MEMBRANE TECHNOLOGY FOR ORGANIC REMOVAL IN WASTEWATER

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Contents

Summary

Microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) are presented in terms of fundamental theory and applications. The efficiency of MF, UF, NF and RO in terms of removal of organic matter, endocrine disrupting chemicals (EDCs)/pharmaceutical and personal care products (PPCPs) removal (representation of small molecular weight (MW) compounds) and MW distribution (different MW sizes) is reviewed. The factors affecting membrane fouling and the removal of different organics are extensively dealt with in this chapter.

1. Introduction

Membrane filtration which is classified into microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) is a pressure driven process in which the membrane acts as a selective barrier to restrict the passage of pollutants such as organics, nutrients, turbidity, microorganisms, inorganic metal ions and other oxygen depleting pollutants, and allows relatively clear water to pass through. With technological advances and the ever-increasing stringency of water quality criteria, membrane processes are becoming a more attractive solution to the challenge of producing high quality water from wastewater reuse processes. A number of books have been published on membrane technology in a wastewater treatment plants (WWTPs) (Vigneswaran and Ben Aim, 1989; Ho and Sirkar, 1992; Mallevialle et al., 1996; Mulder, 1996; Water Environment Federation. 2006). The books cover a wide range of theoretical mass transport and application on MF, UF, NF and RO. However, to date, not many studies have dealt with detailed removal processes of organic matter, endocrine disrupting chemicals (EDCs) and pharmaceuticals and personal care products (PPCPs). This chapter focuses on achieving detailed organic removal in terms of dissolved organic carbon (DOC), molecular weight distribution (MWD) and EDCs/PPCPs. DOC indicates the general level of pollutant removal by different membranes used. EDCs and PPCPs represent removal of the smallest compounds (approximately 150–500 Da). MW distribution provides information on the removal of the different ranges of MW.

2. Overview of Membrane Technology

Membrane technology has been applied in various fields of wastewater reuse. MF and UF membrane systems have already proven their advantages in terms of economic efficiency as well as water quality. NF and RO membranes are also used in a broad range of wastewater reclamation.

MF refers to membranes that have pore diameters from 0.1 to 10 μm (Cheryan, 1998) and is the membrane with the largest pores. It can be used to filter suspended particulates, large colloids, bacteria and organics. The MF is also used as a pretreatment for NF and RO processes. Since the pore size of the MF is relatively large, air backflush or permeate backwash can be used to remove the deposits from the pores and surface of the membrane. Physical sieving is the major separation or rejection mechanism in MF. The deposit or cake on the membrane also acts as a self-rejecting layer, and thus MF can retain even smaller particles or solutes than its pore size. Membrane bioreactor (MBR) technology is the most promising development in biological WWTPs. The principal element of the MBR is MF. Now, when economic reasons no longer limit the application of MBR in industrial and municipal WWTPs, and new requirements are being set for WWTPs, MBR may be the key for direct or indirect recycling of wastewaters. This is because two of their characteristics, namely: (a) the low sludge load in terms of BOD, so that the bacteria are forced to mineralize poorly degradable organic compounds; and (b) the long life of the sludge gives the bacteria time to adapt to the treatment-resistant substances. The use of membrane separation technologies in water industry is gaining popularity due to increasing environmental regulations, and capability of membrane to remove most of the pollutants. However, the limitation of using the membrane separation process is: (i) membrane fouling and hence the feed solution should have low solid contents, and it should be operated at low flow rate to minimize the fouling, and (ii) high capital cost. A recent review on MBR can be found elsewhere (Le-Clech et al., 2006).

UF refers to membranes that have pore diameters from 0.001 to 0.02 microns. UF is generally used for the separation of colloids up to a range of 0.001 to 0.1 microns. It enables the concentration, purification and fractionation of macromolecules such as proteins, dyes, and other polymeric materials. It is widely used in the industrial WWTPs where recycling of raw materials, products, and by-products are of primary concern. For example, it can be used to recover paints in the electrophoretic painting industries, and lignin and lignosulforates from black liquor in the pulp and paper industry. UF is also used as a pretreatment to NF and RO processes (Schafer, 2001).

NF has membrane pore size in the range between UF and RO. Simpson et al. (1987) has defined NF as charged UF and is sometimes referred to as a low pressure RO. The NF can remove 50% of hardness, more than 90% of color causing substances and almost all turbidity. The NF has the advantage of low operating pressure compared to RO, and has a high rejection of organics compared to UF. Both charge and size are important in NF rejection. At a neutral pH, most NF membranes are negatively charged. At lower pH, it is positively charged (Zhu and Elimelech., 1997). Physical sieving is the dominant rejection mechanism for the colloids and large molecules. However, for the ions and lower MW organics, chemical interactions between the solutes and membrane can play an important role in rejection mechanisms.

RO was the first membrane process to be widely commercialized. Reverse osmosis is a reversal of the natural process of osmosis in which water from a dilute solution passes through a semi-permeable membrane into a more concentrated solution due to osmotic pressure. In reverse osmosis, an external pressure greater than the osmotic pressure is applied so that the water from concentrated solution passes into the diluted solution. Thus it can be used to separate salts and low MW pollutants from water and wastewater (Vigneswaran et al, 1991).

There are many references on the limits of applications or boundaries of different membranes (Vigneswaran and Ben Aim, 1989; Ho and Sirkar, 1992; Mallevialle et al., 1996; Mulder, 1996; Fane, 1996; Schafer, 2001). However, since the boundary of each membrane is uncertain, many researchers have used different definitions for the choice of membranes. Hence, it is necessary to put forward a detailed and clear definition for the pore size of the membrane. Table 1 presents the classification of different membranes, and thus would avoid overlapping of the definition of pore sizes for different membranes in terms of the tight and loose membranes.

Membrane Process	**RO**	**NF**		**UF**		**MF**	
		Tight	**Loose**	**Tight**	**Loose**	**Tight**	**Loose**
Molecular Weight Cutoff (Da)	< 200 Da	200 to 300	300 to 1000	1000 to 10000	10000 Da to 100000 Da	100000 Da to 0.01 μm	0.01 μm to 0.05 μm

Table 1 Size range of membrane separation process

The major difference between different membrane processes is shown in Table 2.
MF and UF can be considered as the same group due to its porous membrane type. On the other hand, NF and RO have similar characteristics of membrane material,

transport and solute removal. The major difference between NF and RO is that the membrane can remove more than 50% of divalent ions such as calcium (Ca^{2+}) and magnesium (Mg^{2+}).

The membranes can be designed for different support frames (Cardew and Le, 1998). The advantages and disadvantages of different designs of membrane in terms of MF, UF, NF and RO are given in Table 3. The selection of a membrane module is determined by economic considerations, type of application and the functionality of the module. Membrane modules are available in five different designs, flat sheet (plate and frame), hollow fiber, spiral wound, tubular and capillary (of hollow fiber design). The characteristics of the module which must be considered in a system design include packing density, investment cost, fouling tendency, cleaning, operating costs and membrane replacement cost. The qualitative comparison of various membrane configurations are given in Table 4.

Particular	**MF**	**UF**	**NF**	**RO**
Membrane	Porous isotropic	Porous asymmetric	Finely porous asymmetric/ composite	Nonporous asymmetric/ composite
Transfer mechanism	Sieving and adsorptive mechanisms (the solutes migrate by convection)	Sieving and preferential adsorption	Sieving/electrostatic hydration/ diffusive	Diffusive (solutes migrate by diffusion mechanism)
Law governing transfer	Darcy's law	Darcy's law	Fick's law	Fick's law
Typical solution treatment	Solution with solid particles	Solution with colloids and/or macromolecules	Ions, small molecules	Ions, small molecules
Typical pure water flux (L/m^2h)	500 – 10,000	100 – 2,000	20 – 200	10 - 100
Pressure requirement (atm)	0.5 – 5	1 – 10	7 – 30	20 - 100

Table 2 Difference between MF, UF, NF, and RO

Design	**Advantages**	**Disadvantages**
Flat Sheet	Wide choice of membranes	High cost
	Can be dissembled and cleaned	Replacing membrane is time consuming
	Low energy requirement	Can have seal problem
Hollow Fiber	Very compact system	Can be fouled with particulates
	Low liquid hold-up	Not suitable for viscous systems
	Low capital cost	Limited range of products
	Backflushable	
Spiral Wound	Low hold-up	Can have dead spots
	Compact system	Cannot be backflushed
	Wide range of materials	

	Wide range of sizes	
	Low capital cost	
Tubular	Can tolerate feeds with high suspended solids	High energy requirement
	Can work with viscous and non-Newtonian fluids	High capital cost
	Easy to clean mechanically	Large space demand
		Disassembly long
		High hold-up

Table 3 Advantages and disadvantages of membrane designs

Characteristics	**Tubular**	**Flat sheet**	**Spiral wound**	**Capillary**	**Hollow fiber**
Packing density	Lowest	Lower	Low	High	Higher
Investment cost and Installed area	Highest	Higher	High	Low	Lower
Fouling tendency	Lowest	Lower	Low	High	Higher
Ease of cleaning	Best	Better	Good	Poor	Poorer
Operating cost	Highest	Higher	High	Low	Lower
Membrane replacement	Yes/No	Yes	No	No	No

Table 4. Qualitative comparison of various membrane configurations

2.1 Theory

The application of membrane processes in WWTPs has increased since the appearance of synthetic asymmetric membranes in 1960 (Ridgway et al., 1996). A number of mathematical models have also been developed to describe membrane filtration. The transport models developed have been classified into different groups: i) porous and nonporous membranes, ii) organic and inorganic and iii) different sizes of organic matter. However, it should be noted that a comprehensive understanding of the parameters influencing the mass transfer of solutes is invaluable to a predictive model of membrane filtration.

The transport models developed for nonporous membranes (NF and RO) consist of three types: i) homogeneous membrane models (solution-diffusion, extended solution-diffusion and solution-diffusion-imperfection models), ii) pore-based models (preferential sorption-capillary flow, finely porous and surface force-pore flow models) and iii) irreversible thermodynamic models (Kedem-Katchalsky and Spiegler-Kedem models) (Bhattacharyya and Williams, 1992). The models of porous membranes (UF and MF) can be classified into: i) basic models based on Hagen-Poiseulle equation and Kozeny-Carman relationship), ii) Knudsen flow, iii) friction model and iv) concentration polarization (CP) model (resistance in series model, osmotic pressure model and mechanistic interpretation) (Mulder, 1996).

These models can be divided into four groups in terms of organic and inorganic characteristics of solutes in Figure 1.

(i) the non-charged colloids which follow mainly the CP relationship, convection and diffusion, Nernst-Plank equation, resistance in series and cake filtration theory.

(ii) the charged colloids which involve a relationship of convection and diffusion, Donnan exclusion, extended Nernst-Plank equation, resistance in series and cake filtration theory.

(iii) general organic matter which follows the CP relationship, thermodynamic model, diffusivity, resistance in series and adsorption layers.

(iv) ions (anions) which obey Donnan exclusion and extended Nernst-Plank equation.

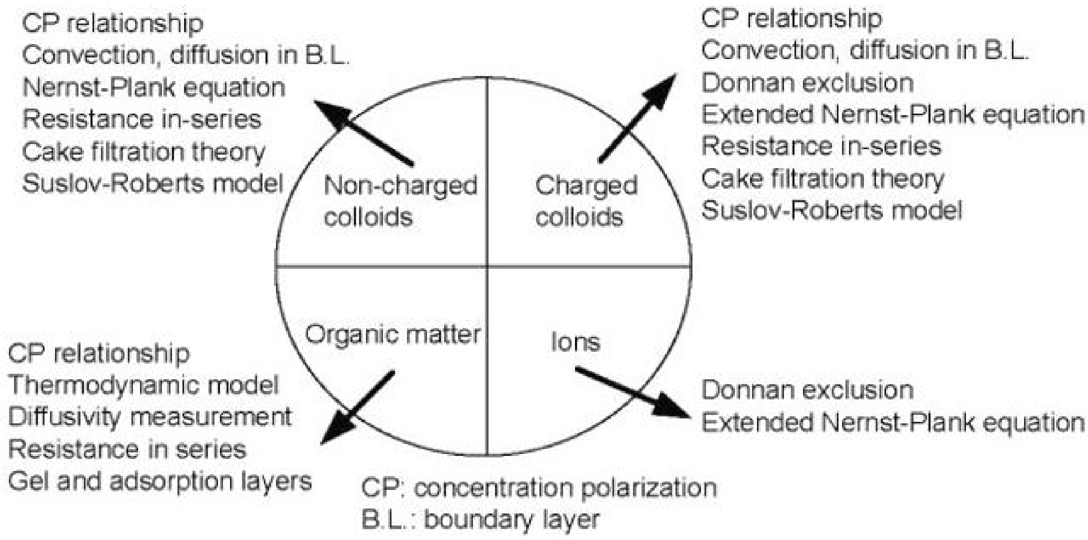

Figure 1 Concept of membrane transport phenomena in terms of different solutes

2.2 Membrane Fouling

Membrane fouling has been the major challenge to the better operation of the membrane processes. Membrane fouling can occur due to the following reasons: (i) biological fouling which is the growth of biological species on the membrane surface, (ii) colloidal fouling which results in a loss of permeate flux through the membrane, (iii) organic fouling due to the deposition of organic substances, and iv) scaling which is defined as the formation of mineral deposits precipitating from the feed stream to the membrane surface (Duranceau, 2001).While biofouling is important in the long term, most likely, biofouling occurs only after organic, inorganic or colloidal fouling. Since interactions between solutes and the membranes are poorly understood, it is possible that effects like charge interactions, bridging, and hydrophobic interactions may play an important role in membrane fouling. Normally, membranes with larger pores exhibit a greater flux decline as filtration proceeds. This is due to internal clogging. However, flux decline is not necessarily due to fouling. Concentration polarization or osmotic pressure or membrane compaction can appear as fouling.

Membrane foulants consist of complex compound, in a range of sizes and affected by the type of transport. As time proceeds, the foulants form different structures, type and layers on the membrane surface, depending on pressure applied and the progress of biofouling. In addition, different membranes and water characteristics form different types of membrane foulants. This leads to a difficulty in understanding the issue of membrane foulants. Table 5 presents the distribution of reasons for selecting membrane process. Although this information applies to drinking water sources, it provides a starting point for ranking of membrane foulants in wastewater sources. The top rank reasons of selecting membrane processes are in the order of biofouling, scaling, organic fouling and particulate fouling (Mickley et al., 1993).

Ranking	Foulants	Foulant type
1	Bacteria	Biofouling (attachment -> growth -> soluble microbial products -> detachment)
	HCO_3	Scaling (precipitation of calcium carbonate by RO concentrate)
2	Organics	Organic fouling (primary cause of chronic fouling)
	Turbidity	Particulate fouling (deposition of suspended matter, colloids and micro-organisms on the membrane)

Table 5 Distribution of reasons for selecting membrane process

Organic fouling which is the initial cause of membrane fouling is related to the molecular size, shape and chemical characteristics (steric, polar, functional group and stability to form hydrogen bond) of organic matter. The organic fouling can occur due to adsorption, precipitation, and the interactions with cations. Therefore, depending on the characteristics of the organics, the membrane type and its operating condition need to be selected. Organic fouling is normally irreversible and needs careful chemical treatment.

Membrane fouling in terms of the size of organic matter is different with MF, UF, NF and RO. Membrane fouling of MF and UF which are porus is significantly affected by suspended solid, particulate organic and large organic matter, while that of NF and RO which are non-porus and come into contact with more smaller size of organic matter due to pretreatment is caused by less than 30 kDa of organic matter. Kim et al. (2008) reported that the inorganic and organic weight percentages of the NF foulant in North Buffalo wastewater, USA after MF and UF pretreatment were nearly equal (56% vs. 44%) on the membrane surface. Extensive autopsy analyses on the NF showed there was between 35-56% less organic carbon where UF pretreatment was used rather than MF. Chellam et al. (1997) found that colloidal materials could cause more fouling than dissolved organic matter in NF. DiGiano et al. (1994) found that the molecular weight greater than 30 kDa of organic matter was responsible for NF fouling, while Kaiya et al. (1996) observed that the organic compounds with MW larger than 100 kDa are major foulants in MF. They further noticed the change in fouling mechanism after 20 h operation of NF, possibly due to the interactions of the hydrophobic and hydrophilic fractions of organics. Membrane fouling would be very severe in positively charged membranes which can attract the negatively charged organics easily (Nystrom et al., 1995).

Many researchers have suggested that the humic substances fraction of organic matter is a major foulant which controls the rate and extent of fouling (Combe et al., 1999; Jones and O'Melia, 2000; Yuan and Zydney, 1999). However, recent studies have reported that hydrophilic (non-humic) organic matter is a more significant foulant. Wiesner et al. (1992) identified that proteins, aminosugars, polysaccharides, and polyhydroxyaromatics were strong foulants. In the studies performed by Lin et al. (2000) and Carroll et al., (2000), the rate of fouling was reduced after coagulation pretreatment. Fan et al., (2001) identified potential foulants in order as hydrophilic neutrals > hydrophobic acids > transphilic acids. Macromolecules of a relatively hydrophilic character (e.g. polysaccharides) were effectively rejected by low-pressure membranes, suggesting that macromolecular compounds and/or colloidal organic matter in the hydrophilic organic fraction may be a problematic foulant for low-pressure membranes. As the filtration through the membrane proceeds, the pore of the membrane is blocked by organic and inorganic substances reducing the effluent flux through the membrane. The blockage of the pores of the membrane is known as membrane fouling. Normally, membranes with larger pores exhibit a greater flux decline. It should be noted that the flux decline is not necessarily due to the membrane fouling only. Concentration polarization, or osmotic pressure or membrane compaction can cause flux decline. Therefore careful experimental study is necessary to distinguish membrane fouling from other effects.

Inorganic ions such as calcium, phosphorus, aluminium and iron etc. were found to enhance the membrane fouling during water treatment processes (Baker et al., 1995). Hong and Elimelech (1997) observed that membrane fouling by organic matter was increased in the presence of calcium ions, at decreased pH, and increased ionic strength. They further noted that permeation drag and electrostatic double layer repulsion controlled the membrane fouling.

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Dr J. Cho, associate professor at Gwangju Institute of Science and Technology, has been studying on research for water reuse using various technologies, including constructed wetland and membrane filtration. He is recently interested in research of ecological engineering as well as related education. He is an editorial board member of Journal, Water Science and Technology, IWA and newsletter editor of Water Reuse Specialty Group in IWA.

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