L)	Specific Cake Resistance (m/Kg)
0	1.33E+15
5	2.18E+14
10	4.52E+13
15	1.94E+13

pH = 7.5	
Fe^{3+} dosage (mg/L)	Specific Cake Resistance (m/Kg)
0	1.20E+15
5	1.63E+14
10	4.81E+13
15	4.31E+13

pH = 8.3	
Fe^{3+} dosage (mg/L)	Specific Cake Resistance (m/Kg)
0	1.13E+15
2	6.32E+14
5	3.71E+14
10	8.77E+13
15	3.77E+13

Figure A24.

pH = 6.4	
Fe^{3+} dosage (mg/L)	Number weighted mean particle diameter (μm)
0	3.06
5	3.69
10	4.35
15	4.7

pH = 7.5	
Fe^{3+} dosage (mg/L)	Number weighted mean particle diameter (μm)
0	2.96
5	3.79
10	4.22
15	4.56

pH = 8.3	
Fe^{3+} dosage (mg/L)	Number weighted mean particle diameter (μm)
0	2.89
1	3.04
2	3.18
5	3.51
10	4.56
15	4.81

Figure A25.

Fe^{3+} = 0 mg/L	
log ΔP (ΔP in KPa)	log α^* (α^* in m/Kg)
1.64	14.91
1.94	15.08
2.15	15.14
2.32	15.20

Fe^{3+} = 5 mg/L	
log ΔP (ΔP in KPa)	log α^* (α^* in m/Kg)
1.62	14.21
1.94	14.33
2.15	14.53
2.22	14.53
2.32	14.68

Fe^{3+} = 10 mg/L	
log ΔP (ΔP in KPa)	log α^* (α^* in m/Kg)
1.65	13.68
2.16	14.05
2.23	14.13
2.32	14.21

Figure A26.

Fe (total) = 0 mg/L		Fe (total) = 7.4 mg/L		Fe (total) = 13.9 mg/L		Fe (total) = 21.2 mg/L	
V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0
0.00114	0.901	0.00303	0.95443	0.01071	0.97826	0	0.99512
0.01003	0.78471	0.00513	0.91648	0.01477	0.89052	0.01261	0.94795
0.01502	0.74621	0.0104	0.83135	0.02051	0.82064	0.01714	0.89785
0.02558	0.64607	0.01514	0.78346	0.02541	0.7305	0.02077	0.87159
0.03055	0.5892	0.02006	0.67087	0.03055	0.66285	0.02569	0.81081
0.04035	0.50553	0.02581	0.61388	0.03518	0.59682	0.03057	0.77057
0.04525	0.458	0.03079	0.52978	0.04007	0.53717	0.03303	0.73502
0.05013	0.39376	0.04424	0.43784	0.04899	0.4469	0.04036	0.66423
0.05502	0.36031	0.05013	0.39376	0.0551	0.39543	0.04539	0.6253
0.06017	0.31267	0.05683	0.37028	0.06364	0.35333	0.05012	0.57109
0.06506	0.27048	0.06783	0.3052	0.07586	0.31259	0.05539	0.49894
0.07239	0.22771	0.0776	0.2734	0.08565	0.28728	0.06505	0.45717
0.08716	0.17886	0.08613	0.23738	0.09628	0.26682	0.0725	0.42492
0.10049	0.14174	0.09349	0.22453	0.10509	0.24244	0.08067	0.37471
0.11038	0.12114	0.10082	0.20766	0.11253	0.22717	0.08715	0.34886
0.11893	0.1127	0.10693	0.19879	0.12108	0.21702	0.09938	0.31425
0.12749	0.10255	0.1139	0.1796	0.12842	0.21228	0.11038	0.27773
0.13603	0.09207	0.12036	0.17042	0.13574	0.20042	0.12004	0.25082
0.14215	0.08253	0.12526	0.16166	0.14186	0.18849	0.12871	0.23418
0.14826	0.08009	0.13015	0.15791	0.14916	0.17862	0.13726	0.22302
0.15437	0.07667	0.13504	0.14877	0.15408	0.17554	0.1458	0.20844
0.16134	0.07247	0.13992	0.13839	0.16019	0.16768	0.15314	0.1982
0.16659	0.06865	0.14481	0.13575	0.1663	0.16346	0.16047	0.18635
0.17148	0.06366	0.1497	0.13076	0.1724	0.15842	0.16659	0.17393
0.17637	0.06293	0.15337	0.12802	0.17729	0.15191	0.1727	0.17052
0.18125	0.06199	0.15825	0.12579	0.18218	0.14904	0.17881	0.16298
0.18614	0.05831	0.16192	0.11997	0.18707	0.14332	0.18492	0.1604
0.18981	0.05718	0.16559	0.11446	0.19318	0.13986	0.19103	0.15559
0.19469	0.05464	0.16925	0.11262	0.19807	0.13762	0.19713	0.1526
0.19836	0.05218	0.17292	0.11085	0.20173	0.13294	0.20202	0.1466
0.20325	0.0509	0.17658	0.11038	0.20662	0.13334	0.20691	0.1411
0.20691	0.05009	0.18025	0.10687	0.21151	0.12996	0.21302	0.13954
0.21058	0.04807	0.18269	0.1033	0.21518	0.12828	0.21791	0.1349
0.21425	0.04816	0.18636	0.10152	0.22006	0.12372	0.2228	0.13017
0.21791	0.04741	0.19002	0.09927	0.22373	0.12109	0.22768	0.13034
0.22158	0.04591	0.19369	0.09657	0.22862	0.1198	0.23257	0.12717
0.22524	0.04514	0.19613	0.0952	0.23228	0.11811	0.23735	0.12399
0.2289	0.04419	0.1998	0.09262	0.23717	0.11554	0.24113	0.12081
0.23135	0.04375	0.20224	0.09178	0.24084	0.11287	0.24601	0.12022
0.23502	0.04242	0.20588	0.08931	0.2445	0.11283	0.24846	0.11808
0.23868	0.04129	0.20835	0.08967	0.24817	0.11025	0.24846	0.11808
0.24235	0.04023	0.2108	0.08736	0.25061	0.10931	0.24846	0.11808
0.24357	0.04037	0.2141	0.08623	0.25061	0.10932	0.24846	0.11808
0.24357	0.04037	0.21691	0.08477	0.25061	0.10932	0.24846	0.11808
0.24357	0.04037	0.21935	0.08216	0.25061	0.10933	0.24846	0.11808
0.24357	0.04037	0.2218	0.08183	0.25061	0.10933	0.24846	0.11808
0.24357	0.04037	0.22424	0.08054	0.25061	0.10933	0.24846	0.11808
0.24357	0.04037	0.22668	0.08063	0.25061	0.10933	0.24846	0.11808
0.24357	0.04037	0.22913	0.07908	0.25061	0.10933	0.24846	0.11808
0.24357	0.04037	0.23279	0.07839	0.25061	0.10933	0.24846	0.11808
0.24443	0.04037	0.23524	0.07688	0.25147	0.10933	0.24846	0.11808
0.24468	0.04037	0.23768	0.07579	0.25172	0.10933	0.24846	0.11808
0.24476	0.04037	0.24012	0.07535	0.2518	0.10933	0.24931	0.11808
0.24478	0.03988	0.24257	0.07389	0.25182	0.10933	0.24957	0.11808
0.24479	0.03973	0.24501	0.07347	0.25183	0.10933	0.24965	0.11808
0.24479	0.03968	0.24623	0.07347	0.25183	0.10933	0.24967	0.11808
0.24479	0.03967	0.24623	0.07327	0.25183	0.10885	0.24968	0.11918
0.24479	0.03966	0.24623	0.07327	0.25183	0.10885	0.24968	0.11951
0.24479	0.03983	0.24623	0.07327	0.25183	0.10885	0.24968	0.11851
0.24479	0.03983	0.24623	0.07327	0.25183	0.10885	0.24968	0.11909
0.24479	0.03983	0.24623	0.07327	0.25183	0.10885	0.24968	0.11909
0.24479	0.03983	0.24623	0.07327	0.25183	0.10885	0.24968	0.11909
0.24479	0.03983	0.24623	0.07327	0.25183	0.10885	0.24968	0.11909
0.24479	0.03983	0.24623	0.07327	0.25183	0.10885	0.24968	0.11799
0.24479	0.03983	0.24623	0.07327	0.25183	0.10885	0.24968	0.11766
0.24479	0.03983	0.24623	0.07327	0.25183	0.10777	0.24968	0.11756
0.24479	0.03983	0.24623	0.07327	0.25183	0.10745	0.24968	0.11753
0.24479	0.03983	0.24623	0.07327	0.25183	0.10736	0.24968	0.11753
0.24479	0.03934	0.24623	0.07327	0.25183	0.10733	0.24968	0.11752
0.24479	0.0392	0.24623	0.07327	0.25183	0.10732	0.24968	0.11752
0.24479	0.03964	0.24623	0.07327	0.25183	0.10732	0.24968	0.11752
0.24479	0.03977	0.24623	0.07327	0.25183	0.10839	0.24968	0.11752
0.24479	0.03981	0.24623	0.07327	0.25183	0.10871	0.24968	0.11752
0.24479	0.03982	0.24623	0.07327	0.25183	0.10881	0.24968	0.11752
0.24479	0.03983	0.24623	0.07327	0.25183	0.10884	0.24968	0.11752
0.24479	0.03983	0.24623	0.07255	0.25183	0.10884	0.24968	0.11752
0.24479	0.03983	0.24623	0.07233	0.25183	0.10885	0.24968	0.11752
0.24479	0.03983	0.24623	0.07226	0.25183	0.10885	0.24968	0.11752
0.24479	0.03983	0.24623	0.07225	0.25183	0.10885	0.24968	0.11752
0.24479	0.03983	0.24623	0.07224	0.25183	0.10885	0.24968	0.11752
0.24479	0.03983	0.24623	0.07224	0.25183	0.10885	0.24968	0.11752
0.24479	0.03983	0.24623	0.07224	0.25183	0.10885	0.24968	0.11752
0.24479	0.03983	0.24623	0.07224	0.25183	0.10885	0.24968	0.11752
0.24479	0.03983	0.24623	0.07224	0.25183	0.10885	0.24968	0.11752
0.24479	0.03983	0.24623	0.07224	0.25183	0.10885	0.24968	0.11752
0.24479	0.03983	0.24623	0.07224	0.25269	0.10885	0.25053	0.11862
0.24479	0.03983	0.24623	0.07224	0.25295	0.10885	0.25079	0.11785
0.24479	0.03983	0.24623	0.07224	0.25302	0.10885	0.25087	0.11762
0.24565	0.03983	0.24623	0.07224	0.25305	0.10885	0.25089	0.11755
0.24591	0.03983	0.24623	0.07224	0.25305	0.10885	0.2509	0.11753
0.24598	0.03983	0.24623	0.07224	0.25306	0.10885	0.2509	0.11752
0.24601	0.03983	0.24623	0.07224	0.25306	0.10885	0.2509	0.11752
0.24601	0.03983	0.24623	0.07224	0.25306	0.10777	0.2509	0.11752
0.24602	0.03983	0.24623	0.07224	0.25306	0.10745	0.2509	0.11752
0.24602	0.03983	0.24623	0.07224	0.25306	0.10736	0.2509	0.11752
0.24602	0.03983	0.24623	0.07224	0.25306	0.10733	0.2509	0.11752
0.24602	0.03983	0.24623	0.07224	0.25306	0.10732	0.2509	0.11752
0.24602	0.03983	0.24709	0.07224	0.25306	0.10732	0.2509	0.11752
0.24602	0.03934	0.24735	0.07224	0.25306	0.10731	0.2509	0.11752
0.24602	0.0392	0.25306	0.10731	0.25306	0.10731	0.2509	0.11752

Figure A27.

pH = 6.4		pH = 7.5		pH = 8.3	
V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0
0	0.991	0.00452	1	0.00776	1.02391
0.00252	0.82537	0.00615	0.90128	0.01264	0.7515
0.005	0.65902	0.01015	0.72489	0.01473	0.75101
0.00736	0.58636	0.01504	0.65117	0.0217	0.68001
0.01066	0.4701	0.02	0.62623	0.02487	0.60938
0.01225	0.46259	0.02611	0.57766	0.03342	0.52939
0.01555	0.41394	0.03066	0.50613	0.04073	0.43476
0.01993	0.32272	0.03215	0.49588	0.04842	0.37834
0.0253	0.27304	0.0358	0.4654	0.05443	0.35304
0.03141	0.23062	0.04033	0.44943	0.05942	0.32972
0.0391	0.19477	0.0486	0.38186	0.06428	0.29732
0.04362	0.16328	0.04888	0.37048	0.06917	0.28578
0.04277	0.1505	0.04896	0.36707	0.07286	0.27178
0.04888	0.13811	0.05752	0.31565	0.07772	0.24618
0.05377	0.13231	0.06487	0.28612	0.0802	0.23335
0.05902	0.11629	0.07697	0.26011	0.08386	0.22812
0.06632	0.11199	0.08306	0.22145	0.0875	0.21611
0.06965	0.10871	0.08919	0.2173	0.09108	0.20413
0.07173	0.10335	0.09408	0.19418	0.09364	0.19278
0.07332	0.09844	0.10009	0.18394	0.0973	0.19441
0.07454	0.09568	0.10017	0.18394	0.09975	0.18554
0.07586	0.0933	0.10387	0.16967	0.10219	0.18479
0.07702	0.0869	0.10875	0.16676	0.10586	0.17627
0.07943	0.08593	0.11353	0.1589	0.1083	0.16863
0.08065	0.08292	0.11731	0.15466	0.11075	0.17138
0.08187	0.07944	0.11731	0.15608	0.11319	0.16573
0.08432	0.07791	0.12097	0.14364	0.11563	0.15877
0.08798	0.07874	0.12464	0.13658	0.11808	0.15346
0.09053	0.07599	0.1283	0.13684	0.12052	0.15465
0.09287	0.07323	0.13197	0.13064	0.12297	0.1478
0.09534	0.07208	0.13564	0.1259	0.12541	0.15446
0.09901	0.0721	0.13808	0.12187	0.12994	0.14844
0.1039	0.07023	0.14174	0.11657	0.13237	0.14408
0.10879	0.06929	0.14541	0.11839	0.1336	0.13807
0.11245	0.06742	0.14786	0.11346	0.13605	0.13477
0.11734	0.06617	0.15116	0.10838	0.13849	0.13462
0.11734	0.06617	0.15396	0.10929	0.14057	0.13312
0.12101	0.06462	0.15727	0.10394	0.14216	0.12757
0.12467	0.06371	0.15971	0.10275	0.1446	0.12943
0.12834	0.0623	0.16251	0.0966	0.14703	0.12943
0.132	0.06165	0.16496	0.10014	0.14827	0.12816
0.13689	0.06086	0.16741	0.09767	0.1507	0.12341
0.14056	0.06033	0.16984	0.09421	0.15193	0.11992
0.14422	0.05954	0.17226	0.09315	0.15438	0.1242
0.14789	0.05793	0.17352	0.09019	0.1556	0.1208
0.15156	0.05719	0.17682	0.08717	0.15803	0.11667
0.15522	0.05647	0.17841	0.08735	0.15926	0.11432
0.15889	0.05632	0.18196	0.08323	0.16171	0.1143
0.16133	0.05565	0.18329	0.083	0.16293	0.11339
0.165	0.05552	0.18574	0.08213	0.16537	0.11134
0.16866	0.0541	0.18818	0.08213	0.1666	0.11133
0.17233	0.05363	0.18818	0.08213	0.16867	0.11101
0.17477	0.05332	0.18818	0.08213	0.17026	0.10743
0.17955	0.05271	0.18818	0.08213	0.17148	0.10707
0.1821	0.05242	0.18818	0.08213	0.17382	0.10735
0.18577	0.05102	0.18818	0.08027	0.17515	0.10535
0.18821	0.04965	0.18818	0.07972	0.17637	0.10535
0.19188	0.04774	0.18818	0.083	0.17871	0.10524
0.19432	0.04586	0.18818	0.08317	0.18003	0.10208
0.19432	0.04586	0.18818	0.08321	0.18126	0.10048
0.19432	0.04586	0.18818	0.08141	0.18369	0.09734
0.19432	0.04586	0.18904	0.08087	0.18493	0.09757
0.19432	0.04586	0.18929	0.0807	0.18615	0.09962
0.19432	0.04586	0.18937	0.08066	0.18737	0.09905
0.19518	0.04586	0.18939	0.08064	0.18978	0.09905
0.19544	0.04586	0.1894	0.08064	0.19093	0.09897
0.19551	0.04586	0.1894	0.08246	0.191	0.09901
0.19554	0.04586	0.1894	0.083	0.19103	0.0976
0.19554	0.04586	0.1894	0.08317	0.19103	0.09718
0.19555	0.04586	0.1894	0.08321	0.19103	0.09706
0.19555	0.04586	0.1894	0.08323	0.19103	0.09702
0.19555	0.04586	0.1894	0.08323	0.19103	0.09701
0.19555	0.04586	0.1894	0.08324	0.19103	0.097
0.19555	0.04586	0.1894	0.08324	0.19103	0.097
0.19555	0.04586	0.1894	0.08324	0.19103	0.097
0.19555	0.04586	0.1894	0.08142	0.19103	0.097
0.19555	0.04586	0.1894	0.08087	0.19103	0.097
0.19555	0.04573	0.1894	0.0807	0.19103	0.097
0.19555	0.04573	0.1894	0.08066	0.19103	0.097
0.19555	0.04573	0.19026	0.08064	0.19103	0.097
0.19555	0.04573	0.19052	0.08064	0.19103	0.097
0.19555	0.04573	0.19059	0.08064	0.19103	0.097
0.19555	0.04573	0.19062	0.08063	0.19103	0.097
0.19555	0.04573	0.19062	0.08063	0.19189	0.097
0.19555	0.04573	0.19062	0.08063	0.19215	0.09559
0.19555	0.04573	0.19062	0.08063	0.19222	0.09516
0.19555	0.04573	0.19063	0.08246	0.19225	0.09645
0.19555	0.04573	0.19063	0.083	0.19225	0.09684
0.19555	0.04573	0.19063	0.08317	0.19226	0.09695
0.19555	0.04536	0.19063	0.08321	0.19226	0.09699
0.19555	0.04524	0.19063	0.08323	0.19226	0.097
0.19555	0.04521	0.19063	0.08323	0.19226	0.097
0.19555	0.0452	0.19063	0.08324	0.19226	0.097
0.19555	0.0452	0.19063	0.08142	0.19226	0.097
0.19555	0.04519	0.19063	0.08087	0.19226	0.09559
0.19555	0.04519	0.19063	0.0807	0.19226	0.09516
0.19555	0.04519	0.19063	0.08066	0.19226	0.09503
0.19555	0.04519	0.19063	0.08064	0.19226	0.095
0.19555	0.04519	0.19148	0.08064	0.19226	0.09499
0.19555	0.04519	0.19174	0.08064	0.19226	0.09498

Figure A28.

ΔP = 41.37 KPa		ΔP = 82.74 KPa		ΔP = 165.47 KPa		ΔP = 206.84 KPa	
V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0
9.97E-04	0.79484	0.00148	0.9752	0	0.86737	0	0.99977
0.00578	0.78869	0.01006	0.84228	0.01017	0.77986	0.00496	0.91259
0.01076	0.72426	0.0207	0.68847	0.01592	0.63644	0.011	0.75275
0.01566	0.70794	0.02569	0.55565	0.02004	0.54229	0.01597	0.66257
0.02044	0.63519	0.03055	0.49115	0.0274	0.50501	0.02112	0.62353
0.02543	0.57221	0.03536	0.4485	0.0347	0.41136	0.02575	0.57993
0.0319	0.46587	0.04281	0.38007	0.04525	0.34565	0.03347	0.45799
0.0401	0.41424	0.05013	0.35225	0.0554	0.29263	0.04041	0.40671
0.04532	0.39164	0.06017	0.30351	0.07016	0.23799	0.05169	0.32073
0.05147	0.35112	0.06638	0.27563	0.08239	0.20129	0.06	0.29018
0.06613	0.2759	0.07577	0.22982	0.09339	0.17602	0.06514	0.2504
0.07835	0.24318	0.0835	0.21689	0.10316	0.1584	0.07625	0.22823
0.08934	0.22228	0.09902	0.188	0.11294	0.15048	0.09702	0.18674
0.09912	0.19572	0.1116	0.16844	0.12138	0.13645	0.11377	0.1612
0.10768	0.18646	0.12382	0.15495	0.12883	0.13047	0.12876	0.13808
0.11623	0.1668	0.13482	0.13938	0.13616	0.1249	0.14102	0.12865
0.12477	0.16093	0.14423	0.13376	0.14349	0.11578	0.15323	0.11953
0.13208	0.15569	0.15315	0.12212	0.1496	0.10989	0.16544	0.11064
0.13822	0.14188	0.16291	0.11787	0.15693	0.10665	0.17634	0.10342
0.14552	0.13394	0.17026	0.11341	0.16304	0.10208	0.18501	0.0989
0.15167	0.1314	0.17881	0.10608	0.16914	0.1	0.19478	0.09447
0.15778	0.12874	0.18614	0.10343	0.17404	0.09879	0.20456	0.09029
0.16389	0.12676	0.19347	0.09971	0.18015	0.09489	0.2131	0.08773
0.16999	0.11739	0.2008	0.09588	0.18622	0.08932	0.229	0.08078
0.17488	0.11658	0.20814	0.09413	0.19115	0.08928	0.23755	0.07905
0.18099	0.11243	0.21425	0.09172	0.19722	0.08588	0.24488	0.07663
0.18588	0.11035	0.22158	0.08835	0.20203	0.08555	0.25221	0.07404
0.19077	0.10894	0.22769	0.08662	0.20581	0.08199	0.25955	0.07179
0.19651	0.10263	0.2338	0.0839	0.2107	0.08067	0.26565	0.07127
0.2014	0.10095	0.23991	0.08217	0.21558	0.07815	0.27176	0.06911
0.20543	0.09929	0.24602	0.08146	0.22047	0.07482	0.27176	0.06911
0.21032	0.0978	0.25202	0.07887	0.22536	0.07539	0.27176	0.06911
0.21517	0.09543	0.25701	0.07855	0.22903	0.07431	0.27176	0.06911
0.21887	0.09408	0.26312	0.07636	0.23391	0.07304	0.27176	0.06911
0.22376	0.09161	0.26801	0.07531	0.23758	0.07196	0.27176	0.06982
0.22743	0.08994	0.27412	0.07312	0.24243	0.07093	0.27176	0.07004
0.23195	0.08786	0.27534	0.07369	0.24613	0.06907	0.27176	0.0701
0.23598	0.08543	0.27534	0.07312	0.25102	0.06849	0.27176	0.06941
0.24087	0.08583	0.27534	0.07296	0.25469	0.06778	0.27176	0.0692
0.24453	0.08386	0.27534	0.0729	0.25835	0.06715	0.27176	0.06914
0.2482	0.08161	0.27534	0.07289	0.26202	0.06608	0.27262	0.06912
0.25187	0.08089	0.2762	0.07288	0.26568	0.06545	0.27288	0.06911
0.25553	0.0795	0.27646	0.07288	0.26935	0.06372	0.27295	0.06911
0.2592	0.07847	0.27653	0.07288	0.27302	0.06261	0.27298	0.06911
0.26286	0.07747	0.27656	0.07288	0.27302	0.06255	0.27298	0.06911
0.26653	0.07648	0.27656	0.07288	0.27302	0.06253	0.27299	0.06911
0.2702	0.07448	0.27656	0.07288	0.27302	0.06252	0.27299	0.06911
0.27386	0.07517	0.27657	0.07288	0.27302	0.06252	0.27299	0.06911
0.27631	0.07437	0.27657	0.07288	0.27387	0.06321	0.27299	0.06911
0.27631	0.07437	0.27657	0.07288	0.27413	0.06342	0.27299	0.06911
0.27631	0.07437	0.27657	0.07288	0.27421	0.06348	0.27299	0.06911
0.27631	0.07437	0.27657	0.07288	0.27423	0.0635	0.27299	0.06911
0.27631	0.07437	0.27657	0.07288	0.27424	0.06351	0.27299	0.0684
0.27631	0.07437	0.27657	0.07288	0.27424	0.06351	0.27299	0.0689
0.27631	0.07437	0.27657	0.07288	0.27424	0.06301	0.27299	0.06905
0.27631	0.07437	0.27657	0.07345	0.27424	0.06301	0.27299	0.06909
0.27631	0.07422	0.27657	0.07305	0.27424	0.06301	0.27299	0.06911
0.27631	0.07422	0.27657	0.07293	0.27424	0.06301	0.27299	0.06911
0.27631	0.07422	0.27657	0.0729	0.27424	0.06301	0.27299	0.06911
0.27631	0.07422	0.27657	0.07289	0.27424	0.06301	0.27384	0.06911
0.27631	0.07422	0.27657	0.07288	0.27424	0.06301	0.2741	0.06911
0.27631	0.07422	0.27742	0.07288	0.27424	0.06301	0.27418	0.06911
0.27631	0.07422	0.27768	0.07288	0.27509	0.06301	0.2742	0.06911
0.27631	0.07422	0.27775	0.07288	0.27535	0.06301	0.27421	0.06911
0.27631	0.07422	0.27778	0.07232	0.27543	0.0637	0.27421	0.06911
0.27631	0.07422	0.27778	0.07215	0.27545	0.06391	0.27421	0.06911
0.27631	0.07422	0.27779	0.07209	0.27546	0.06328	0.27421	0.06911
0.27631	0.07422	0.27779	0.07265	0.27546	0.06309	0.27421	0.06911
0.27631	0.07422	0.27779	0.07281	0.27546	0.06303	0.27421	0.06911
0.27631	0.07422	0.27779	0.07286	0.27546	0.06302	0.27421	0.0684
0.27631	0.07422	0.27779	0.07287	0.27546	0.06301	0.27421	0.06819
0.27631	0.07422	0.27779	0.07288	0.27546	0.06301	0.27421	0.06812
0.27631	0.07422	0.27779	0.07288	0.27546	0.06301	0.27421	0.0681
0.27631	0.07422	0.27779	0.07288	0.27546	0.06301	0.27421	0.0681
0.27631	0.07422	0.27779	0.07288	0.27546	0.06301	0.27421	0.0681
0.27631	0.07422	0.27779	0.07288	0.27546	0.06301	0.27421	0.0681
0.27631	0.07422	0.27779	0.07288	0.27546	0.06301	0.27421	0.0681
0.27631	0.07422	0.27779	0.07288	0.27546	0.06301	0.27506	0.0681
0.27716	0.07422	0.27779	0.07288	0.27546	0.06301	0.27532	0.0681
0.27742	0.07422	0.27779	0.07288	0.27546	0.06301	0.2754	0.0681
0.27749	0.07422	0.27779	0.07288	0.27546	0.06301	0.27542	0.0681
0.27752	0.07422	0.27779	0.07288	0.27546	0.06301	0.27543	0.0681
0.27752	0.07422	0.27779	0.07288	0.27546	0.06301	0.27543	0.0681
0.27753	0.07422	0.27779	0.07288	0.27632	0.06301	0.27543	0.06739
0.27753	0.07422	0.27779	0.07288	0.27657	0.06232	0.27543	0.06717
0.27753	0.07422	0.27779	0.07288	0.27665	0.06211	0.27543	0.06711
0.27753	0.07422	0.27779	0.07232	0.27667	0.06205	0.27543	0.06709
0.27753	0.07422	0.27779	0.07215	0.27668	0.06203	0.27543	0.06708
0.27753	0.07422	0.27779	0.07209	0.27668	0.06203	0.27543	0.06708
0.27753	0.07422	0.27864	0.07208	0.27668	0.06272	0.27543	0.06708
0.27753	0.07422	0.2789	0.07207	0.27668	0.06292	0.27543	0.06708
0.27753	0.07422	0.27898	0.07207	0.27668	0.06298	0.27543	0.06708
0.27753	0.07422	0.279	0.07207	0.27668	0.063	0.27543	0.06708
0.27753	0.07422	0.27901	0.07207	0.27668	0.06301	0.27543	0.06708
0.27753	0.07422	0.27901	0.07207	0.27668	0.06301	0.27543	0.06708
0.27753	0.07422	0.27901	0.07207	0.27668	0.06301	0.27543	0.06708
0.27753	0.07422	0.27901	0.07207	0.27668	0.06301	0.27629	0.06708
0.27753	0.07422	0.27901	0.07253	0.27668	0.06301	0.27654	0.06708
0.27753	0.07422	0.27901	0.07253	0.27668	0.06301	0.27662	0.06708
0.27753	0.07422	0.27901	0.07253	0.27668	0.06301	0.27664	0.06708

Figure A29.

Theoretical Values			
Generation Time (s)	Fe^{2+} (mg/L)	Generation Time (s)	Fe^{3+} (mg/L)
4.06	1	6.09	1
8.12	2	12.18	2
20.3	5	30.45	5
40.6	10	60.9	10
60.9	15	91.35	15
81.2	20		
93.38	23		

Experimental Values					
pH = 6.4		pH = 7.5		pH = 8.3	
Generation Time (s)	Fe(total) (mg/L)	Generation Time (s)	Fe(total) (mg/L)	Generation Time (s)	Fe(total) (mg/L)
6	1.32	6	1.21	6	1.72
12	2.69	12	2.69	12	3.13
30	6.94	30	6.86	30	7.04
61	14.7	61	13.75	61	13.54
91	23.14	91	21.08	91	21.13

Figure A30.

pH = 6.4	
Fe (total) (mg/L)	Fe^{2+} Percentage
1.32	44.44
2.69	39.99
6.94	31.99
14.7	16.09
23.14	15.95

pH = 7.5	
Fe (total) (mg/L)	Fe^{2+} Percentage
1.21	6.49
2.69	2.04
6.86	0.78
13.75	0.45
21.08	1.09

pH = 8.3	
Fe (total) (mg/L)	Fe^{2+} Percentage
1.78	2.98
3.2	2.12
7.13	1.22
13.62	0.6
21.19	0.3

Figure A31.

pH = 6.4	
Fe (total) (mg/L)	Number weighted mean particle diameter (μm)
0	2.74
6.5	2.8
13.2	2.89
20.9	3.12

pH = 7.5	
Fe (total) (mg/L)	Number weighted mean particle diameter (μm)
0	2.82
7.4	2.95
13.9	3.08
21.2	3.64

pH = 8.3	
Fe (total) (mg/L)	Number weighted mean particle diameter (μm)
0	2.78
1.8	2.84
3.2	2.86
7.1	2.9
13.6	3.1
21.2	3.7

Figure A32.

Raw Water		Chemical Coagulation		Electrocoagulation	
V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0
0.00722	0.81464	0.00244	0.9584	0.00722	0.81535
0.01952	0.75365	0.00611	0.8907	0.01063	0.72434
0.03055	0.69826	0.01099	0.87092	0.01552	0.66852
0.03507	0.65438	0.01588	0.86154	0.02041	0.59264
0.04032	0.60957	0.01955	0.85317	0.03044	0.51372
0.04642	0.56778	0.02555	0.82808	0.0391	0.45258
0.0501	0.513	0.02932	0.83602	0.05377	0.39223
0.05621	0.47359	0.03299	0.80249	0.0611	0.37029
0.06232	0.41337	0.03788	0.80034	0.06721	0.34567
0.06843	0.37029	0.04155	0.78734	0.07332	0.30312
0.07332	0.34301	0.04643	0.78606	0.07821	0.27698
0.08309	0.30106	0.05129	0.78288	0.08309	0.26661
0.08798	0.26082	0.05499	0.7738	0.08798	0.26279
0.09165	0.2518	0.05865	0.76066	0.09287	0.25178
0.09898	0.22912	0.06343	0.75067	0.09776	0.2427
0.10142	0.21883	0.0672	0.74884	0.10253	0.23162
0.10509	0.20949	0.07087	0.73705	0.10631	0.22716
0.1112	0.19583	0.07454	0.72403	0.10998	0.21386
0.11364	0.19213	0.07821	0.71291	0.11364	0.21194
0.11609	0.18329	0.08309	0.70194	0.11731	0.20199
0.11972	0.17882	0.08676	0.69044	0.12097	0.19885
0.12216	0.1714	0.09042	0.6792	0.12464	0.19973
0.12708	0.16449	0.09408	0.68086	0.12831	0.19191
0.13197	0.15666	0.09776	0.67088	0.13196	0.18408
0.13564	0.14812	0.10142	0.67085	0.13442	0.18316
0.13808	0.14671	0.10506	0.64935	0.13808	0.1776
0.14052	0.14466	0.10839	0.65168	0.14175	0.17429
0.14297	0.13954	0.11205	0.6426	0.14419	0.16903
0.14419	0.13826	0.11486	0.63109	0.14786	0.16729
0.1503	0.13162	0.11853	0.62229	0.1503	0.16581
0.15274	0.12992	0.12219	0.62158	0.15397	0.1587
0.15397	0.12427	0.12464	0.61374	0.15641	0.16014
0.15641	0.12533	0.12831	0.61374	0.15885	0.15847
0.16008	0.12144	0.13186	0.59447	0.16241	0.15377
0.1613	0.1222	0.13442	0.58674	0.16496	0.15256
0.16374	0.11841	0.13807	0.57893	0.16741	0.15095
0.16496	0.11537	0.14138	0.57561	0.16985	0.14682
0.16741	0.11638	0.14419	0.5713	0.1723	0.14555
0.1723	0.10984	0.14663	0.57122	0.17474	0.1439
0.17352	0.10924	0.1503	0.55382	0.1783	0.14038
0.18084	0.10295	0.15274	0.55592	0.18084	0.13959
0.18329	0.10275	0.15519	0.54988	0.18329	0.13653
0.18574	0.10069	0.15885	0.5397	0.18574	0.1354
0.1894	0.09847	0.16251	0.53303	0.18696	0.13502
0.19063	0.09847	0.16496	0.52678	0.1894	0.13428
0.19673	0.09433	0.16741	0.52651	0.19185	0.13204
0.2004	0.09218	0.16985	0.51621	0.19429	0.13204
0.20284	0.09028	0.17348	0.51056	0.19673	0.12981
0.20407	0.08818	0.17596	0.50337	0.19917	0.12757
0.21017	0.08608	0.17841	0.50316	0.2004	0.12533
0.21129	0.08608	0.17841	0.50316	0.20284	0.12533
0.21262	0.08608	0.18196	0.49038	0.20529	0.12533
0.21506	0.08167	0.18451	0.48565	0.20773	0.12309
0.21995	0.07963	0.18696	0.48857	0.21017	0.12044
0.22117	0.07923	0.1894	0.47347	0.2114	0.12005
0.22484	0.07768	0.19185	0.47487	0.21384	0.12005
0.22606	0.07711	0.19429	0.47272	0.21629	0.11791
0.22728	0.0798	0.19673	0.47203	0.21873	0.11576
0.22851	0.0798	0.19918	0.46238	0.21995	0.11548
0.22973	0.07789	0.20162	0.46362	0.2224	0.11546
0.23095	0.07789	0.20407	0.45717	0.22484	0.11338
0.23217	0.07627	0.20651	0.45091	0.22728	0.11128
0.23217	0.07789	0.20895	0.44674	0.22851	0.11128
0.23339	0.07789	0.2114	0.44316	0.23095	0.10918
0.23706	0.07627	0.21384	0.4386	0.23339	0.10918
0.23828	0.07507	0.21629	0.43965	0.23462	0.1089
0.2395	0.07507	0.21873	0.43618	0.23706	0.10834
0.24073	0.07507	0.22106	0.43865	0.23828	0.10697
0.24195	0.07478	0.2224	0.43083	0.24073	0.10644
0.24195	0.0729	0.22484	0.43082	0.24314	0.10564
0.24317	0.07333	0.22728	0.42416	0.24439	0.10512
0.24561	0.07276	0.22973	0.42574	0.24684	0.10434
0.24684	0.07206	0.23217	0.41908	0.24806	0.10505
0.24927	0.0709	0.23461	0.41872	0.2505	0.10474
0.24928	0.06961	0.23669	0.41515	0.25172	0.10308
0.25172	0.06837	0.23828	0.41515	0.25417	0.10308
0.25294	0.0686	0.24073	0.40849	0.25539	0.10272
0.25417	0.06882	0.24317	0.40729	0.25783	0.10224
0.25417	0.06904	0.24525	0.4034	0.25905	0.10061
0.25661	0.06789	0.24684	0.39956	0.26028	0.10061
0.25905	0.06724	0.24928	0.39485	0.26272	0.09903
0.26028	0.06619	0.2505	0.39568	0.26394	0.09899
0.2615	0.06642	0.25294	0.39215	0.26639	0.09903
0.26394	0.06498	0.25539	0.38994	0.26883	0.09632
0.26516	0.06555	0.25661	0.38631	0.26883	0.09744
0.26883	0.06361	0.25905	0.38562	0.27127	0.09546
0.27005	0.06437	0.2615	0.38482	0.2725	0.09649
0.27127	0.06352	0.26394	0.38638	0.27372	0.09565
0.2725	0.06327	0.26516	0.38092	0.27457	0.09546
0.27372	0.0631	0.26761	0.38058	0.27494	0.09528
0.27494	0.06163	0.27004	0.37551	0.27616	0.0938
0.27738	0.06241	0.27127	0.37369	0.27727	0.094
0.27861	0.06224	0.27127	0.37369	0.27738	0.094
0.27983	0.06071	0.27127	0.37369	0.27861	0.09376
0.28105	0.06165	0.27127	0.37369	0.27983	0.09353
0.28227	0.06148	0.27127	0.37369	0.27983	0.09353
0.28349	0.06006	0.27127	0.37369	0.28105	0.09331
0.28472	0.06099	0.27213	0.37369	0.28227	0.09183
0.28594	0.05958	0.27239	0.37369	0.28227	0.09331
0.28716	0.05942	0.27246	0.37369	0.28349	0.0931

Figure A33.

Raw Water		Chemical Coagulation		Electrocoagulation	
V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0	V/A (m^3/m^2)	J/J_0
0	0.89172	0	1	0	0.97416
0.00585	0.70307	0.00245	0.92892	0.00251	0.93408
0.01074	0.66935	0.00578	0.79302	0.00585	0.81339
0.01563	0.58797	0.01066	0.68149	0.0083	0.68357
0.02052	0.55455	0.01555	0.55451	0.01071	0.63352
0.02541	0.48197	0.02322	0.45431	0.01563	0.50298
0.03066	0.37061	0.03791	0.33196	0.02199	0.42429
0.04007	0.30889	0.04891	0.28336	0.03429	0.3267
0.04774	0.24323	0.05991	0.23973	0.04522	0.27401
0.05877	0.19317	0.06843	0.20687	0.05388	0.23811
0.06973	0.17589	0.0758	0.19467	0.0624	0.20295
0.07832	0.15604	0.08313	0.19017	0.0694	0.18891
0.08564	0.14079	0.09035	0.1713	0.07587	0.17174
0.09176	0.11801	0.09535	0.15827	0.08198	0.16668
0.09787	0.107	0.10146	0.14812	0.08809	0.15455
0.10398	0.10568	0.10757	0.14276	0.0942	0.15218
0.10887	0.10615	0.11246	0.13568	0.09909	0.14001
0.11376	0.09986	0.11734	0.12765	0.1052	0.13941
0.11864	0.08629	0.12223	0.12674	0.11006	0.13116
0.12231	0.08106	0.12712	0.12106	0.11375	0.12866
0.12719	0.07643	0.13079	0.11565	0.11864	0.12439
0.13086	0.0723	0.13567	0.11133	0.12352	0.11882
0.13453	0.0743	0.14056	0.10608	0.1272	0.1121
0.13931	0.07068	0.14423	0.10713	0.13208	0.10899
0.14272	0.06739	0.14789	0.10515	0.13575	0.11007
0.14553	0.06235	0.15156	0.10287	0.13942	0.10495
0.14919	0.06221	0.15522	0.09861	0.14308	0.1003
0.15285	0.05702	0.15889	0.09452	0.14675	0.10134
0.1553	0.05702	0.16256	0.09515	0.15041	0.09975
0.15897	0.05339	0.16622	0.09204	0.15408	0.09777
0.16141	0.05702	0.16989	0.09203	0.15775	0.09456
0.16386	0.05184	0.17233	0.08891	0.16019	0.09456
0.16752	0.05184	0.176	0.08693	0.16386	0.09136
0.16997	0.05184	0.17966	0.08471	0.16752	0.08777
0.17363	0.05184	0.18211	0.08339	0.17119	0.0869
0.17607	0.05113	0.18577	0.08111	0.17484	0.08724
0.17852	0.04601	0.18822	0.08037	0.1773	0.08489
0.18096	0.05099	0.19188	0.07822	0.17974	0.08258
0.18341	0.04557	0.19433	0.07896	0.18341	0.08185
0.18585	0.04555	0.19799	0.07651	0.18585	0.08113
0.18829	0.04147	0.20044	0.07412	0.18952	0.08007
0.19074	0.04147	0.20288	0.07424	0.19196	0.07903
0.19318	0.04147	0.20532	0.07331	0.1944	0.07628
0.19563	0.04147	0.20899	0.07167	0.19806	0.07558
0.19807	0.04147	0.21143	0.07155	0.20051	0.0757
0.20051	0.04147	0.21388	0.07062	0.20407	0.07454
0.20295	0.03629	0.21632	0.0702	0.20662	0.07452
0.20418	0.03629	0.21877	0.06865	0.20907	0.07268
0.20662	0.04147	0.22121	0.06826	0.21151	0.07226
0.20907	0.04147	0.22365	0.06748	0.21396	0.07144
0.21029	0.03629	0.2261	0.0671	0.2164	0.07008
0.21273	0.03629	0.22854	0.06581	0.21884	0.06877
0.21518	0.03672	0.23221	0.06544	0.22129	0.06836
0.21759	0.03629	0.23465	0.06507	0.22373	0.06798
0.21884	0.03629	0.2371	0.06471	0.22617	0.0676
0.22129	0.03633	0.23954	0.06346	0.22862	0.06631
0.22373	0.04147	0.24195	0.06239	0.23106	0.06592
0.22495	0.03629	0.24321	0.0615	0.23351	0.06502
0.2274	0.03784	0.24565	0.0615	0.23595	0.06411
0.22862	0.03112	0.24809	0.06061	0.23839	0.06383
0.23106	0.03629	0.25054	0.06061	0.24073	0.06319
0.23347	0.0311	0.25298	0.05883	0.24206	0.06236
0.23473	0.0311	0.25532	0.05883	0.24573	0.06228
0.23595	0.0311	0.25665	0.05883	0.24806	0.06136
0.23839	0.0311	0.25909	0.05794	0.24939	0.06045
0.23962	0.0311	0.26153	0.05794	0.25184	0.06042
0.24206	0.0311	0.26276	0.05756	0.25428	0.05953
0.24328	0.03111	0.2652	0.05635	0.25672	0.05915
0.24569	0.0311	0.26764	0.0555	0.25795	0.05878
0.24695	0.02956	0.27009	0.0555	0.26039	0.0579
0.24817	0.0311	0.27253	0.05464	0.26283	0.05702
0.25061	0.0311	0.27375	0.05464	0.26528	0.05729
0.25184	0.0311	0.2762	0.05435	0.2665	0.05641
0.25417	0.0311	0.27864	0.05352	0.26894	0.05585
0.2555	0.02606	0.27986	0.05352	0.27102	0.05585
0.25672	0.0311	0.28231	0.05268	0.27261	0.05499
0.25917	0.0311	0.28475	0.05268	0.27505	0.05499
0.26039	0.02592	0.28597	0.05268	0.27628	0.05585
0.26039	0.0311	0.28842	0.05185	0.27872	0.05499
0.26125	0.0311	0.28964	0.05187	0.28116	0.05499
0.2615	0.0311	0.29208	0.05185	0.28361	0.05413
0.26158	0.0311	0.29449	0.05171	0.28483	0.05413
0.2616	0.0311	0.29575	0.05062	0.28727	0.05313
0.26161	0.0311	0.29819	0.05119	0.28935	0.05286
0.26161	0.0311	0.29942	0.04985	0.29094	0.05259
0.26161	0.0311	0.30186	0.05042	0.29327	0.05206
0.26161	0.0311	0.30308	0.04936	0.2946	0.0518
0.26161	0.0311	0.30553	0.04992	0.29705	0.05071
0.26161	0.0311	0.30675	0.04954	0.29827	0.04988
0.26161	0.0311	0.30919	0.04918	0.30071	0.04845
0.26161	0.0311	0.31041	0.04804	0.30071	0.04751
0.26161	0.0311	0.31041	0.04804	0.30071	0.04727
0.26161	0.0311	0.31041	0.04804	0.30071	0.04727
0.26161	0.0311	0.31041	0.04804	0.30071	0.04727
0.26161	0.0311	0.31041	0.04804	0.30157	0.04727
0.26161	0.0311	0.31041	0.04804	0.30183	0.04727
0.26161	0.0311	0.31041	0.04804	0.3019	0.04727
0.26161	0.0311	0.31041	0.04804	0.30193	0.04727
0.26161	0.0311	0.31041	0.04804	0.30193	0.04727
0.26161	0.0311	0.31041	0.04804	0.30194	0.04727

Figure A34.

pH = 6.4

Chemical Coagulation		Electrocoagulation	
Fe^{3+} Concentration (mg/L)	UV_{254} removal (%)	Fe^{3+} Concentration (mg/L)	UV_{254} removal (%)
0	0.00	0	0.00
5	30.73	4.59	15.70
10	40.78	11.39	28.49
15	60.34	18.59	50.00

Figure A35.

pH = 6.4

Chemical Coagulation		Electrocoagulation	
Fe^{3+} Concentration (mg/L)	DOC removal (%)	Fe^{3+} Concentration (mg/L)	DOC removal (%)
0	0.00	0	0.00
5	26.33	4.59	16.42
10	36.04	11.39	27.79
15	43.82	18.59	37.05

Figure A36.

pH = 6.4

Chemical Coagulation		Electrocoagulation	
Fe^{3+} Concentration (mg/L)	Microfiltered water SUVA (L/mg-m)	Fe^{3+} Concentration (mg/L)	Microfiltered water SUVA (L/mg-m)
0	3.46	0	3.62
5	2.97	4.59	3.65
10	2.82	11.39	3.59
15	2.23	18.59	2.88

Figure A37.

pH = 7.5

Chemical Coagulation		Electrocoagulation	
Fe^{3+} Concentration (mg/L)	UV_{254} removal (%)	Fe^{3+} Concentration (mg/L)	UV_{254} removal (%)
0	0.00	0	0.00
5	22.22	7.33	25.58
10	34.19	13.78	40.70
15	46.15	21.1	53.49

Figure A38.

pH = 7.5

Chemical Coagulation		Electrocoagulation	
Fe^{3+} Concentration (mg/L)	DOC removal (%)	Fe^{3+} Concentration (mg/L)	DOC removal (%)
0	0.00	0	0.00
5	16.52	7.33	18.02
10	23.04	13.78	24.16
15	32.39	21.1	29.90

pH = 7.5

Chemical Coagulation		Electrocoagulation	
Fe^{3+} Concentration (mg/L)	Microfiltered water SUVA (L/mg-m)	Fe^{3+} Concentration (mg/L)	Microfiltered water SUVA (L/mg-m)
0	3.35	0	3.41
5	2.66	7.33	3.09
10	2.40	13.78	2.66
15	2.25	21.1	2.43

Figure A40.

Fe^{3+} dose (mg/L)	Volume filtered per unit membrane area (cm^3/cm^2)	MS2 Log removal
0	5	0.228
	24	0.439
	36	0.447
2	5	4.649
	25	4.549
	43	4.623
5	5	6.491
	25	7.213
	44	7.866

Figure A41.

Fe^{3+} dose (mg/L)	Volume filtered per unit membrane area (cm^3/cm^2)	MS2 Log removal
0	5	0.600
	24	0.908
	36	1.077
2	5	0.790
	25	1.185
	43	1.196
5	5	1.349
	25	2.095
	44	2.077
10	5	3.277
	25	5.135
	45	4.922

Figure A42.

Fe^{3+} dose (mg/L)	MS2 Log removal	
	pH 6.4	pH 7.5
0	0.228	0.600
2	4.649	0.790
5	6.491	1.349
10	7.69	3.277

Figure A43.

Fe^{3+} dose (mg/L)	MS2 Log removal	
	pH 6.4	pH 7.5
0	0.439	0.908
2	4.549	1.185
5	7.213	2.095
10	7.600	5.135

Figure A44.

Fe^{3+} dose (mg/L)	MS2 Log removal	
	pH 6.4	pH 7.5
0	0.447	1.077
2	4.623	1.196
5	7.866	2.077
10	7.900	4.922

Figure A45.

Fe^{3+} dose (mg/L)	Volume filtered per unit membrane area (cm^3/cm^2)	MS2 Log removal
0	5	0.034
	24	0.240
	44	0.344
1	5	0.216
	25	0.402
	44	0.220
2	5	4.340
	25	4.332
	44	4.756
3	5	5.101
	25	5.017
	44	5.199

Figure A46.

Fe^{3+} dose (mg/L)	Volume filtered per unit membrane area (cm^3/cm^2)	MS2 Log removal
0	5	0.011
	24	0.013
	43	0.008
1	5	0.207
	25	0.558
	43	0.898
2	5	1.205
	25	1.722
	43	2.203
3	5	2.618
	25	2.745
	43	3.317

Fe^{3+} dose (mg/L)	**Volume filtered per unit membrane area (cm^3/cm^2)**	**MS2 Log removal**
0	5	0.908
	24	1.616
	41	1.743
2	5	2.992
	24	4.214
	41	4.899
5	5	4.677
	24	5.899
	41	6.677

Fe^{3+} dose (mg/L)	**Volume filtered per unit membrane area (cm^3/cm^2)**	**MS2 Log removal**
0	5	0.923
	24	1.787
	41	2.698
2	5	1.883
	24	2.505
	41	2.857
5	5	2.116
	24	4.388
	41	4.784

Fe^{3+} dose (mg/L)	**Volume filtered per unit membrane area (cm^3/cm^2)**	**MS2 Log removal**
0	5	0.250
	24	0.780
	41	1.875
2	5	1.428
	24	2.187
	41	3.539
5	5	3.070
	24	4.434
	41	4.670
10	5	3.107
	24	4.922
	41	5.360

pH	Fe^{3+} dose (mg/L)	MS2 log removal
6.4	0	0.91
	2	2.99
	5	4.68
7.5	0	0.92
	2	1.88
	5	2.12
8.3	0	1.25
	2	1.43
	5	3.07

pH	Fe^{3+} dose (mg/L)	MS2 log removal
6.4	0	1.62
	2	4.21
	5	5.9
7.5	0	1.79
	2	2.51
	5	4.39
8.3	0	0.78
	2	2.19
	5	4.43

pH	Fe^{3+} dose (mg/L)	MS2 log removal
6.4	0	1.74
	2	4.9
	5	6.68
7.5	0	2.7
	2	2.86
	5	4.7
8.3	0	1.87
	2	3.54
	5	4.67

pH	Fe (total) mg/L	% Fe(II) remaining
7.5	1.67	9%
	1.97	2%
	4.43	0%
	9.68	1%
	16.25	3%
6.4	1.17	72%
	2.22	69%
	4.95	54%
	9.43	24%
	15.70	13%

Run Number	% Fe(II) remaining
1	62%
2	58%
3	49%
4	47%
5	45%
6	45%
7	39%
8	48%
9	53%
10	45%
11	46%
12	49%
13	53%
14	41%
15	43%
16	46%
17	48%
18	34%
19	43%
20	54%
21	56%
22	53%
23	55%
24	57%
25	52%
26	44%

Run No.	% Fe (II) remaining
1	10%
2	11%
3	13%
4	12%
5	10%
6	11%
7	10%
8	11%
9	12%
10	10%

Fe dose (mg/L)	**MS2 Log removal**
0	0.059
1	0.140
2	0.224
5	0.287
10	0.298
15	0.441

Fe(tot) generated (mg/L)	Volume filtered per unit membrane area (cm^3/cm^2)	MS2 Log removal
0.62	5	1.004
	25	0.570
	39	0.559
2.89	5	3.565
	25	3.485
	39	3.084
6.02	6	4.927
	25	5.814
	39	5.706
12.8	5	5.972
	25	6.602
	39	7.176

Fe dose (mg/L)	Volume filtered per unit membrane area (cm^3/cm^2)	MS2 Log removal
0.53	5	0.860
	25	0.958
	39	1.081
2.41	5	1.422
	25	1.468
	39	1.530
5.39	6	3.030
	25	2.982
	39	3.252
11.61	5	3.461
	25	4.085
	39	4.137

pH	Fe(III) dose (mg/L)	MS2 log removal
6.4	0.62	1.00
	2.89	3.56
	6.02	4.93
	12.8	5.97
7.5	0.53	0.86
	2.41	1.42
	5.39	3.03
	11.61	3.46

pH	Fe(III) dose (mg/L)	MS2 log removal
6.4	0.62	0.57
	2.89	3.48
	6.02	5.81
	12.8	6.60
7.5	0.53	0.96
	2.41	1.47
	5.39	2.98
	11.61	4.09

pH	Fe dose (mg/L)	MS2 log removal
6.4	0.62	0.56
	2.89	3.08
	6.02	5.71
	12.8	7.18
7.5	0.53	1.08
	2.41	1.53
	5.39	3.25
	11.61	4.14

Fe dose (mg/L)	Volume filtered per unit membrane area (cm^3/cm^2)	MS2 Log removal
1.87	5	0.057
	24	1.164
	39	1.258
2.96	5	1.214
	24	1.656
	40	1.612
6.11	5	1.739
	24	1.817
	39	1.848
13.04	5	1.153
	24	1.716
	39	1.630

Fe dose (mg/L)	Volume filtered per unit membrane area (cm^3/cm^2)	MS2 Log removal
0.42	5	0.515
	24	1.281
	39	1.472
3.28	5	1.081
	24	1.360
	40	1.433
6.3	5	1.107
	24	1.700
	39	1.804
11.48	5	0.789
	24	1.348
	39	1.433

Fe dose (mg/L)	Volume filtered per unit membrane area (cm^3/cm^2)	MS2 Log removal
1.24	5	0.365
	24	1.633
	39	1.906
3.16	5	1.028
	24	1.379
	39	1.340
7.07	5	1.588
	24	1.503
	39	1.610
12.24	5	0.833
	24	2.165
	39	2.419

Water sample	Volume filtered per unit membrane area (cm^3/cm^2)	MS2 Log removal
Synthetic Water	5	5.97
	25	6.64
	39	7.27
Lake Water	5	1.16
	24	1.72
	39	1.63

Water sample	Volume filtered per unit membrane area (cm^3/cm^2)	MS2 Log removal
Synthetic Water	5	3.46
	25	4.09
	39	4.14
Lake Water	5	0.79
	24	1.35
	39	1.44

pH	MS2 log removal		Fe(Tot) (mg/L)
6.4	7.24	CC-MF	5
	5.81	EC-MF	6.02
7.5	2.2	CC-MF	5
	2.9	EC-MF	5.39

pH	MS2 log removal		Fe(Tot) (mg/L)
6.4	5.9	CC-MF	5
	1.82	EC-MF	6.11
7.5	4.39	CC-MF	5
	1.7	EC-MF	6.3

Ferrous dose (mg/L)	MS2 Log removal	
	Synthetic Water	Lake Water
0	0.023	0.039
2	0.893	0.021
5	2.743	0.133
10	3.337	0.213

DOC Conc (mg/L)	MS2 Log removal	Std Deviation
0	3.50	0.02
0.54	3.51	0.04
0.78	3.25	0.02
1.02	2.52	0.05
1.31	1.10	0.03
1.59	0.48	0.05
2.66	0.01	0.07
1.10	1.26	0.05
0.53	1.27	0.10
0.86	2.60	0.01
1.32	1.25	0.16
1.47	1.03	0.09

DOC/Fe in CC Feed	%Fe in filtrate	%DOC in filtrate
0.2	0%	83%
0.4	0%	57%
0.7	0%	68%
1.7	0%	73%
3.1	93%	100%
8.8	94%	91%

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