



# The effect of coupling coagulation and flocculation with membrane filtration in water treatment: A review

TorOve Leiknes\*

*NTNU- Norwegian University of Science and Technology, Department of Hydraulic and Environmental Engineering, S. P. Andersensvei 5, N-7491 Trondheim, Norway. E-mail: [torove.leiknes@ntnu.no](mailto:torove.leiknes@ntnu.no)*

## Abstract

Water supply and sanitation demands are foreseen to face enormous challenges over the coming decades to meet the fast growing needs in a global perspective. Significant growth in the industry is predicted and membrane separation technologies have been identified as one of the possible solutions to meet future demands. Application and implementation of membrane technology is expected both in production of potable water as well as in treatment of wastewater. In potable water production membranes are substituting conventional separation technologies due to the superior performance, potential for less chemical use and sludge production, as well as the potential to fulfill hygienic barrier requirements. Membrane bio-reactor (MBR) technology is probably the membrane process which has had most success and has the best prospects for the future in wastewater treatment. Trends and developments indicate that this technology is becoming accepted and is rapidly becoming the best available technology for many wastewater treatment applications. A major drawback of MBR systems is membrane fouling. Studies have shown that fouling mitigation in MBR systems can potentially be done by coupling coagulation and flocculation to the process.

**Key words:** coagulation and flocculation; membrane filtration; potable water; wastewater

## Introduction

A recent global water supply and sanitation assessment reports that the percentage of people served with some form of improved water supply rose from 79% (4.1 billion) in 1990 to 82% (4.9 billion) in 2000. For sanitation purposes an increase from 55% (2.9 billion people served) to 60% (3.6 billion) was registered during the same period. At the beginning of 2000 one-sixth (1.1 billion people) of the world's population was without access to improved water supply and two-fifths (2.4 billion people) lacked access to improved sanitation (WHO/UNIFEC, 2000). At the Second World Water Forum, the Hague, 17–22 March, 2000, targets were presented in the report VISION 21 where the aim was by 2015 to reduce by half the proportion of people without access to sanitation and access to adequate quantities of affordable and safe water, and by 2025 to provide water, sanitation, and hygiene for all (WHO/UNIFEC, 2000). To achieve these targets, it is required providing water supply services to 280.000 people and sanitation facilities to 384.000 people every day for the next 15 years. To reach universal coverage by the year 2025, almost 3 billion people will need to be served with water supply and more than 4 billion with sanitation. The water supply and sanitation sector will face enormous challenges over the coming decades where the water industry will need to meet the fast growing needs in a

global perspective. With this perspective, the global water market has been estimated at a total value of around 224 billion EUR with an anticipated annual growth of around 16%–20% depending on the market segment. Drinking water production is stipulated to have the largest growth with a doubling of the market value in the period 2000–2015. The market for wastewater treatment is the largest sector with an anticipated growth of around 43% for the same period.

Membrane separation technologies have been identified as one of the possible solutions to meet the future demands with regard to water supply and sanitation. Recent studies have stipulated a yearly increase of 7.8% in the demand for membrane materials with a total value of membrane systems (including equipment such as pumps and piping) reaching USD 4.8 billion in 2004. Water and wastewater treatment has been identified as the largest end use for membrane materials, where they will continue to dominate. Implementation and expansion in consumer applications combined with replacement sales to municipal and industrial customers are suggested as the main reasons for this. Cross-flow membrane systems are expected to grow from 4.8 billion EUR in 2004 to 6.5 billion EUR in 2007 on a global basis where desalination has been reported to represent about 1/3 of this growth. The fastest growing segment, however, has been predicted to be the development of membrane bioreactor systems for wastewater treatment with an estimated annual growth rate of 15%.

\* Corresponding author. E-mail: [torove.leiknes@ntnu.no](mailto:torove.leiknes@ntnu.no)

## 1 Membranes in drinking water production

Reverse osmosis (RO) process for desalination of brackish water or sea water to produce potable water is a well established industry. The cost of desalination is expected to drop drastically, as presented at the Fourth World Water Forum, Mexico, 2006. The reports about the implementation of large desalination plants to produce fresh water can often be found in the news media. Although membrane technology applied to production of potable water by desalination is well known, the application of membrane systems in drinking water treatment in general has become more common in recent years. The most common form of drinking water treatment for surface water sources involves chemical/physical removal of particulate matter by coagulation, flocculation, sedimentation, and filtration processes, along with disinfection to inactivate any remaining pathogenic microorganisms. Due to their separation capabilities membranes are currently gaining more interest to substitute conventional separation technologies.

Studies have been conducted to investigate coupling coagulation and flocculation with membrane filtration. Membranes together with appropriate pre-treatment options for fouling minimization, such as coagulation, can be efficiently used for the removal of natural organic matter (NOM) and for the treatment of surface water (Meyn *et al.*, in press). Particle removal mechanisms in membrane filtration differ from those in conventional technologies, ultimately affecting coagulant dosages, points of application, type of coagulant applied, and so on. One of the expected benefits of the membrane process is a reduction of coagulant demand. A recent study has shown that the combination of coagulation, flocculation, and membrane filtration is an efficient and reliable treatment option for surface waters with high NOM concentrations. DOC and color removal were achieved at relatively low coagulant concentrations. Results showed that the coagulant dosage can be reduced while still obtaining high DOC removal if operating at optimal pH conditions. An example of results obtained is illustrated in Fig. 1.

Further studies were conducted to investigate the significance of the flocculation step when coupling coagulation/flocculation with membrane filtration. The objective

was to investigate the overall performance of the process based on the hypothesis that particle characteristics necessary of efficient membrane filtration differ from conventional separation technologies. Three different flocculation modes were tested in parallel to evaluate how these affect removal efficiencies as well as membrane fouling. Flocculation was done with a two stage and one stage paddle flocculation, and in-line flocculation. Typical results are illustrated in Fig. 2. In-line flocculation was found to have similar removal efficiencies at significantly shorter hydraulic retention times (HRT) compared to conventional flocculation. The average shear rate ( $G$ ) value and the retention time for in-line flocculation do not have a significant impact on the membrane fouling rate. The results indicate that coagulation and flocculation coupled with membrane filtration should be based on different design parameters compared to those applied for conventional systems. The use of an efficient in-line flocculation strategy was found to have the potential of giving compact treatment plant designs compared to conventional systems. Further assessment and investigations of these effects are currently being conducted.

An issue in drinking water treatment is the requirement to secure the necessary hygienic barrier effects in a treatment process. Microfiltration (MF) has the capacity to remove pathogenic microorganisms such as protozoa,

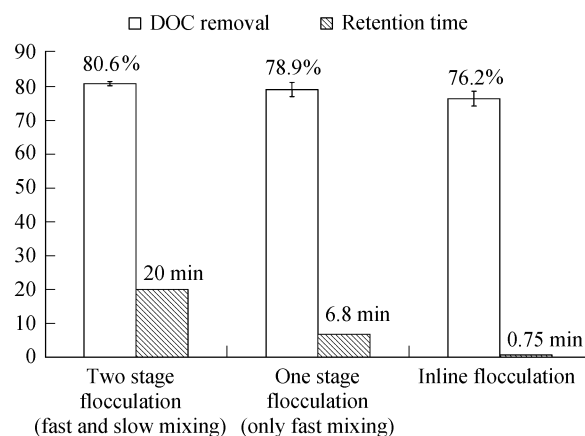


Fig. 2 Illustration of effect of flocculation conditions on DOC removal in surface water treatment.

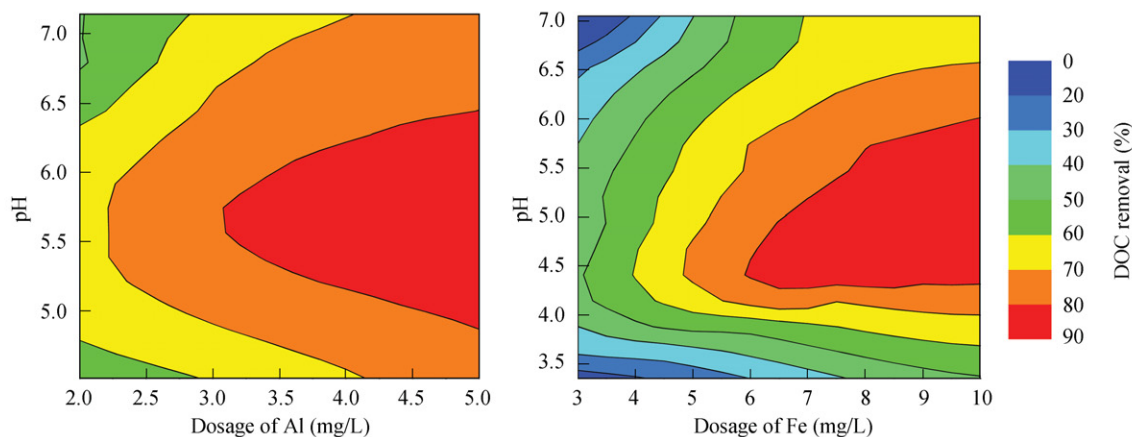


Fig. 1 Illustration of percentage DOC removal for various dosages of Al and Fe based coagulants (Meyn *et al.*, in press).

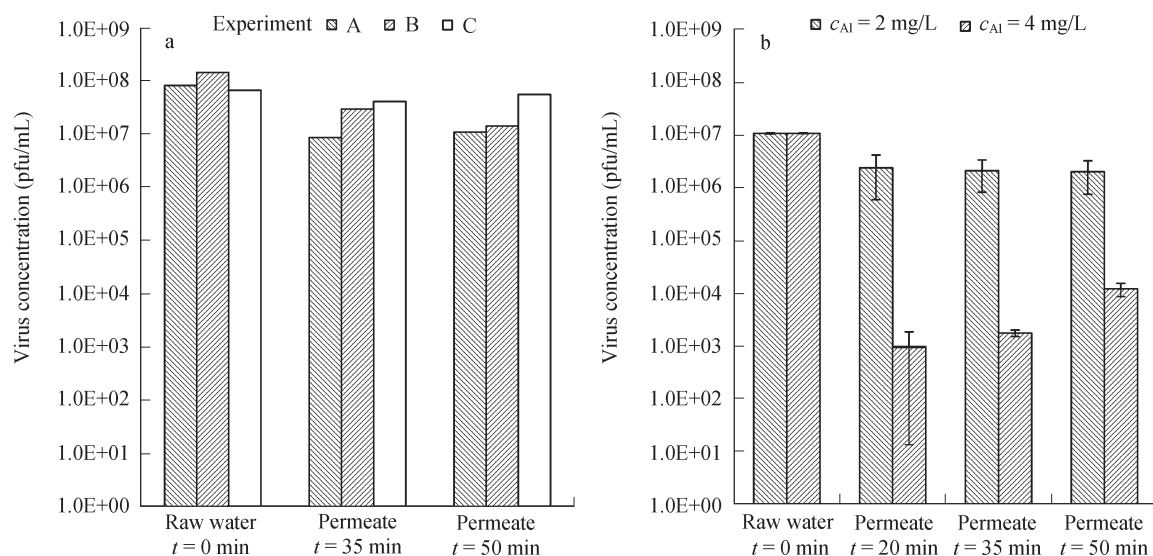
bacteria, and viruses depending on the type of membrane applied. Nanofiltration (NF) technology is generally accepted as being able to remove virus whereas ultrafiltration (UF) technology has been shown to meet current water regulation for turbidity and Giardia. Depending on membrane properties and characteristics, UF membranes are generally capable of removing bacteria while MF membranes also could perform well, though to less degree. Applying MF/UF systems as a hygienic barrier is therefore of concern. More efficient virus retention can potentially be expected when coupling coagulation/flocculation with UF/MF membrane filtration where viruses are adsorbed to or included in larger flocks that are retained by the membrane. Studies related to this topic have shown that without coagulation/flocculation, no virus removal is generally observed for MF membranes and only a minor removal for the UF membranes. With coagulation/flocculation in combination with both UF and MF membrane filtration an effective hygienic barrier against MS2 virus was found, where the MF membrane retained the virus to a similar extent as the UF membrane (Fiksdal and Leiknes, 2006). Figure 3 illustrates the removal of MS2 virus in a MF filtration unit without coagulant and with coagulation/flocculation using two different dosages. Insignificant virus removal is observed when no coagulant is applied. The results indicate that when applying coagulation/flocculation at optimal conditions, a hygienic barrier effect is achieved for the treatment scheme.

## 2 Membranes in wastewater treatment

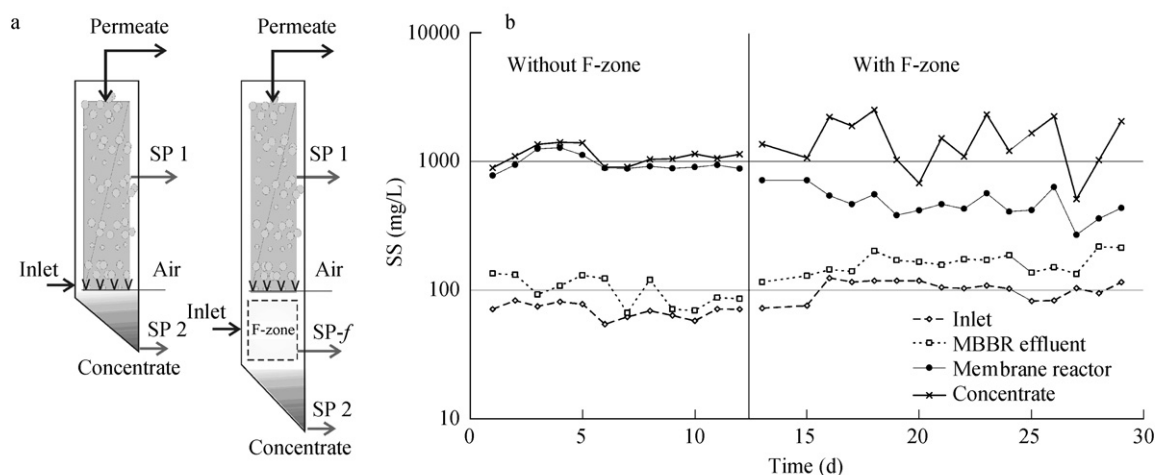
Membrane bioreactor (MBR) is commonly understood as the combination of membrane filtration and biological treatment using activated sludge (AS) where the membrane primarily serves to replace the clarifier in the wastewater treatment system (Gunder and Krauth, 1998; Van der Roest *et al.*, 2002). The first generation of MBRs (late 1970s and 1980s) applied the use of cross-flow operated membranes installed in units outside the activated sludge tank with

high flow velocity circulation pumps. A disadvantage of the cross-flow membranes is the high energy required to generate sufficient sludge velocities across the membrane surface and this process option was therefore considered nonviable for treating municipal wastewater. The development of submerged low pressure configurations in the late 1980s to 1990s, by immersing the membranes into the activated sludge tank, was an important step in making viable commercial solutions for the MBR process (Bouhabila *et al.*, 1998; Cote *et al.*, 1998; Davies *et al.*, 1998). Today a variety of process configurations exist where the membrane is installed either in an external unit or immersed in the aeration tank, where the systems are designed to be operated under low-pressure vacuum. Compared to conventional activated sludge systems, several advantages of AS-MBRs have been identified (Judd, 2006), which have promoted the development of commercial AS-MBR options. These include compact units with small footprints, complete solids removal, effluent disinfection, operation at higher suspended biomass concentrations resulting in long sludge retention times, low sludge production, and no problems with sludge bulking.

A major drawback of MBRs is membrane fouling, which is common for all membrane systems (Judd, 2006). Deposition of solids as a cake layer, pore plugging/clogging by colloidal particles, adsorption of soluble compounds and biofouling are some of the main forms of fouling that have been identified. The significance of colloids and submicron particles on membrane fouling has been reported in literature where different estimations of the total measured fouling caused by these particles vary between 25% and 50% (Wisniewski and Grasmick, 1996; Defrance *et al.*, 2000; Bouhabila *et al.*, 2001; Bae and Tak, 2005). From the results found in literature it is apparent that reduction of submicron particles around the membranes would be desirable in operation of MBR systems. Fouling mitigation in MBR systems can potentially be done by coupling coagulation and flocculation to the process. There are studies to be found in the literature



**Fig. 3** Illustration of MS2 virus removal in a microfiltration unit. (a) without coagulant; (b) with coagulation/flocculation using two dosages.



**Fig. 4** Schemes of BF-MBR reactors with and without an integrated flocculation zone (F-zone) (a) and the variations of suspended solids (SS) over time in those reactors (b). SP: sampling point.

that have investigated improved process performance by using flux-enhancing additives (i.e., coagulants) as well as improved reactor designs to induce flocculation, thereby reducing the number of submicron particles around the membrane. A comprehensive review of these studies is beyond the scope of this article. An example of including flocculation to improve the process performance in a biofilm-MBR is included to illustrate the potential of coupling flocculation with membrane processes (Ivanovic *et al.*, 2008). A BF-MBR process with two different membrane reactors was investigated, one with and one without a flocculation zone, respectively. Analysis of the water in the different flow streams (feedwater, from biofilm reactor, around membrane, concentrate etc.) was done with respect to several parameters and related to membrane performance (i.e., fouling), on the hypothesis that colloids are mainly responsible for fouling. There was no clear correlation found between values measured for suspended solids (SS) and non-filtered and filtered chemical oxygen demand (COD/FCOD) and membrane performance, however, the particle size distribution (PSD) measurements showed a relatively good relationship. In cases where the differential number percentage of submicron particles was high an increase in membrane fouling rates was observed. An illustration of the two reactor configurations with an immersed membrane module and corresponding SS values measured over time at different sampling points (SP) is shown in Fig. 4.

Introducing the feed water inlet port of the membrane reactor through a flocculation zone integrated in the membrane reactor resulted in a reduction of the number of submicron particles and reduction of SS concentrations around the membrane area. The effect was improved membrane performance (i.e., lower fouling rates) and a means for better fouling control in the BF-MBR process. The results confirm that submicron particles in the feed water to the membrane reactor are a significant foulant and that a reduction of this component in the water had a positive effect on membrane performance. A side effect of the alternative membrane reactor design was improved concentrate characteristics with respect to sludge treatment

in that better dewatering and filterability characteristics were measured. Overall, the alternative reactor design and enhanced flocculation resulted in less fouling are therefore, potential strategies for fouling control and minimization by reduction of submicron particles in the wastewater effluent (Ivanovic *et al.*, 2008).

#### Acknowledgments

The essence of material presented is based on ongoing Ph.D studies by Thomas Meyn and Igor Ivanovic at NTNU – Norwegian University of Science and Technology, Department of Hydraulic and Environmental Engineering, and Master's Thesis work undertaken by various MSc students associated with their research projects.

#### References

- Bae T H, Tak M T, 2005. Interpretation of fouling characteristics of ultrafiltration membranes during the filtration of membrane bioreactor mixed liquor. *Journal of Membrane Science*, 264(1-2): 151–160.
- Bouhabila E H, Ben Aïm R, Buisson H, 1998. Microfiltration of activated sludge using submerged membrane with air bubbling (application to wastewater treatment). In: *Proceedings of Conference of Membrane in Drinking and Industrial Water Production*, Amsterdam, 1998.
- Bouhabila E H, Ben Aïm R, Buisson H, 2001. Fouling characterisation in membrane bioreactors. *Separation and Purification Technology*, 22: 123–132.
- Cote P, Buisson H, Praderie M, 1998. Immersed membranes activated sludge process applied to the treatment of municipal wastewater. *Water Science and Technology*, 38(4-5): 437–442.
- Davies W J, Le M S, Heath C R, 1998. Intensified activated sludge process with submerged membrane microfiltration. *Water Science and Technology*, 38(4-5): 421–428.
- Defrance L, Jaffrin M Y, Gupta B, Paullier P, Geaugey V, 2000. Contributions of various constituents of activated sludge to membrane bioreactor fouling. *Bioresource Technology*, 73: 105–112.
- Fiksdal L, Leiknes T, 2006. The effect of coagulation with MF/UF membrane filtration for the removal of virus in drinking water. *Journal of Membrane Science*, 279: 364–

- 371.
- Gunder B, Krauth K, 1998. Replacement of secondary clarification by membrane separation – results with plate and hollow fiber modules. *Water Science and Technology*, 38(4-5): 383-393.
- Ivanovic I, Leiknes T, Ødegaard H, 2008. Fouling control by reduction of submicron particles in a BF-MBR with an integrated flocculation zone in the membrane reactor. *Separation Science and Technology*, 43: 1871–1883.
- Judd Simon, 2006, *The MBR Book-Principles and Applications of Membrane Bioreactors in Water and Wastewater Treatment*. Oxford, UK: Elsevier Ltd.
- Meyn T, Leiknes T, Ødegaard H. Coagulation/flocculation – ceramic membrane filtration for removal of natural organic matter (NOM) under Norwegian conditions. *Water Science and Technology* (in press).
- Van der Roest H F, Lawrence D P, van Bentem A G N, 2002. *Membrane Bioreactors for Municipal Wastewater Treatment*. STOWA Report. London: IWA Publishing.
- WHO/UNICEF (the World Health Organization/the United Nations Children's Fund), 2000. *Global water supply and sanitation assessment report*. WHO/UNICEF Joint Monitoring Programme for Water Supply and Sanitation. ISBN 92 4 156202 1.
- Wisniewski G, Grasmick A, 1996. Floc size distribution in a membrane bioreactor and consequences for membrane fouling. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 138: 403–411.