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Coagulation-Ceramic Membrane Filtration for U.S. Surface Water Treatment Summary

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OBJECTIVES

The objective of this project was to conduct a systematic pilot-scale investigation of a hybrid coagulation-ceramic membrane treatment system to gain fundamental insights about necessary pretreatment conditions, fouling mechanisms, and contaminant removal capabilities, using two U.S. surface water sources. A two-phase plan was implemented for each of the three coagulants considered in this study: aluminum sulfate, aluminum chlorohydrate, and ferric chloride. The first phase involved the optimization of the coagulation pretreatment conditions that provided the best performance in terms of particle removal, organics removal, and membrane fouling. The second phase involved a comprehensive performance evaluation of the optimized system. The removal of precursors of selected regulated and emerging disinfection by-products as well as selected microorganisms and surrogates from the two U.S. surface waters was determined.

BACKGROUND

Compared to conventional treatment methods, membranes can reliably provide an absolute barrier against particles and microorganisms, and membranes are less impacted by fluctuating source water quality. However, U.S. membrane applications for drinking water treatment almost exclusively use the cheaper and more common polymeric membranes. Recent reductions in production costs have sparked a new interest in the application of ceramic membranes for drinking water treatment. Ceramic membranes are far superior to polymeric membranes in terms of structural, chemical, and thermal stability. This suggests that ceramic membranes can operate at higher fluxes and tolerate extreme cleaning procedures without compromising membrane integrity.

Membrane fouling is one of the most prevailing problems associated with any membrane system. Membrane fouling is caused by the accumulation or adsorption of contaminants on the membrane surface or within the pores. Membrane fouling can decrease the permeability of the membrane surface, decrease the quality of product water, increase operating pressures, increase downtime, and shorten the useful life of the membrane. Therefore, much research has been devoted to understand fouling mechanisms and investigate methods to decrease or inhibit

membrane fouling. Coagulation pretreatment has been accepted as an effective solution to control membrane fouling in low pressure membrane systems. Membrane performance is generally improved since particles are incorporated into relatively large flocs that are rejected by the membrane and flushed via a simple hydraulic backwash. However, the conditions at which a coagulant is dosed are critical since improper application of coagulation can lead to increased membrane fouling.

Despite their numerous known benefits over polymeric membranes, there is a severe lack of research knowledge and drinking water industry experience with ceramic membranes, particularly in the United States. Applicable research studies that demonstrate whether ceramic membranes can meet the current U.S. drinking water quality standards are limited. Even less is known about optimizing the chemical pretreatment scheme for ceramic membrane systems or the primary mechanisms responsible for fouling of ceramic membranes.

APPROACH

Ceramic membrane hollow fibers with a nominal pore size of 0.1 μm and a total surface area of 0.8 m^2 were used in this study. Two identical process trains were operated simultaneously in constant flux mode to directly treat raw surface water at each of the locations considered. Coagulation pretreatment was implemented in only one of the trains, and its impact on membrane fouling and the performance of the overall system was investigated. Membrane fouling was quantified in terms of pressure increase per unit time. A distinction was made between the types of fouling encountered during each experiment. Fouling that could be effectively removed via a simple hydraulic backwash was termed reversible fouling, while irreversible fouling occurred when a simple backwash was not enough to recover the loss in permeability. Total fouling is the sum of the two types of fouling. Based on a range of common coagulation pretreatment conditions found in the literature, a 3x3 matrix of coagulant doses at different pH conditions was assessed. After the best combination of coagulant dose and pH was identified, a comprehensive performance evaluation was completed with the selected pretreatment conditions.

Fundamental fouling characteristics of ceramic membranes were investigated in a lab-scale filtration system. Ceramic microfiltration, ultrafiltration, and nanofiltration membranes were adapted, and the effects of pore size, particle loading, solution chemistry, and coagulation were evaluated. Solutions containing model compounds (such as polyethylene glycol (PEG) or Suwannee River humic acid [SRHA]), a synthetic river water, and real surface waters were tested. Model analyses were conducted including a resistance-in-series model, combined pore blockage-cake filtration model, filtration power laws, and a unified membrane fouling index (UMFI) model. A systematic comparison was made between ceramic membranes and their polymeric counterparts for better understanding of the fouling behavior of ceramic membranes.

RESULTS AND CONCLUSIONS

Aluminum Sulfate Pretreatment

Aluminum sulfate (alum) optimization experiments using river water showed that with a dose of 1.3 mg/L as Al (14 mg/L as alum) at pH 6.0, approximately 83 percent of the total membrane fouling rate was reduced compared to the control train. At this condition, the total fouling rate was 0.15 psi/hr with coagulation pretreatment and 0.81 psi/hr without coagulation pretreatment. The rate of irreversible fouling gradually decreased as the experiment progressed until its effects became negligible. At this point, the pressure stabilized. Almost 98 percent of

total fouling was reduced with a dose of 3.5 mg/L as Al (38 mg/L as alum); however the overall cost of treatment and the possibility of elevated residual aluminum concentrations were considered when selecting the optimal treatment condition.

The comprehensive performance evaluation (CPE) with alum was performed at a dose of 1.3 mg/L as Al at a pH of 6.0. The total fouling rate was found to be 95 percent lower with coagulation pretreatment than without coagulation pretreatment. Residual aluminum concentrations were below their maximum contaminant level (MCL). Very little or no removal of UV₂₅₄ absorbance and dissolved organic carbon (DOC) was achieved by the control train, but an average of 60 and 30 percent UV₂₅₄ absorbance and DOC was removed by the experimental train, respectively. Regulated disinfection by-product (DBP) concentrations were above their MCLs without coagulation pretreatment and well below their MCLs with coagulation pretreatment. DBP removals were in the range of 30 to 50 percent with coagulant pretreatment and negligible without coagulation pretreatment. Both treatment trains demonstrated effective removal of particles as small as 0.75 µm.

The overall results suggest that coagulation pretreatment with alum was not only effective in decreasing the effects of irreversible fouling, but also in removing DBP precursors that would not have been removed otherwise. The coagulant dose used at the pilot plant was 43 percent lower than the dose used at Catawba River Water Treatment Plant (25 mg/L as alum + polymer aid).

Aluminum Chlorohydrate Pretreatment

Preliminary aluminum chlorohydrate (ACH) optimization experiments showed that membrane fouling remained primarily irreversible under all coagulation conditions studied and in some cases was worse than in the control train. Increasing the detention time between the point of coagulant addition and the membrane showed some improvement; therefore optimization experiments were repeated with an approximately 2 minute detention between the point of coagulant addition and the membrane filter.

The best coagulation condition was observed at a dose of 1.3 mg/L as Al at a pH of 6.0. Total fouling rates were 0.7 psi and 1.5 psi/hr for the experimental and control trains, respectively. Although experimental train fouling rates were lower compared to the control train, the pressure never stabilized and fouling proceeded irreversibly.

The comprehensive performance evaluation with ACH was performed at a dose of 1.3 mg/L as Al at a pH of 6.0. The experimental train membrane fouling was irreversible and the system lasted only four hours before reaching the pressure limit. Membrane fouling rates were four times higher with the experimental train than the control train. It was clear that the addition of the coagulant led to the increased fouling rates. There was very little to no removal of DBP precursors by either the experimental or the control trains. Aluminum chlorohydrate was reported by many authors to have been an effective coagulant for the pretreatment of ceramic membranes; however, aluminum chlorohydrate did not perform as well as expected in this study. Several potential solutions were tested but did not result in any significant improvements in the performance of the system. Ultimately, downstream conductivity instrumentation was installed and showed that the syringe pump was inconsistently and inaccurately dosing the coagulant during the filtration cycle. Results comparable to those achieved with aluminum sulfate and ferric chloride were observed when a smaller syringe was used or the volumetric flow rate was increased by diluting the coagulant. Both options increased the velocity at which the syringe plunger was depressed.

Ferric Chloride Pretreatment

Ferric chloride concentrations were selected on an equimolar basis relative to aluminum sulfate. Optimization results showed that ferric chloride was not effective at the highest pH (7.0) and lowest dose (1.0 mg/L as Fe). On the other hand, approximately 95 percent of the membrane fouling rate was reduced at a dose of 2.6 mg/L as Fe at pH 5.5. Ferric chloride performed well only over a narrower range of conditions relative to alum.

A ferric chloride comprehensive performance evaluation was performed at a coagulant dose of 2.6 mg/L as Fe at a pH of 5.5. The results showed that experimental train membrane fouling rates were approximately 92 percent lower than the control train. The total rate of fouling was found to be 2 psi/hr for the control train and 0.2 psi/hr with the experimental train. Both influent and effluent metal concentrations were below their MCL. Average UV₂₅₄ absorbance and DOC removals were approximately 38 and 27 percent, respectively. On the other hand, the removals were negligible without coagulation pretreatment. DBP concentrations were 20 to 45 percent lower with the experimental train than the control train; however, because the water had a low organic carbon concentration, effluent DBP concentrations for both trains were below their MCL values. The overall results indicated that ferric chloride was effective in reducing the rate of membrane fouling and removing DBP precursors; thus making it a viable coagulant pretreatment method for ceramic membrane filtration.

Fouling characteristics of ceramic membranes

The effects of membrane pore size and particle loading in the feed solutions were similar to results reported in other studies utilizing polymeric membranes, influencing the fouling rate and the mechanisms. It was noted, however, that the fouling of the ceramic membranes was less severe and mostly removable by physical cleaning, while there were significant resistances removable only by chemical cleaning and irreversible fouling for the polymeric membranes. Cake filtration was more pronounced with ceramic membranes but pore blockage mechanisms were mainly responsible for the fouling of polymeric membranes. It was suggested that even though the effects of solution chemistry on the fouling behavior with ceramic membranes were mostly similar with their polymeric counterparts in terms of trends, the extents of such effects varied depending on water quality parameters. The effects of pH and calcium ions were pronounced while that of ionic strength was less noticeable. Less fouling tendency and better cleaning efficiency were observed with the ceramic membrane, which was a promising finding for ceramic membrane application to surface water treatment. The surface properties of ceramic membranes made of metal oxides (such as being more hydrophilic and having a higher isoelectric points than polymeric membranes) seem to be responsible for the difference in the fouling mechanisms, the lower fouling potential, and more efficient cleaning than their polymeric counterparts.

APPLICATIONS AND RECOMMENDATIONS

This report serves as a valuable reference for future ceramic membrane research projects as well as for regulators, consulting engineers, and drinking water utilities to successfully implement and develop this technology. Coagulation pretreatment guidelines for ceramic membranes with common U.S. coagulants were established. The performance capabilities of ceramic membranes to remove precursors of disinfection by-products were demonstrated under

optimized coagulation pretreatment conditions. Fouling mechanisms of ceramic membranes were quantitatively identified and modeled.

Additional research is recommended to compare ceramic membranes to polymeric membrane hybrid systems on the basis of performance and overall costs. Such research should be performed at the pilot-scale with membranes having identical pore sizes and configurations. A comprehensive performance evaluation should be performed after each system has been optimized. A detailed cost analysis should also be incorporated into the study. Such a study may provide an accurate assessment of the effects of membrane material in real-world applications. Water utilities and professional consultants may benefit from such a study as it would help them to compare and select the most suitable membrane systems for different source waters.

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