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Terminology for Membrane Filtration

Term	Definition
Asymmetric membrane	Membrane whose morphology (structure) varies significantly across the thickness of the membrane.
Cross-flow filtration	Filtration technique in which the feed stream is pumped at high velocity parallel to the membrane surface to reduce the collection of retained species at the membrane surface.
Dalton	Unit for molecular weight, equal to one-twelfth of the mass of a carbon-12 atom. Also equal to the molar mass in units of grams per mole. Equivalent to atomic mass units (amu).
Dead-end filtration	Filtration technique in which the feed stream is directed toward and perpendicular to the membrane surface.
Fouling	Process resulting in loss of performance of a membrane due to the deposition of suspended or dissolved substances on its external surfaces, at its pore openings, or within its pores.
Homogeneous membrane	Membrane with consistent morphology and transport properties throughout its thickness.
Lumen	Bore, or cavity, in the center of a hollow fiber membrane.
Molecular weight cutoff	See <i>Retention rating</i> .
Packing density	Membrane area per unit volume in a membrane module.
Permeability	Specific flux of clean, deionized water through a new membrane.

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Term	Definition
Permeate	Water and permeable components that pass through a membrane.
Retentate	Solution containing water and impermeable components retained on the feed side of a semipermeable membrane.
Retention rating	Designation for the size of materials retained by a membrane. The retention rating is called the pore size in micrometers for microfiltration (MF) membranes and the molecular weight cutoff (MWCO) in daltons for ultrafiltration (UF) membranes.
Semipermeable membrane	Membrane that is permeable to some components in a feed solution and impermeable to other components.
Specific flux	Flux divided by transmembrane pressure.
Straining	Process in which particles are retained because they are physically larger than the void spaces in the filter medium (often called sieving).
Transmembrane pressure	Differential pressure between the feed and permeate sides of a membrane.

Note: Additional membrane nomenclature is available in Koros et al. (1996) and ASTM (2001b).

Membrane processes are modern physicochemical separation techniques that use differences in permeability (of water constituents) as a separation mechanism. During membrane treatment, water is pumped against the surface of a membrane, resulting in the production of product and waste streams, as shown on Fig. 12-1. The membrane, typically a synthetic material less than 1 mm thick, is *semipermeable*—meaning that it is highly permeable to some components in the feed stream and less permeable (or impermeable) to others. During operation, permeable components pass through the membrane and impermeable components are retained on

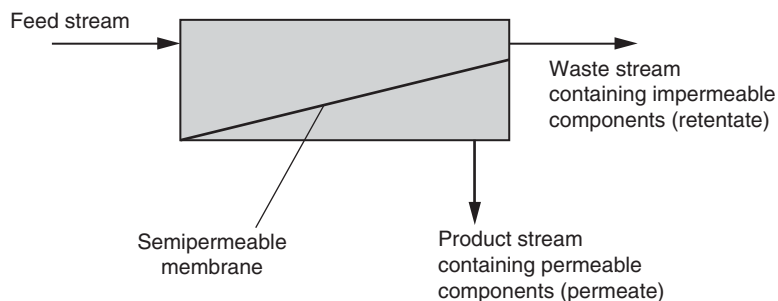


Figure 12-1
Schematic of separation process through semipermeable membrane.

the feed side. As a result, the product stream is relatively free of impermeable constituents and the waste stream is concentrated in impermeable constituents.

12-1 Classification of Membrane Processes

Four types of pressure-driven membranes are currently used in municipal water treatment: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) membranes. The hierarchy of membrane processes is shown on Fig. 12-2. The distinction between the types of membranes is somewhat arbitrary and subject to differing interpretations, but the membranes are loosely identified by the types of materials rejected, operating pressures, and nominal pore dimensions (which are identified on an order-of-magnitude basis on Fig. 12-2). A “loose” NF membrane marketed by one manufacturer might be substantially similar to a “tight” UF membrane marketed by another manufacturer. As used in water treatment, these membranes can be classified into two distinct physicochemical processes: (1) membrane filtration and (2) reverse osmosis.

Membrane Filtration

Membrane filtration is the focus of this chapter and encompasses the use of MF and UF membranes. Filtration can be broadly defined as a process that separates suspended particles (a dispersed solid phase) from a liquid phase by passage of the suspension through a porous medium (either membranes or granular media). In membrane filtration, the feed stream is a suspension,

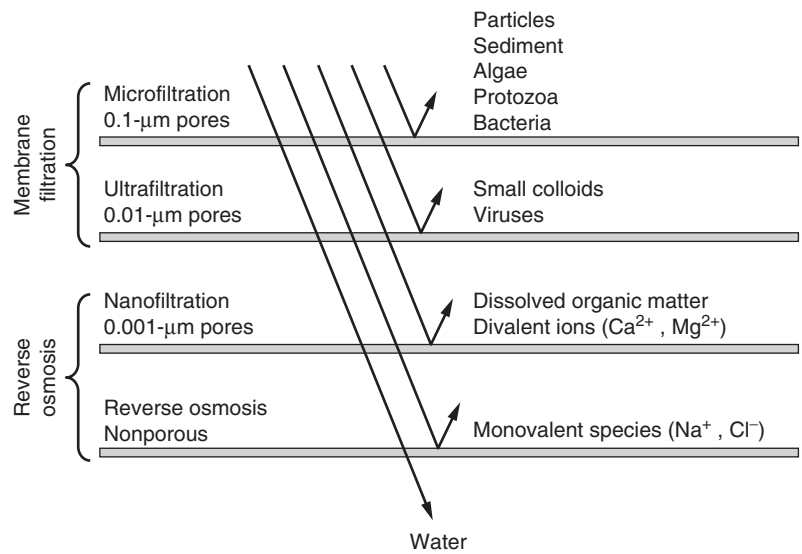


Figure 12-2
Hierarchy of pressure-driven membrane processes.

or two-phase system, in which the dispersed solid phase to be separated may include sediment, algae, bacteria, protozoa, viruses, or colloids. The primary goal of membrane filtration is to produce a product stream (water) from which the targeted solids have been completely removed, which is similar to the goal of granular filtration. While used for similar purposes, MF and UF membranes have important differences that will be described later in this chapter.

The other fundamental physicochemical membrane process is *reverse osmosis*. Reverse osmosis is the focus of Chap. 17 and encompasses the use of NF and RO membranes. *Osmosis* is the preferential diffusion of water through a semipermeable membrane in response to a concentration gradient. In reverse osmosis, the feed stream is a solution, or single-phase system, in which the constituents targeted for removal are truly dissolved solutes (ions such as sodium, chloride, calcium, or magnesium, and dissolved NOM). The primary goal of reverse osmosis is to reduce the concentration of these solutes in the product water. Reverse osmosis membranes are used to produce potable water from ocean or brackish water and to remove specific dissolved contaminants (e.g., pesticides, arsenic, nitrate, radionuclides). Nanofiltration membranes are used to soften hard waters (remove calcium and magnesium ions), freshen brackish waters, and reduce the concentration of NOM to control disinfection by-product (DBP) formation.

Reverse Osmosis

The differences between membrane filtration and reverse osmosis are substantial. The predominant removal mechanism in membrane filtration is straining, or size exclusion, so the process can theoretically achieve perfect exclusion of particles regardless of operational parameters such as influent concentration and pressure. Mass transfer in reverse osmosis, however, involves a diffusive mechanism so that separation efficiency is dependent on influent solute concentration, pressure, and water flux rate. Differences between membrane filtration and reverse osmosis are evident in the materials used for the membranes, the configuration of the membrane elements, the equipment used, the flow regimes, and the operating modes and procedures. Additional comparisons between membrane filtration and reverse osmosis are detailed in Table 12-1. It should be noted that membranes are used for many purposes in a wide variety of fields and industries, and the distinction between membrane types as used in water treatment may not be appropriate in other industries. For instance, UF membranes are used in food-processing and pharmaceutical industries for purifying, concentrating, and fractionating concentrated solutions of macromolecules such as proteins and polysaccharides; UF membrane use in those industries involves phenomena (such as concentration polarization) described in Chap. 17.

Differences between Membrane Processes

Table 12-1

Comparison between membrane filtration and reverse osmosis

Process Characteristic	Membrane Filtration	Reverse Osmosis
Objectives	Particle removal, microorganism removal	Seawater desalination, brackish water desalination, softening, NOM removal for DBP control, specific contaminant removal
Target contaminants	Particles	Dissolved solutes
Membranes types	Microfiltration, ultrafiltration	Nanofiltration, reverse osmosis
Typical source water	Fresh surface water (TDS < 1000 mg/L)	Ocean or seawater, brackish groundwater (TDS = 1000–20,000 mg/L), colored groundwater (TOC > 10 mg/L)
Membrane structure	Homogeneous or asymmetric	Asymmetric or thin-film composite
Most common membrane configuration	Hollow fiber	Spiral wound
Dominant exclusion mechanism	Straining	Differences in solubility or diffusivity
Removal efficiency of targeted impurities	Frequently 99.9999% or greater	Typically 50–99%, depending on objectives
Most common flow pattern	Dead end	Tangential
Operation includes backwash cycle	Yes	No
Influenced by osmotic pressure	No	Yes
Influenced by concentration polarization	No	Yes
Noteworthy regulatory issues	Challenge testing and integrity monitoring	Concentrate disposal
Typical transmembrane pressure	0.2–1 bar (3–15 psi)	5–85 bar (73–1200 psi)
Typical permeate flux	30–170 L/m ² · h (18–100 gal/ft ² · d)	1–50 L/m ² · h (0.6–30 gal/ft ² · d)
Typical recovery	>95%	50% (for seawater) to 90% (for colored groundwater)
Competing processes	Granular filtration	Carbon adsorption, ion exchange, precipitative softening, distillation

Table 12-2

Non-pressure-driven membrane processes

Membrane Process	Driving Force
Dialysis	Concentration gradient
Electrodialysis	Electrical potential
Electrodialysis reversal	Electrical potential
Pervaporation	Pressure gradient
Forward osmosis	Osmosis
Membrane distillation	Vapor pressure
Thermosmosis	Temperature gradient

It should be noted that membrane filtration and reverse osmosis are both pressure-driven membrane processes. Driving forces other than pressure are used in other membrane processes, including some that are occasionally used in water treatment, such as electrodialysis. Other membrane processes (not covered in this text because of their limited applicability in water treatment) and their driving forces are identified in Table 12-2.

12-2 History of Membrane Filtration in Water Treatment

Microporous membranes were first patented in the 1920s (Belfort et al., 1994) and were limited primarily to laboratory use until the 1950s. They were used primarily for enumerating bacteria, removing microorganisms and particles from liquid and gas streams, fractionating and sizing macromolecules such as proteins, and diffusion studies. The U.S. Public Health Service (U.S. PHS) adopted membrane filtration as a method for identifying coliform bacteria in 1957.

In the 1950s, industrial users started applying membrane filtration to larger scale industrial use, with one common use being sterilization of liquid pharmaceuticals and intravenous solutions. Membrane filtration was used in food-processing industries for clarifying, concentrating, purifying, or sterilizing various products such as fruit juices, dairy products, vegetable oils, and alcoholic beverages. Membrane filtration also began to be used for industrial process and waste treatment—such as oily wastewater treatment, caustic acid, and brine recovery—and treatment or recovery of various other industrial waste streams.

The first interest in membrane filtration for potable water production began in the 1980s as utilities and regulators became increasingly concerned about microbiological contamination. Advances in industrial equipment design and operation, including the introduction of dead-end flow regimes, outside-in hollow-fiber flow configurations, and backwashing systems, made

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Drinking Water
Treatment**

the production of drinking water by membrane filtration an economically realistic possibility. Very small utilities, in particular, began to consider membrane filtration. Rapid granular filter equipment was expensive and required a level of operator attention and sophistication that was sometimes unaffordable for small communities, and membrane filtration offered an attractive, highly automated, operationally simple alternative. The first membrane filtration plant used for drinking water production in the United States was a 225-m³/d (0.06-mgd) plant at Keystone Resorts in Colorado in 1987 (U.S. EPA, 2001). Similar developments occurred in Europe, and a 250-m³/d (0.07-mgd) UF plant was installed in France in 1988 (Anselme et al., 1999).

The passage of the SWTR (U.S. EPA, 1989) in 1989 provided utilities with another reason to consider membrane filtration. Regulatory agencies were focusing greater attention on microorganisms in water supplies and lower turbidity levels were required. Membrane filtration offered the potential of higher quality treatment than granular filtration. Still, the use of membrane filtration grew slowly, and by 1993 there were only eight systems installed in the United States, all considerably smaller than 3800 m³/d (1 mgd) (U.S. EPA, 2001).

Effectiveness of Membrane Filtration for Removing Protozoa

As noted in Chap. 1, an outbreak of cryptosporidiosis in Milwaukee, Wisconsin, in 1993 caused over 400,000 illnesses and 50 deaths (Craun et al., 1998; U.S. EPA, 1998). During the incident, *Cryptosporidium* oocysts had passed through the conventional water treatment plant, including the rapid granular filters. The outbreak underscored the fact that the effluent water quality from rapid granular filters is dependent on proper chemical conditioning of the feed water, which is ultimately dependent on operators' judgment, experience, and knowledge of water chemistry. In contrast, membrane filtration removes particles by straining so that complete removal of protozoa is virtually guaranteed as long as the membranes are intact.

Afterward, the view that membranes provided superior filtration helped fuel a rapid increase in the installation of membrane filtration plants, with growth rates in installed capacity of 50 to 100 percent per year over the next several years. Costs of membrane filtration facilities dropped dramatically during this period as a result of advances in technology, mass production, and the entry of additional manufacturers into the market. A survey of equipment manufacturers revealed over 700 membrane filtration facilities in operation worldwide by the end of 2003. In North America, 213 plants [with capacity greater than 379 m³/d (0.1 mgd)] had a total installed capacity of 2.3 million m³/d (620 mgd) by the end of 2003 (Adham et al., 2005). Interest in membrane filtration has continued unabated since then. Membrane filtration is now considered a viable option for any surface water treatment facility of any size. The decision to use granular filtration or membrane filtration in any particular facility depends on site-specific circumstances.

12-3 Principal Features of Membrane Filtration Equipment and Operation

Membrane filtration occurs when water is forced through a thin wall of porous material. The filter medium is not woven or fibrous like cloth but is a continuous mass with tortuous interconnecting voids, as shown in the scanning electron microscope (SEM) images on Fig. 12-3. Nearly all membrane filtration systems installed in the United States use polymeric membranes. Polymeric membranes are almost always configured as hollow fibers, as shown on Figs. 12-4a and 12-4b. The fibers have an outside diameter ranging from about 0.65 to 2 mm and a wall thickness (i.e., membrane thickness) ranging from about 0.1 to 0.6 mm. Although the hollow fiber configuration is the most common used in water treatment, other configurations exist and are in widespread use in other industries. Membrane filtration is a rapidly evolving field, and other configurations might be used in the future. Ceramic membranes are used in some systems in Japan, and the first large ceramic membrane system in the United States was designed for Parker, Colorado, and is expected to be operational in 2012. Ceramic membranes have a tubular configuration with many parallel channels in a rigid monolithic element, as shown on Fig. 12-4c. The configuration has a strong effect on the *packing density*, or membrane area per unit of volume of equipment module, which can be

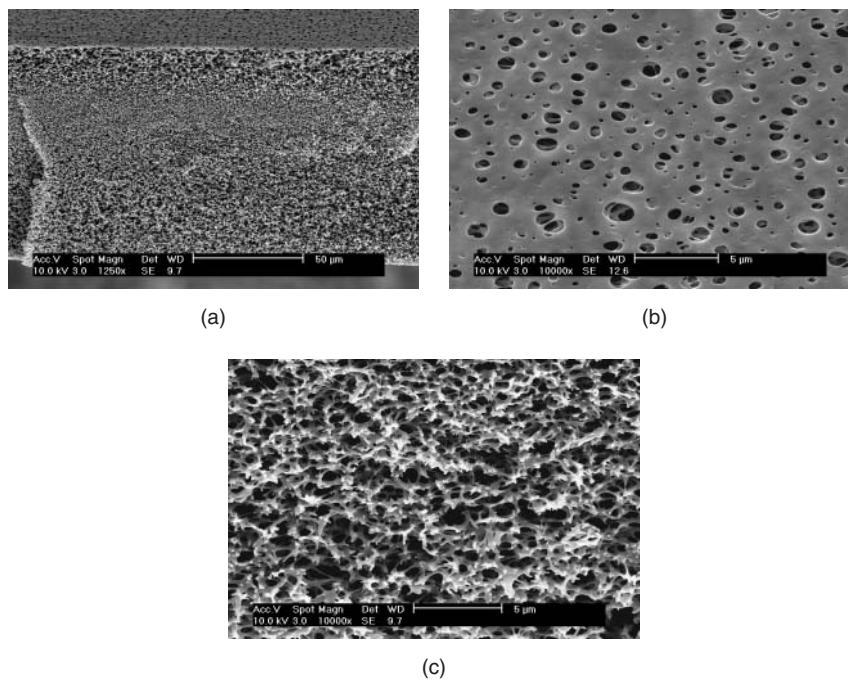


Figure 12-3
Scanning electron microscope images of a 0.2-μm polyethersulfone microfiltration membrane: (a) cross section of the entire membrane, (b) high magnification of the membrane surface, and (c) high magnification of the membrane internal structure.

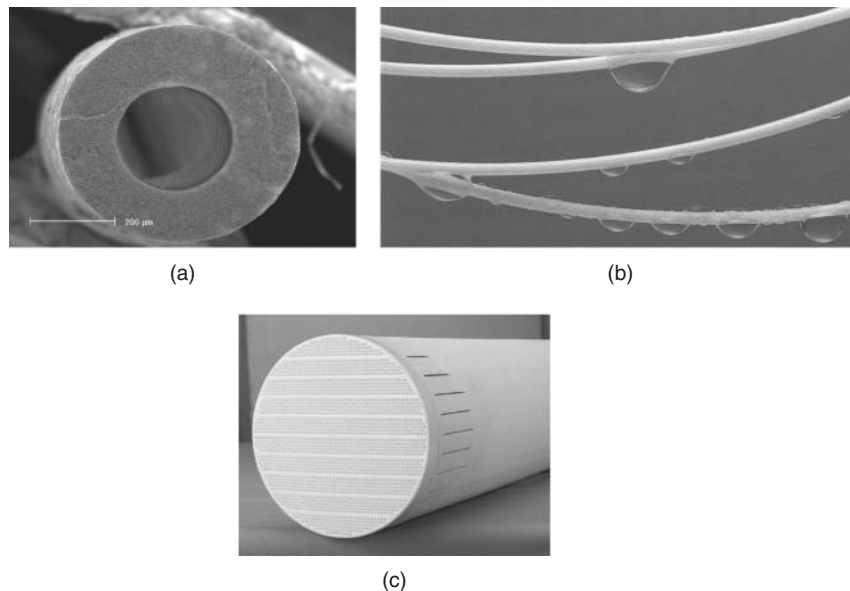


Figure 12-4
(a) Scanning electron microscope image of end view of a hollow-fiber membrane (courtesy of US Filter Memcor Products), (b) water permeating hollow-fiber membranes (courtesy of Suez Environnement), and (c) end view of a ceramic tubular membrane (courtesy of NKG).

an important consideration in the cost effectiveness of membrane plants. Other membrane configurations are described in Table 12-3.

The water passing through the membrane is called permeate, and water remaining on the feed side is called retentate. As solids accumulate against the filter medium, the head across the membrane required to maintain constant flux increases. The difference in pressure between the feed and permeate is known as the transmembrane pressure. The transmembrane pressure is between 0.2 and 1 bar (3 and 15 psi) for most membrane filtration systems. Keeping pressure below 1 bar (15 psi) helps minimize membrane fouling.

Membrane filters operate over a cycle consisting of two stages, just like granular filters: (1) a filtration stage, during which particles accumulate, and (2) a backwash stage, during which the accumulated material is flushed from the system. During the backwash cycle, air and/or water is used to remove accumulated solids. Typical permeate flux, operating pressure, and duration of filter and backwash cycles, along with a comparison to rapid granular filtration, are presented in Table 12-4. Although the backwash removes accumulated solids, a gradual but continuous loss of performance is observed over a period of days or weeks, as shown on Fig. 12-5. The loss of performance, or fouling, is due to slow adsorption or clogging of material that cannot be removed during backwash. Fouling affects the cost effectiveness of membrane filtration and will be discussed in detail later in this chapter. Fouling is minimized by periodically adding chemicals to the backwash cycle, known as chemically enhanced backwash (CEB),

Table 12-3
Membrane configurations

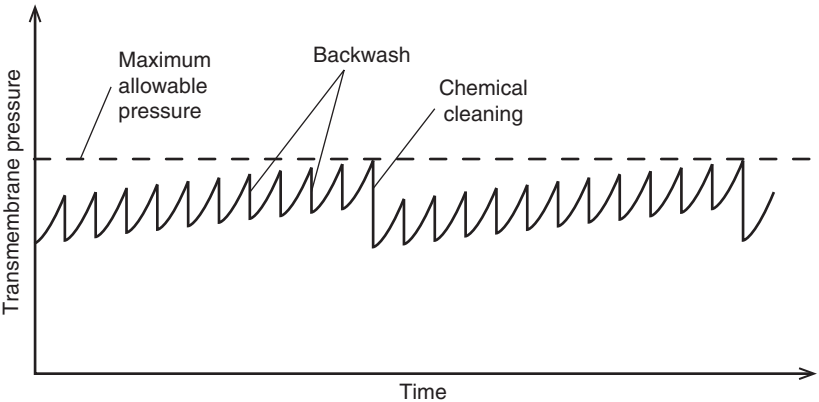
Configuration	Description
Hollow fiber	Membranes are cast as hollow tubes and filtration occurs as water passes through the wall of the fibers (see Fig. 12-4b). The module packing density (specific surface area) is 750–1700 m ² /m ³ .
Tubular	Membranes are constructed as a monolithic structure with one or more channels through the structure (see Fig. 12-4c). Ceramic membranes are typically tubular membranes. These membranes can be operated at a high cross-flow velocity, which is ideal for applications where the particle concentration is high. The module packing density is up to 400–800 m ² /m ³ .
Flat sheet	Membranes are cast as a sheet and used as a single layer or as a stack of sheets. Common in laboratory separations but not as common at an industrial scale. Packing density depends on spacing of the sheets.
Spiral wound	Flat-sheet membranes, stacked in layers separated by permeate and retentate spacers, then rolled around a central tube so that the permeate travels in a spiral flow path toward the central collection tube. Common in NF and RO membranes but not in wide use for membrane filtration due to clogging of flow paths with particulate matter and problems with backwashing effectively. See Chap. 17 for additional details on the construction of spiral-wound elements. The packing density is 700–1000 m ² /m ³ .
Hollow fine fiber	Membranes cast as hollow tubes with an outside diameter of 0.085 mm (about the thickness of human hair). Hollow fine fibers are used only as RO membranes; see Chap. 17 for additional details. The packing density is 5600–7400 m ² /m ³ .
Track etched	Flat-sheet membranes that are cast as a dense sheet of polymer material and exposed to a radioactive beam, which damages the material along “tracks,” or straight pathways through the material. The material is then immersed in an etching bath that dissolves the material along the pathways, widening the tracks to form pores of uniform cylindrical dimensions. The result is a flat-sheet membrane with a narrow, controllable, and extremely uniform pore size distribution, which is advantageous in laboratory separations. Track-etched membranes are not currently used in industrial-scale applications.

Table 12-4
Operating characteristics of membrane and rapid granular filters

Criteria	Membrane Filtration	Rapid Granular Filtration
Filtration rate (permeate flux)	0.03–0.17 m/h ^a (0.01–0.07 gpm/ft ²)	5–15 m/h ^a (2–6 gpm/ft ²)
Operating pressure	0.2–1 bar (7–34 ft)	0.18–0.3 bar (6–10 ft)
Filtration cycle duration	30–90 min	1–4 d
Backwash cycle duration	1–3 min	10–15 min
Ripening period	None	15–120 min
Recovery	>95 %	>95 %
Filtration mechanism	Straining	Depth filtration

^aConventional units for membrane permeate flux are L/m² · h and gal/ft² · d. The conversions to the units shown in this table are 1 L/m² · h = 10^{−3} m/h and 1440 gal/ft² · d = 1 gpm/ft².

Figure 12-5
Transmembrane pressure development during membrane filtration.



and periodic chemical cleaning, known as the clean-in-place (CIP) cycle. CIP typically involves soaking the membranes for several hours in one or more warm solutions containing surfactants, acids, or bases. The cleaning frequency may range from a few days to several months, depending on the membrane material, operating conditions, and raw-water quality. The membranes degrade over a longer period of time, and replacement may be necessary after a period of 5 to 10 years.

The increase in transmembrane pressure when filters are operated in a constant-flux, rising-pressure mode is shown on Fig. 12-5. Full-scale facilities are operated in this mode because of production capacity requirements. In contrast, laboratory studies are sometimes performed in a

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constant-pressure, declining-flux mode to accommodate equipment capabilities and data analysis procedures.

As shown in Table 12-4, the flux through a membrane filter is typically about two orders of magnitude lower than the flux through a rapid granular filter; consequently, a membrane filtration plant needs 100 times the filter area of a rapid granular filtration plant to produce the same quantity of water. One characteristic of membrane filtration plants, however, is that they are frequently more compact than granular filtration plants. This apparent contradiction is possible because membrane plants are constructed by packing thousands of hollow fibers into modules; thus, 1 m² of floor space at a membrane plant may contain more than 100 m² of membrane area. Membrane modules are available in two basic configurations: pressure-vessel systems or submerged systems.

Module Configuration

PRESSURE-VESSEL CONFIGURATION

Pressure-vessel modules are generally 100 to 300 mm (4 to 12 in.) in diameter, 0.9 to 5.5 m (3 to 18 ft) long, and arranged in racks or skids. Typical pressure-vessel membrane elements are shown on Fig. 12-6. A single module has thousands of fibers and typically contains between 40 and 80 m² (430 and 860 ft²) of filter area. The rack or skid is the basic production unit, and all modules within one rack are operated in parallel simultaneously (see Fig. 12-7). Racks can contain between 2 and 100 modules, depending on capacity requirements. Feed pumps typically deliver water

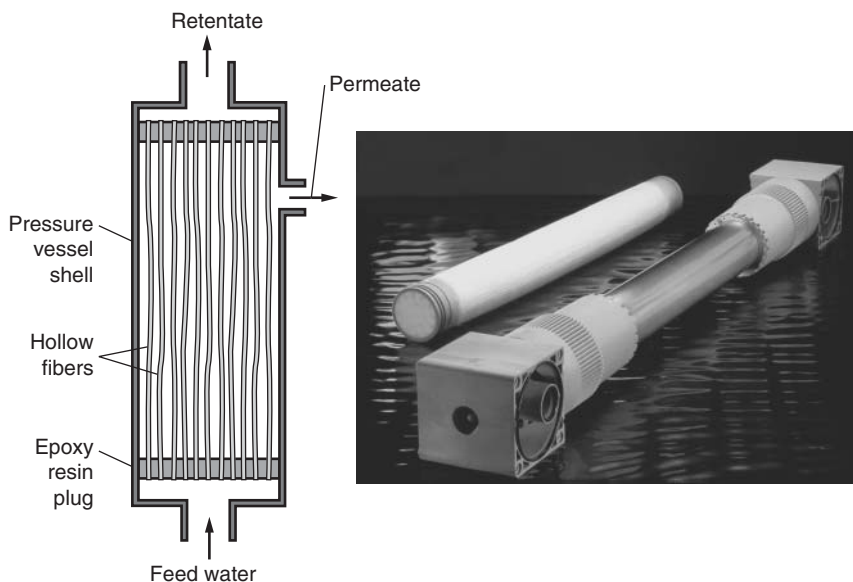


Figure 12-6
Pressure-vessel configuration for membrane filtration: (a) schematic of a single cross-flow membrane module and (b) photograph (courtesy of US Filter Memcor Products).

Figure 12-7
Full-scale membrane
filtration facility using the
pressure-vessel
configuration.



to a common manifold that supplies each rack. Each module must be piped individually for feed and permeate water, so large racks involve a substantial number of piping connections. Transmembrane pressure is developed by a feed pump that increases the feed water pressure, while the permeate stays at near-atmospheric pressure. Pressure-vessel systems typically operate at transmembrane pressures between about 0.4 and 1 bar (6 and 15 psi).

SUBMERGED CONFIGURATION

Submerged systems (also called immersed membranes) are modules of membranes suspended in basins containing feed water, as shown on Fig. 12-8. The basins are open to the atmosphere, so pressure on the influent side is limited to the static pressure provided by the water column. Transmembrane pressure is developed by a pump that develops suction on the permeate side of the membranes; thus submerged systems are sometimes called suction- or vacuum-based systems. Net positive suction head (NPSH) limitations on the permeate pump restrict submerged membranes to a maximum transmembrane pressure of about 0.5 bar (7.4 psi), and they typically operate at a transmembrane pressure of 0.2 to 0.4 bar (3 to 6 psi). Submerged systems are configured with multiple basins so that individual basins can be isolated for cleaning or maintenance without shutting down the entire plant. Each basin typically has its own permeate pump.

Because clean water is extracted from the feed basin through the membranes and solids are returned directly to the feed tank during the backwash

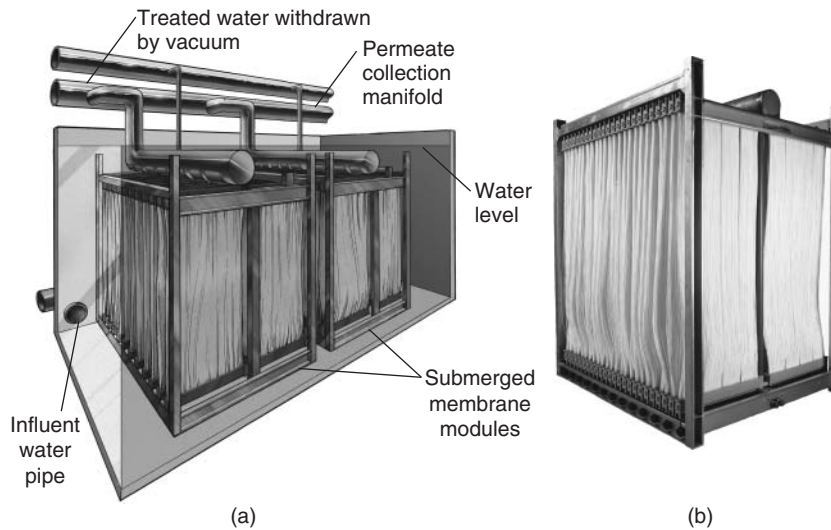


Figure 12-8
Submerged configurations for membrane filtration: (a) schematic of a submerged membrane module and (b) photograph of a single module. (© 2011 General Electric Company. All rights reserved. Reprinted with permission.)

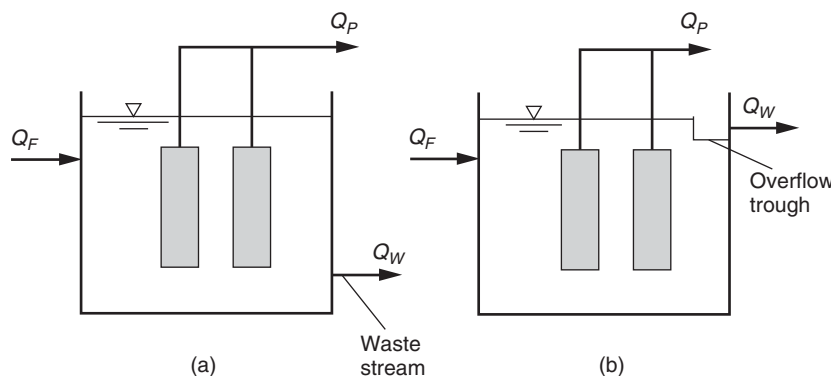


Figure 12-9
Feed-and-bleed and semibatch modes of operation. In feed-and-bleed, Q_P and Q_W are both continuous, the sum of the two flows equals Q_F . In semibatch, Q_P is continuous and equal to Q_F , Q_W only flows when solids are being wasted.

cycle, the solids concentration in the feed tank can be significantly higher than in the raw water. A high solids concentration can be advantageous when using treatment additives (i.e., coagulants or PAC) to remove dissolved contaminants but can have an adverse impact on the solids loading on the membrane during filtration. Two basic strategies are used to maintain the proper solids concentration in the feed tank, as shown on Fig. 12-9: (1) the feed-and-bleed strategy and (2) the semibatch strategy. In the feed-and-bleed strategy, a small waste stream is continuously drawn from the feed tank. The average solids concentration in the tank will be a function of the size of the waste stream:

$$C_w = \left(\frac{Q_f}{Q_w} \right) C_f \quad (12-1)$$

where C_f = solids concentration in influent, mg/L
 C_w = solids concentration in tank and waste stream, mg/L
 Q_f = influent flow rate, m³/h
 Q_w = waste flow rate, m³/h

Some guide books, such as the *Membrane Filtration Guidance Manual* (U.S. EPA, 2005), refer to the ratio C_w/C_f , and therefore the ratio Q_f/Q_w , as the volume concentration factor (VCF).

The semibatch strategy operates without a continuous waste stream, and the feed and permeate flows are at the same rate. As a result, solids accumulate in the feed tank during the filtration cycle. During the backwash cycle, the volume of water in the tank increases due to addition of the backwash flow (raw water continues to flow to the tank during the backwash cycle), and the excess water (and solids) exits the basin through an overflow trough or port.

In currently available equipment, submerged systems tend to accommodate larger modules than pressure-vessel systems. Furthermore, submerged systems have substantially fewer valves and piping connections. As larger membrane plants are designed and built, membrane manufacturers have tried to improve the economy of scale by developing larger modules to reduce the number of individual modules and piping connections necessary in large facilities, and these trends are expected to continue to lead to the development of larger modules.

Flow Direction through Hollow Fibers

Filtration occurs as water passes through the wall of the hollow fiber. Some manufacturers have designed membrane systems to filter from outside to inside (the feed water is against the shell, or outside the fiber, and the permeate is in the lumen, or inside the fiber), and other manufacturers have designed systems to filter in the opposite direction (inside out). The advantages and disadvantages of each flow configuration are described in Table 12-5. Pressure vessels use either outside-in or inside-out membranes, while submerged systems use only outside-in membranes. The difference in flow that can be achieved with outside-in versus inside-out systems is demonstrated in Example 12-1.

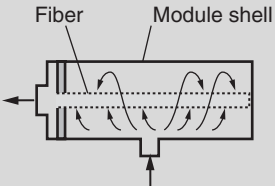
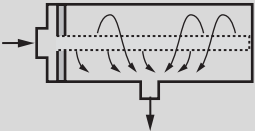
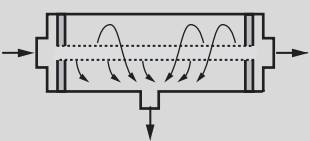
Cross-Flow and Dead-End Flow Regimes

Permeate flux and fouling are affected by the flow regime of the feed water near the membrane surface. Two filtration strategies, cross-flow filtration and dead-end filtration, have been developed to influence this flow regime and are shown schematically on Fig. 12-10.

CROSS-FLOW FILTRATION

In cross-flow filtration, the feed water is pumped at a high rate through the lumen of inside-out membrane fibers. The cross-flow velocity, typically 0.5

Table 12-5
Comparison of hollow-fiber membrane configurations

Configuration	Advantages	Disadvantages
<p>Outside in</p> 	<ul style="list-style-type: none"> <input type="checkbox"/> Can treat more water at same flux because outside of fiber has more surface area <input type="checkbox"/> Less sensitive to presence of large solids in the feed water 	<ul style="list-style-type: none"> <input type="checkbox"/> Cannot be operated in cross-flow mode
<p>Inside out (dead-end mode)</p> 	<ul style="list-style-type: none"> <input type="checkbox"/> Less expensive to operate than inside out in cross-flow mode 	<ul style="list-style-type: none"> <input type="checkbox"/> Large solids in feed water can clog lumen <input type="checkbox"/> Can treat less water at same flux because inside of fiber has less surface area
<p>Inside out (cross-flow mode)</p> 	<ul style="list-style-type: none"> <input type="checkbox"/> Can be operated at higher flux with high-turbidity feed water because cross-flow velocity flushes away solids and reduces impact of particles forming cake at membrane surface 	<ul style="list-style-type: none"> <input type="checkbox"/> Large solids in feed water can clog lumen <input type="checkbox"/> Can treat less water at same flux because inside of fiber has less surface area <input type="checkbox"/> Pumping costs associated with recirculating feed water through lumen can be expensive

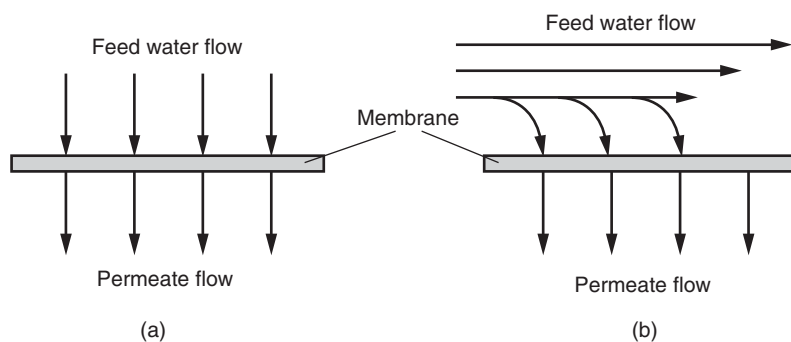


Figure 12-10
Flow regimes in membranes:
(a) dead-end filtration and
(b) cross-flow filtration.

Example 12-1 Comparison of outside-in and inside-out filtration

A Dow Filmtec SFX-2860 membrane module contains 5760 fibers. The fibers are 1.87 m long with an outside diameter of 1.3 mm and inside diameter of 0.7 mm. Calculate the water production from one module if the flux is $75 \text{ L/m}^2 \cdot \text{h}$ and the flow direction is (1) outside in and (2) inside out. Compare the two answers.

Solution

1. Compute the product water flow for outside-in flow.

- a. Determine the outside surface area per fiber:

$$\begin{aligned} a(\text{per fiber}) &= \pi dL = \pi(1.3 \text{ mm})(1.87 \text{ m})(10^{-3} \text{ m/mm}) \\ &= 7.64 \times 10^{-3} \text{ m}^2/\text{fiber} \end{aligned}$$

- b. Compute the product water flow:

$$\begin{aligned} Q &= Ja = (75 \text{ L/m}^2 \cdot \text{h})(7.64 \times 10^{-3} \text{ m}^2/\text{fiber})(5760 \text{ fibers}) \\ &= 3300 \text{ L/h} \end{aligned}$$

2. Compute the product water flow for inside-out flow.

- a. Determine the inside surface area per fiber:

$$\begin{aligned} a(\text{per fiber}) &= \pi dL = \pi(0.7 \text{ mm})(1.87 \text{ m})(10^{-3} \text{ m/mm}) \\ &= 4.11 \times 10^{-3} \text{ m}^2/\text{fiber} \end{aligned}$$

- b. Compute the product water flow:

$$\begin{aligned} Q &= Ja = (75 \text{ L/m}^2 \cdot \text{h})(4.11 \times 10^{-3} \text{ m}^2/\text{fiber})(5760 \text{ fibers}) \\ &= 1780 \text{ L/h} \end{aligned}$$

3. Compare the outside-in and inside-out flow configurations:

$$\text{Ratio} = (3300/1780) \times 100\% = 186\%$$

Comment

Operating at the same flux, the outside-in system produces nearly double the product water flow (86 percent more) as the inside-out system. Based on the results presented in this example, membrane systems cannot be compared or specified on the basis of flux if the flow configuration is different (the total flow per module and cost per module would be more important indicators than flux).

to 1 m/s (1.6 to 3.3 ft/s), is parallel to the membrane surface and about four orders of magnitude greater than the superficial velocity of water toward the membrane surface. The velocity parallel to the membrane surface creates a shear force that reduces the development of a surface cake (Wiesner and Chellam, 1992). Because many solids are carried away with the retentate instead of accumulating on the membrane surface, the system can be operated at a higher flux or with longer intervals between backwashes. The retentate is recirculated to the feed water, so cross-flow filtration requires a substantial recirculation flow—the permeate flow is typically less than 25 percent of the feed flow. The recirculation requirements can be prohibitive in a large facility—a 50,000-m³/d (13.2-mgd) membrane filtration plant must recirculate 200,000 to 250,000 m³/d (53 to 66 mgd) to maintain sufficient cross-flow velocity.

The retentate can be returned directly to the feed line to the membrane modules or to a mixing basin upstream of the modules. In either case, the solids content of the feed water will increase due to the recirculation. Either the feed-and-bleed or the semibatch procedure can be used to control the solids content in the recycle line.

DEAD-END FILTRATION

Dead-end filtration operates without a cross-flow component to the feed stream. The bulk feed water flow is transverse (perpendicular) to and toward the membrane surface during dead-end filtration, so all solids accumulate on the membrane during the filtration cycle and are removed during the backwash cycle. The greater solids accumulation during the filter run may result in lower average flux values than those achieved with cross-flow filtration.

PRACTICAL CONSIDERATIONS IN WATER TREATMENT

The dead-end flow regime is most common in membrane filtration for water treatment, in contrast to many industrial applications of microfiltration and ultrafiltration. Many industrial feed streams have high solids concentrations (e.g., the solids concentration in many food-processing operations can be 1 to 30 percent), and cross-flow operation is critical for achieving reasonable flux and filter run length. Surface waters are fairly dilute (many membrane plants operate with feed water turbidity of 100 NTU or less, which corresponds to a solids concentration on the order of 0.01 percent) so the advantages of cross-flow filtration are less significant. In addition, the piping and pumping costs of recirculating a large fraction of the feed water become prohibitive as the facility size gets larger, and water treatment facilities are built with considerably higher capacity than most industrial applications. The electrical costs of cross-flow pumping can triple the operating costs (Glucina et al., 1998) over dead-end operation. Some cross-flow systems are designed to operate in a dead-end mode by

closing a valve in the retentate line when raw-water quality conditions permit (turbidity is low) and switch to a cross-flow mode only when necessary to maintain flux.

Comparison to Rapid Granular Filtration

A comparison between membrane filtration and rapid granular filtration has been presented in Table 12-4. Membrane filtration has several advantages over granular filtration. Effective rapid filtration with granular media, as noted in Chap. 11, depends on properly destabilizing particles with a coagulant to facilitate the attachment process. The void spaces in a membrane filter are much smaller; particles are literally strained from the water so destabilization is not necessary. As a result, membrane filtration plants do not require coagulation, flocculation, and sedimentation facilities for effective particle removal. These differences can reduce requirements for chemical storage and handling and residual-handling facilities and allow membrane plants to be more compact and automated. Furthermore, the more compact installation can result in considerable cost savings in densely populated areas or other areas where land costs are high.

The most significant advantage, however, is that the filtered water turbidity from membrane filters is independent of the concentration of particulate matter in the feed. The effluent from rapid granular filters is not independent of influent quality. Changes in raw-water chemistry without changes in pretreatment (i.e., adjustment of the coagulant dose) can cause the rapid granular filtration process to fail. Rapid granular filtration is sensitive to fluctuations in raw-water quality and the experience of the plant operators.

12-4 Properties of Membrane Materials

An understanding of the mechanisms that control membrane filtration begins with an understanding of the filtration medium. Important material properties, membrane chemistry, and physical structure are discussed in this section. Although MF and UF membranes are used for similar purposes in water treatment, some of the properties are different.

Material Properties

Membrane performance is affected strongly by the physical and chemical properties of the material. The ideal membrane material is one that can produce a high flux without clogging or fouling and is physically durable, chemically stable, nonbiodegradable, chemically resistant, and inexpensive. Important characteristics of membrane materials, methods of determination, and effects on membrane performance are described in Table 12-6.

One of the important characteristics in Table 12-6 with respect to membrane fouling is hydrophobicity. Hydrophilic materials, which like contact

Table 12-6Important properties of membrane materials^a

Property	Method of Determination	Impact on Membrane Performance
Retention rating (pore size or molecular weight cut-off)	Bubble point, challenge tests	Controls the size of material retained by the membrane, making it one of the most significant parameters in membrane filtration. Also affects head loss.
Hydrophobicity	Contact angle	Reflects the interfacial tension between water and the membrane material. Hydrophobic materials “dislike” water; thus, constituents from the water accumulate at the liquid–solid interface to minimize the interfacial tension between the water and membrane. In general, hydrophobic materials will be more susceptible to fouling than hydrophilic materials.
Surface or pore charge	Streaming potential	Reflects the electrostatic charge at the membrane surface. Repulsive forces between negatively charged species in solution and negatively charged membrane surfaces can reduce fouling by minimizing contact between the membrane and fouling species. In UF, electrostatic repulsion can reduce the passage of like-charged solutes. Membranes fabricated of uncharged polymers typically acquire some negative charge while in operation.
Surface roughness	Atomic force microscopy	Affects membrane fouling; some studies have shown rough materials will foul more than smooth materials.
Porosity (surface and bulk)	Thickness/weight measurements	Affects the head loss through the membrane; higher porosity results in lower head loss.
Thickness	Thickness gauge, electron microscopy	Affects the head loss through the membrane; thinner membranes have lower head loss.
Surface chemistry	ATR/FTIR, SIMS, XPS	Affects fouling and cleaning by influencing chemical interactions between the membrane surfaces and constituents in the feed water.
Chemical and thermal stability	Exposure to chemicals and temperature extremes	Affects the longevity of the membrane; greater chemical and temperature tolerance allows more aggressive cleaning regimes with less degradation of the material.
Biological stability	Exposure to organisms	Affects the longevity of the membrane; low biological stability can result in the colonization and physical degradation of the membrane material by microorganisms.

(continues)

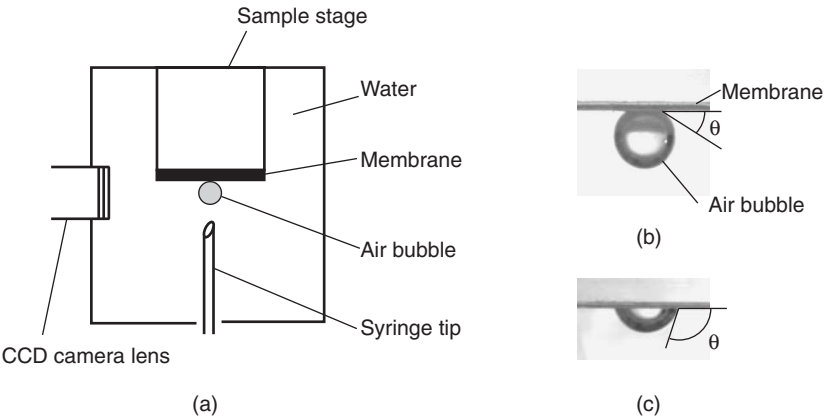
Table 12-6 (Continued)

Property	Method of Determination	Impact on Membrane Performance
Chlorine/oxidant tolerance	Exposure to chlorine/oxidants	Affects the ability to disinfect the membrane equipment. Routine disinfection prevents microbial growth on the permeate side of membrane surfaces and prevents biological degradation of membrane materials (increasing the longevity of the membrane).
Mechanical durability	Mechanical tests	Affects the ability of the material to withstand surges due to operation of valves and pumps.
Internal physical structure, tortuosity	Electron microscopy	Affects the hydrodynamics of flow and particle capture. There are no standard procedures for quantifying the tortuosity or internal structure of membranes.
Cost	Material cost	Affects the cost of the membrane system.

^aAbbreviations: ATR/FTIR = attenuated total reflectance/Fourier transform infrared spectrometry, SIMS = secondary ion mass spectrometry, XPS = X-ray photoelectron spectrometry.

with water, tend to have low fouling tendencies, whereas hydrophobic materials may foul extensively. Hydrophobicity is quantified by contact angle measurements, in which a droplet of water or bubble of air is placed against a membrane surface, and the angle between the surface and water or air is measured. Hydrophobic surfaces have a high contact angle (the water beads like on a freshly waxed car), whereas hydrophilic surfaces have a low contact angle (the water droplets spread out). Techniques for measuring contact angle are demonstrated on Fig. 12-11. Contact angle measurements vary widely because of differences in measurement techniques and variables such as surface roughness but typically range from about 40° to 50° for cellulose acetate to about 110° for polypropylene (Cheryan, 1998).

Figure 12-11
Captive bubble contact angle measurements for determination of hydrophobicity: (a) contact angle measurement apparatus, (b) hydrophilic surface (low contact angle), and (c) hydrophobic surface (high contact angle).



Hydrophobicity is affected strongly by the chemical composition of the polymer comprising the material. Polymers that have ionized functional groups, polar groups (water is very polar), or oxygen-containing and hydroxyl groups (for hydrogen bonding) tend to be very hydrophilic. Unfortunately, chemical properties that improve hydrophilicity tend to reduce the chemical, mechanical, and thermal stability because water molecules act as plasticizers for hydrophilic materials (Anselme and Jacobs, 1996).

Lacking the existence of a perfect material, a variety of materials have been used. The two most common materials in early commercial membrane filtration systems were cellulose acetate (CA) and polypropylene (PP), but their use has been declining. The most common polymeric materials currently used in water treatment are polyvinylidene fluoride (PVDF), polysulfone (PS), and polyethersulfone (PES). Ceramic membrane may also be gaining in popularity. Some membrane manufacturers consider the composition of their membranes to be proprietary and do not release information on their material chemistry. The chemical structure of these polymers is shown on Fig. 12-12 and important properties are given in Table 12-7.

Material Chemistry

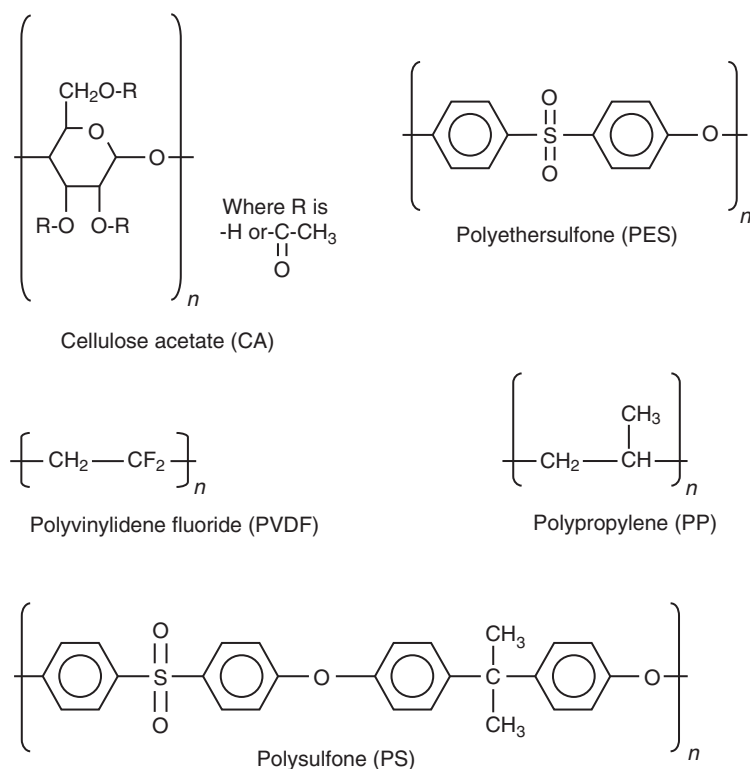


Figure 12-12
Chemical structure of common polymeric MF and UF membrane materials.

Table 12-7

Characteristics of common membrane materials

Membrane Material	Characteristics
Cellulose acetate (CA)	CA is the most hydrophilic of common industrial-grade membrane materials, which helps to minimize fouling and maintain high flux values. The material is easy to manufacture, inexpensive, and available in a wide range of pore sizes. Has been losing favor for membrane filters because of higher susceptibility to biological degradation, lack of tolerance to continuous exposure or high concentrations of free chlorine, gradual decline in the flux over its lifetime due to compaction, and lack of tolerance to aggressive cleaning chemicals or temperatures above 30°C.
Polysulfone (PS)/polyethersulfone (PES)	PS and PES are moderately hydrophobic and have excellent chemical tolerance and biological resistance. They can withstand free chlorine contact to 200 mg/L for short periods of time for cleaning, pH values between 1 and 13, and temperatures to 75°C. Aggressive cleaning and disinfecting is possible.
Polyvinylidene fluoride (PVDF)	PVDF is moderately hydrophobic and has excellent durability, chemical tolerance, and biological resistance. It can withstand continuous free chlorine contact to any concentration, pH values between 2 and 10, and temperatures to 75°C. Aggressive cleaning and disinfecting is possible.
Polypropylene (PP)	PP is the most hydrophobic of common industrial-grade membrane materials. Only MF membranes are available in PP; the material is too hydrophobic to allow water to pass through the small pore spaces in UF membranes. It is durable, chemically and biologically resistant, and tolerant of moderately high temperatures and pH values between 1 and 13, which allows aggressive cleaning. It has been losing favor for membrane filters because it is not tolerant to chlorine, which hinders the ability to control biological growth.
Ceramic	Ceramic membranes are configured as rigid monolithic elements. The material is hydrophilic, rough, and can withstand high operating pressure and temperature. They have excellent chemical and pH tolerance. Aggressive cleaning and disinfecting is possible.

Membrane Structure

The structure, porosity, and transport properties of most MF membranes are relatively constant throughout their depth (this structure is called homogeneous). Theoretically, homogeneous membranes perform identically regardless of which direction filtration is proceeding. In contrast, UF membranes have an asymmetric (also called anisotropic or “skinned”) structure, which means that the morphology varies significantly across the depth of the membrane. A homogeneous membrane was shown on Fig. 12-4a, and the structure of an asymmetric membrane, consisting of an active layer and a support layer, is shown on Fig. 12-13. The active and support layers have separate functions.

Filtration occurs at the active layer in asymmetric membranes, which is a thin skin with low porosity and very small void spaces. The low porosity and small pores generate significant resistance to flow, which must be minimized by making the active layer as thin as possible. The active layer is so thin that it has no mechanical durability. Thus, the remainder of the membrane is a highly porous layer that provides support but produces very little hydraulic resistance. The support layer accounts for the majority of the membrane thickness. Asymmetric membranes allow the filtration and mechanical properties to be designed separately. Filtration through an asymmetric membrane is not the same in both directions. Filtration in the “wrong” direction would cause the voids in the support layer to become clogged and may cause the active layer to separate from the rest of the membrane. To prevent clogging, some commercial asymmetric membranes have active layers on both surfaces of the membrane with a support layer sandwiched between the two active layers.

Microfiltration and UF membranes have different porosities. The porosity of MF membranes varies widely, and values ranging from 30 to 90 percent have been reported. Theoretically, the porosity of homogeneous membranes should be constant throughout the depth of the membrane.

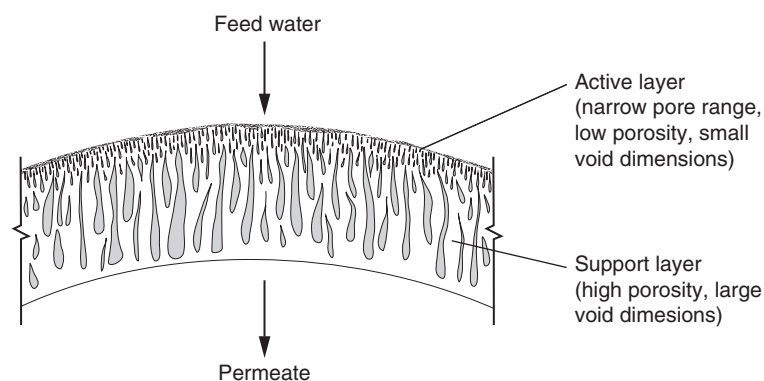


Figure 12-13
Structure of an asymmetric UF membrane.

The porosity of the active layer of UF membranes is low and ranges from 0.5 to 10 percent, whereas the porosity in the support structure is considerably higher (50 to 90 percent).

12-5 Particle Capture in Membrane Filtration

For regulatory purposes in the United States, membrane filtration is defined as “a pressure or vacuum driven separation process in which particulate matter larger than 1 μm is rejected by an engineered barrier primarily through a size exclusion mechanism and which has a measurable removal efficiency of a target organism that can be verified through the application of a direct integrity test” (U.S. EPA, 2006, p. 702). The principles by which membranes are rated, particles are captured, and performance is demonstrated is discussed in this section.

Retention Rating

One of the most significant parameters in membrane filtration is the size of material retained. Microfiltration and UF membranes are currently rated with different systems, making them difficult to compare. The retention rating of MF membranes is based on the diameter of material that is retained by the membrane; for example, a 0.2- μm MF membrane should hypothetically retain 100 percent of 0.2- μm -diameter particles. The MF retention rating is frequently called the nominal pore diameter or pore size throughout the membrane industry, but those terms are misnomers. As was shown on Fig. 12-3, the “pores” in MF membranes are tortuous voids with a wide size distribution, not cylindrical holes of a particular diameter. It is generally accepted that the average void space dimension is somewhat larger than the membrane retention rating (Cheryan, 1998). The pore size value is a nominal rating, so some particles smaller than the pore size can be retained and some particles larger may be able to penetrate the membrane. The retention rating for MF membranes used in water treatment is typically between 0.1 and 1 μm . Membranes with retention ratings in this range will completely retain bacteria and protozoan structures such as *Giardia lamblia* cysts and *Cryptosporidium parvum* oocysts. Because of their small size, viruses may not be completely retained by MF membranes.

Membrane manufacturers use two approaches for defining the retention rating of UF membranes. Some manufacturers use a pore size rating similar to MF membranes, with pore sizes of 0.01 to 0.04 μm being common. For others, the retention rating for UF membranes is based on the molecular weight of material retained by the membrane and is called the molecular weight cutoff (MWCO) or nominal molecular weight limit (NMWL). The first applications of UF membranes were for fractionating macromolecules, so the original focus for classifying UF membranes was on molecular weight rather than size. Membrane filtration for water treatment, however, is principally concerned with retaining materials of a particular

size, such as viruses, which can be as small as 0.025 μm . Unfortunately, the diameter of solids retained by a UF membrane is only loosely related to the MWCO value and depends on various physical and chemical properties (shape, electrostatic charge, etc.) of the solid. The standard procedure for determining the MWCO value of a UF membrane involves filtration of dextran solutions with varying average molecular weights (ASTM, 2001c). Dextran is a branched polysaccharide that might be expected to have substantially different physical and chemical properties from viruses or other particles. In addition, the MWCO of UF membranes is based on the molecular weight at 90 percent rejection. The difference between the rating of MF and UF membranes is shown on Fig. 12-14.

The hydrodynamic diameter of molecules can be roughly estimated from the molecular weight. For instance, data in Ioan et al. (2000) suggest the following empirical relationship between the hydrodynamic diameter and molecular weight of dextran:

$$d_H = 0.11 (\text{MW})^{0.46} \quad (12-2)$$

where d_H = hydrodynamic diameter of dextran molecule, nm
MW = molecular weight, g/mol

Researchers have attempted to use relationships such as Eq. 12-2 to estimate the pore size of UF membranes. In addition, analytical techniques such as electron microscopy, atomic force microscopy, thermoporometry, and biliquid permporometry have been used to estimate the pore size of UF membranes (Kim et al., 1994).

The MWCO for UF membranes range from about 1000 daltons (Da) to about 500,000 Da. These MWCO values correspond to an ability to retain particles ranging from about 1 to 30 nm in diameter (Cheryan, 1998). Comparing these values to the size of viruses, it is clear that some UF membranes can completely retain viruses but others may not, depending

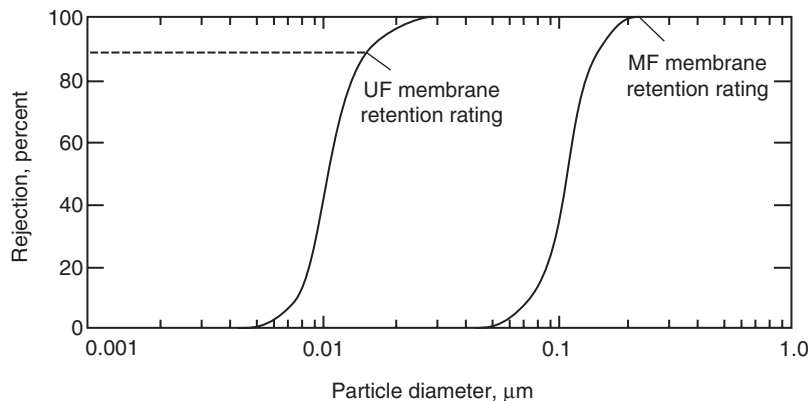


Figure 12-14
Determination of retention ratings for MF and UF membranes.

on the MWCO. Studies have shown, for instance, greater than 7-log removal of MS2 bacteriophage (a model virus) with a 100,000-Da UF membrane but less than 1-log removal with a 500,000-Da UF membrane (Jacangelo et al., 1995).

It should be noted that there is overlap between the size of pores of MF and UF membranes, and there are no standard specifications that classifies a particular product as one or the other. Often, the classification of a particular product as an MF or UF membrane depends on the marketing strategy of the manufacturer and whether the retention rating was measured as a pore size or MWCO. Furthermore, classification as an MF or UF membrane does not guarantee a particular level of removal efficiency for specific pathogen organisms. For that, challenge testing is required, as described later in this section.

Rejection and Log Removal

The fraction of material removed from the permeate stream is called rejection:

$$R = 1 - \frac{C_p}{C_f} \quad (12-3)$$

where R = rejection, dimensionless

C_p = permeate concentration, mol/L or mg/L

C_f = feed water concentration, mol/L or mg/L

Rejection can be calculated for bulk measures of particulate matter (e.g., turbidity, particle counts) or individual components of interest (e.g., *Cryptosporidium* oocysts). In membrane filtration, the concentration of some components in the permeate can be several orders of magnitude lower than in the feed. Many significant figures must be retained to quantify rejection if Eq. 12-3 is used. In these cases, the log removal value (LRV) (see Sec. 4-5) is used:

$$\text{LRV} = \log(C_f) - \log(C_p) = \log\left(\frac{C_f}{C_p}\right) \quad (12-4)$$

A comparison of the calculation of rejection and log removal value is demonstrated in Example 12-2.

Filtration Mechanisms

The primary mechanism for removing particles from solution in membrane filtration is straining, but removal is also affected by adsorption and cake formation. These removal mechanisms are depicted on Fig. 12-15.

STRAINING

Straining (also called sieving or steric exclusion) is the dominant filtration mechanism in membrane filtration. Nominally, particles larger than the retention rating of the membrane collect at the surface while water and

Example 12-2 Calculation of rejection and log removal value

During testing of a prototype membrane filter, bacteriophage concentrations of 10^7 mL^{-1} and 13 mL^{-1} were measured in the influent and effluent, respectively. Calculate the rejection and log removal value.

Solution

1. Calculate rejection using Eq. 12-3:

$$R = 1 - \frac{C_p}{C_f} = 1 - \frac{13 \text{ mL}^{-1}}{10^7 \text{ mL}^{-1}} = 0.9999987$$

2. Calculate log removal value using Eq. 12-4:

$$\text{LRV} = \log \left(\frac{C_f}{C_p} \right) = \log \left(\frac{10^7 \text{ mL}^{-1}}{13 \text{ mL}^{-1}} \right) = 5.89$$

Comment

Note that seven significant digits are necessary to express rejection adequately in arithmetic units, but only three significant digits are necessary to express log removal value for this example. Also note that $\text{LRV} = 5$ corresponds to 99.999 percent and $\text{LRV} = 6$ corresponds to 99.9999 percent rejection (i.e., the log removal value equals the “number of 9’s”).

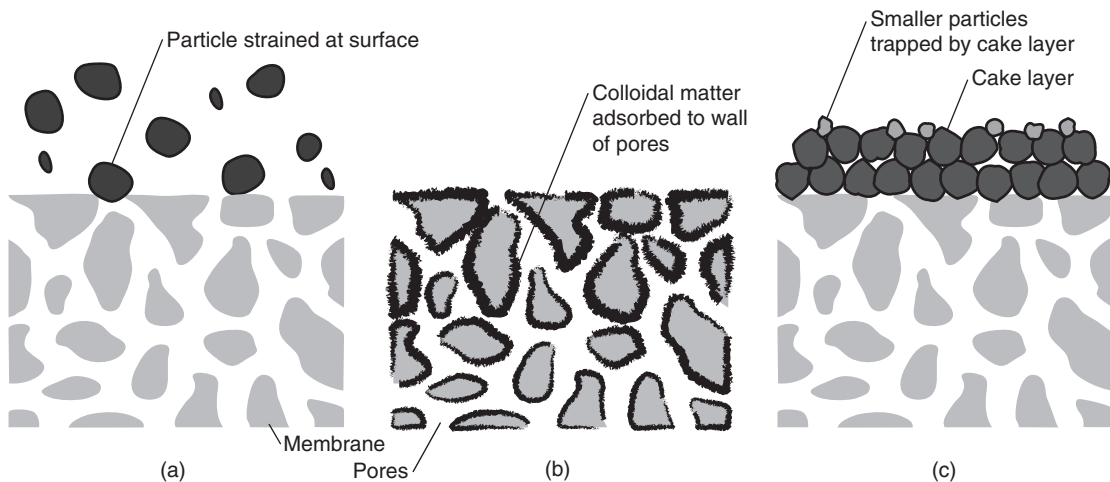


Figure 12-15

Mechanisms for rejection in membrane filtration. (a) Straining occurs when particles are physically retained because they are larger than the pores. (b) Adsorption occurs when material small enough to enter pores adsorbs to the walls of the pores. (c) Cake filtration occurs when particles that are small enough to pass through the membrane are retained by a cake of larger material that collects at the membrane surface.

smaller particles pass through. This view, however, suggests that the relationship between particle size and retention rating is a step function, with $R = 100\%$ for all particles larger than the retention rating and $R = 0\%$ for all smaller particles. As shown on Fig. 12-14, particle removal is not a simple step function at the retention rating. This nonideal performance occurs when the particle size is close to the membrane retention rating and is caused by the variability of pore size dimensions, nonspherical shape of the particles, and other interactions such as electrostatic repulsion.

As is evident from Fig. 12-3c, the tortuous interconnecting voids in membrane filters have a distribution of sizes, including some larger than the retention rating. Thus, particles smaller than the retention rating may be trapped in smaller passageways, and larger particles may pass through the membrane in other areas. The existence of larger void spaces is particularly important when high rejection values are required.

A second source of nonideal rejection arises from particle dimensions. Particles in natural systems can have shape characteristics significantly different from the materials used to determine the retention rating. Rod-shaped bacteria and linear macromolecules may be very long in one dimension and considerably smaller in others and may not be adequately described by an average diameter. Rod-shaped bacteria near the retention rating of the membrane may or may not be rejected depending on orientation when they approach the membrane. Some small particles, particularly large proteins or other macromolecules, may be flexible, assuming a spherical shape when in solution but becoming more linear when forced through a membrane under pressure. Thus, particles that appear to be slightly larger than the retention rating may pass through the membrane.

Interactions between particles and the membrane can affect rejection when the particle size is close to the membrane retention rating. Typically, both particles and membrane surfaces are negatively charged. Electrostatic interactions may prevent the particles from entering the pores even if the physical size would permit passage. Ferry (1936) reported nonideal behavior even under near-ideal filtration conditions (filtration of a monodisperse dispersion through a track-etched membrane with uniform cylindrical pores).

ADSORPTION

Natural organic matter adsorbs to membrane surfaces (Jucker and Clark, 1994). Thus, soluble materials may be rejected even though their physical dimensions are orders of magnitude smaller than the membrane retention rating. Adsorption may be an important rejection mechanism during the early stages of filtration with a clean membrane. The adsorption capacity is quickly exhausted, however, and adsorption is not an effective mechanism in long-term operation of membrane filters. Nevertheless, adsorption has a profound impact on membrane operations. Adsorbed material can reduce the size of voids throughout the membrane, increasing the ability of the

membrane to retain smaller material by straining. In addition, adsorption has been implicated as a prime cause of membrane fouling by NOM.

CAKE FORMATION

During filtration, a clean membrane will quickly accumulate a cake of solids at the surface due to straining. This surface cake acts as a filtration medium, providing another mechanism for rejection. The surface cake is often called a “dynamic” membrane, because its filtering capability varies with time, growing in thickness during filtration but being partially or wholly removed during backwashing. Mathematical modeling of cake filtration is similar to granular filtration, and equations such as the Kozeny equation (see Chap. 11) are used to calculate the additional resistance to flow resulting from the formation of surface cakes.

The principal microorganisms of concern in water treatment are (1) protozoa and helminths, (2) bacteria, and (3) viruses. The removal of each of these is considered below.

Removal of Microorganisms

REMOVAL OF PROTOZOA AND HELMINTHS

Giardia lamblia cysts and *C. parvum* oocysts, which are highly resistant to chemical disinfectants, are at least 10 times larger than the retention ratings of MF and UF membranes. Rejection of greater than 7 log has been observed for both MF and UF membranes (Jacangelo et al., 1991, 1995), as would be expected due to straining. Indeed, reported rejection values are limited only by the influent concentration of organisms. For instance, if 10^6 cysts/L are measured in the feed and the detection limit in the permeate is 1 cyst/L, the calculated log removal value is $LRV \geq 6$.

REMOVAL OF BACTERIA

Bacteria range in size from 0.1 to 100 μm (Madigan et al., 1997). Being considerably larger than the retention rating, complete rejection via straining is expected for UF membranes. In addition, most species of bacteria should be rejected completely by MF membranes through straining. In many studies, bacteria are removed to below the detection limit; in some cases, rejection was higher than 8 log (U.S. EPA, 2001). In other studies, reported rejection was lower because of contamination and regrowth interferences. Some bacterial species are near the retention ratings of MF membranes. In these cases, lower rejection may be possible due to the factors mentioned earlier.

REMOVAL OF VIRUSES

The smallest viruses have a diameter of about 25 nm. At this size, viruses are considerably smaller than the retention rating of MF membranes and are similar to that of UF membranes. For MF membranes, straining, adsorption,

and cake filtration all contribute to rejection, and virus rejection can vary from $LRV < 1$ to $LRV > 4$. Madaeni et al. (1995) studied poliovirus rejection by 0.2- μm MF membranes and found complete rejection ($LRV \geq 4.5$) during the first 15 min of filtration, but rejection quickly dropped to $LRV = 1.31$ after 30 min of filtration and gradually rose to $LRV = 1.71$ during the next 5 h. Adsorption appears to be significant in initial rejection, but the adsorption capacity was quickly exhausted. After 30 min, rejection may have been due to straining through small void spaces or continued adsorption. The gradual increase in rejection over time suggests that straining was increasing as the pores became more restricted with previously deposited virus particles. Jacangelo et al. (1995) also observed a gradual increase in virus rejection that correlated with a decrease in specific flux over a period of 45 days. Fouling caused a reduction in the pore diameter or formation of a surface cake that was able to increase virus rejection. Cake filtration also plays an important role in virus rejection by MF membranes. Preloading an MF membrane with kaolinite clay (Jacangelo et al., 1995) and increasing the turbidity of the feed water (Madaeni et al., 1995) result in higher rejection of viruses. Increased rejection of viruses with higher turbidity may also be due to attachment of the viruses to the particle prior to filtration.

Ultrafiltration membranes with low MWCO ratings may be able to achieve complete rejection of viruses, but UF membranes with higher MWCO ratings might not. It was noted earlier that the pore size of UF membranes may range from 1 to 30 nm depending on the MWCO. Jacangelo et al. (1995) found complete rejection ($LRV > 7.2$) of MS2 bacteriophage, a model virus with a diameter of about 25 nm, with a 100,000-Da UF membrane but $LRV < 1$ with a 500,000-Da UF membrane.

The overall implication for water treatment is that straining is only effective for particles significantly larger than the retention rating of the membrane. Cake filtration and adsorption may provide added rejection of smaller material, but are not considered effective removal mechanisms from a regulatory point of view. Thus, MF membranes should not be relied upon for complete removal of viruses (although some removal credit may be warranted), and poor selection of UF membranes may also provide insufficient removal for viruses. To validate the ability of MF and UF membranes to remove specific microorganisms, challenge testing is performed.

Challenge Testing

Challenge testing is a process in which the ability of a membrane product to remove specific target organisms is determined in carefully controlled tests. Specific requirements in the Long Term 2 Enhanced Surface Water Treatment Rule (LT2ESWTR) (U.S. EPA, 2005) are focused on challenge testing for *Cryptosporidium* removal efficiency, but the general methods could be used for other microbial contaminants such as viruses, bacteria, or other protozoa. Challenge tests establish the maximum removal credit

allowed for a particular product. Once performance has been verified, site-specific testing by utilities is not required. The LT2ESWTR requires that the modules tested be similar in design, materials, and construction to full-scale modules, although the test modules do not necessarily need to be full size. The number of modules testing should be determined on a scientifically defensible basis to provide results that take manufacturing variability into account. Operating conditions representative of full-scale operation, such as maximum flux and recovery are necessary. The target microorganisms or particles should be measured directly and not inferred from a surrogate like turbidity. The concentration of particles in the feed water should be 6.5 log units higher than the detection limit in the permeate water to ensure that high removal efficiency can be demonstrated. Finally, nondestructive performance tests (NDPT) (such as bubble point or pressure decay tests, discussed later) should be performed at the same time, so that the NDPT can be performed on modules after manufacture and related to the results of the challenge testing.

12-6 Hydraulics of Flow through Membrane Filters

The flow of water through MF and UF membranes follows the fundamental law for flow through porous media known as Darcy's law:

$$v = k_p \frac{h_L}{L} \quad (12-5)$$

where v = superficial fluid velocity, m/s
 k_p = hydraulic permeability coefficient, m/s
 h_L = head loss across porous media, m
 L = thickness of porous media, m

The hydraulic permeability coefficient in Darcy's law is an empirical parameter that is used to describe the proportionality between head loss and fluid velocity and is dependent on media characteristics such as porosity and specific surface area. Although flow through membranes follows Darcy's law, the standard equation for membrane flow is written in a substantially different form. Flow is expressed in terms of volumetric flux J rather than superficial velocity, the driving force is expressed as transmembrane pressure ΔP rather than head loss (which are related by $\Delta P = \rho_w g h_L$), and media characteristics are expressed as a resistance coefficient (the inverse of a permeability coefficient). In addition, the membrane flow equation includes the fluid viscosity explicitly (Darcy's law buries it in the permeability coefficient) because viscosity has a significant impact on flux and is easy to determine (via temperature). Finally, the membrane flux equation

incorporates the membrane thickness into the resistance coefficient. The equation for membrane flux is

$$J = \frac{Q}{a} = \frac{\Delta P}{\mu \kappa_m} \quad (12-6)$$

where J = volumetric water flux through membrane, $\text{L}/\text{m}^2 \cdot \text{h}$ or m/s

Q = flow rate, L/h

a = membrane area, m^2

ΔP = differential pressure across membrane, bar

μ = dynamic viscosity of water, $\text{kg}/\text{m} \cdot \text{s}$

κ_m = membrane resistance coefficient, m^{-1}

The membrane resistance coefficient can be calculated from laboratory experiments so that flux through a new membrane can be determined for other pressure or temperature conditions.

The linear relationship between flux and pressure in Eq. 12-6 suggests that the flux can be maximized by operating at the highest possible transmembrane pressure. While this relationship may be true for deionized water, high-pressure operation may not be preferable during filtration of natural waters. Fouling can be exacerbated by high-pressure operation (Cheryan, 1998), so a balance must be struck between flux and fouling. Fouling will be presented in detail later in this chapter.

Ideally, it would be desirable to calculate flux from measurable parameters that describe the internal structure of MF and UF membranes, such as porosity, nominal pore diameter, specific surface area, and membrane thickness, as is done for clean-bed head loss in granular filtration. These parameters, however, are difficult to measure, and the amorphous internal structure of MF and UF membranes (refer to Fig. 12-3c) cannot be described mathematically with any great accuracy. In addition, it will be shown later in this chapter that the volumetric flux through a full-scale membrane filter is influenced more by fouling than by the intrinsic clean-membrane resistance. As a result, currently no reliable models allow flux to be calculated from easily measurable fundamental parameters. To account for membrane fouling, Eq. 12-6 can be extended by adding additional resistance terms in the denominator, as will be presented later in the chapter. Calculation of the membrane resistance coefficient is demonstrated in Example 12-3.

Temperature and Pressure Dependence

During operation, changes in permeate flux due to fouling are monitored to determine when cleaning is necessary. Because flux is dependent on pressure and water viscosity (see Eq. 12-6), determination of the extent of fouling is confounded by simultaneous changes in pressure and temperature (which changes viscosity). In temperate climates, water temperatures can vary by more than 20°C between summer and winter. Due to the

Example 12-3 Calculation of membrane resistance coefficient

An MF membrane is tested in a laboratory by filtering clean, deionized water and the flux is found to be $850 \text{ L/m}^2 \cdot \text{h}$ at 20°C and 0.9 bar . Calculate the membrane resistance coefficient.

Solution

Rearrange Eq. 12-6 to solve for the membrane resistance coefficient. The dynamic viscosity of water at 20°C , from Table C-1 in App. C, is $1.00 \times 10^{-3} \text{ kg/m} \cdot \text{s}$. Also recall that $1 \text{ bar} = 100 \text{ kPa} = 10^5 \text{ N/m}^2 = 10^5 \text{ kg/s}^2 \cdot \text{m}$.

$$\begin{aligned} \kappa_m &= \frac{\Delta P}{\mu J} = \frac{(0.9 \times 10^5 \text{ kg/s}^2 \cdot \text{m})(3600 \text{ s/h})(10^3 \text{ L/m}^3)}{(1.00 \times 10^{-3} \text{ kg/m} \cdot \text{s})(850 \text{ L/m}^2 \cdot \text{h})} \\ &= 3.81 \times 10^{11} \text{ m}^{-1} \end{aligned}$$

temperature dependence of water viscosity, the flux through a membrane can be 70 percent higher in the summer than in the winter. Temperature variations are usually accommodated by calculating the equivalent flux at a standard temperature:

$$J_s = J_m \left(\frac{\mu_m}{\mu_s} \right) \quad (12-7)$$

where J_m = flux at measured temperature, $\text{L/m}^2 \cdot \text{h}$

J_s = flux at standard temperature (typically 20°C), $\text{L/m}^2 \cdot \text{h}$

μ_m = dynamic viscosity of water at measured temperature, $\text{kg/m} \cdot \text{s}$

μ_s = dynamic viscosity of water at standard temperature, $\text{kg/m} \cdot \text{s}$

The dynamic viscosity can be obtained from tabular data or calculated from one of a variety of expressions that relate the viscosity of water to temperature. A relationship often used in membrane operations is (ASTM, 2001a)

$$J_s = J_m (1.03)^{T_s - T_m} \quad (12-8)$$

where T_m = measured temperature, $^\circ\text{C}$

T_s = standard temperature, $^\circ\text{C}$

When using a standard temperature of 20°C , Eq. 12-8 is accurate to within 5 percent over a temperature range of 1 to 28°C , which covers most natural waters. At temperatures above 28°C , Eq. 12-8 becomes increasingly inaccurate and a more rigorous expression should be used.

The factor in Eq. 12-8 accounts only for the effect of water viscosity. Temperature may also have an effect on the membrane material, such as swelling of the material at higher temperature. Some manufacturers have developed temperature correction formulas that account for changes in both the water viscosity and material properties. Manufacturer or site-specific correction equations should be used, if available.

Flux is normalized for pressure by calculating specific flux, which is the flux at a standard temperature divided by the transmembrane pressure:

$$J_{sp} = \frac{J_s}{\Delta P} \quad (12-9)$$

where J_{sp} = specific flux at standard temperature, $L/m^2 \cdot h \cdot bar$

The specific flux is called membrane permeability when clean water (reagent-grade water, typically deionized to a resistivity of $>10 M\Omega \cdot cm$ and $DOC < 0.2 mg/L$) is being filtered through a new, unused membrane. Membrane permeability is measured in laboratory experiments but rarely or never determined in full-scale installations because a large supply of deionized water is not available. Specific flux and membrane permeability are typically reported in units of $L/m^2 \cdot h \cdot bar$ or $gal/ft^2 \cdot d \cdot atm$.

When flux has been normalized to account for temperature and pressure variations, the effect of fouling can be determined, as illustrated in Example 12-4.

12-7 Membrane Fouling

Fouling is widely perceived to be the most significant issue affecting the design and operation of membrane filtration facilities (AWWA, 1992, 1998, 2005a). The results of a laboratory experiment of natural water filtration through a membrane are shown on Fig. 12-16. In this experiment, the membrane lost about half of its flow capacity in just a few hours. The individual steep trend lines represent individual 30-min filter runs. Backwash after each filter run recovers most of the lost flux as solids are flushed from the membrane surface; however, not all of the flux is recovered and a longer-term decline in performance is also evident. Fouling can be more dramatic in laboratory tests conducted at constant pressure (as this experiment was), but the overall trends shown in Fig. 12-16 are routinely observed in full-scale operating membrane filtration facilities. Fouling is characterized by the mechanism (pore blockage, pore constriction, and cake formation), by whether it can be removed (i.e., reversible or irreversible), and by the material causing it (particles, biofouling, and natural organic matter). This section examines each of these views of membrane fouling, followed by an introduction of several ways to model fouling.

Example 12-4 Calculation of specific flux

A membrane plant has a measured flux in March of $80 \text{ L/m}^2 \cdot \text{h}$ at 0.67 bar and 7°C . Four months later, in July, the measured flux is $85 \text{ L/m}^2 \cdot \text{h}$ at 0.52 bar and 19°C . Has a change in specific flux occurred? What is the change in percent? Has fouling occurred?

Solution

1. Calculate the specific flux in March.
 - a. Calculate the flux in March at a standard temperature of 20°C using Eq. 12-8:

$$\begin{aligned} J_s &= J_m(1.03)^{T_s - T_m} = (80 \text{ L/m}^2 \cdot \text{h})(1.03)^{20^\circ\text{C} - 7^\circ\text{C}} \\ &= 117 \text{ L/m}^2 \cdot \text{h} \end{aligned}$$

- b. Calculate the specific flux in March using Eq. 12-9:

$$J_{sp} = \frac{J_s}{\Delta P} = \frac{117 \text{ L/m}^2 \cdot \text{h}}{0.67 \text{ bar}} = 175 \text{ L/m}^2 \cdot \text{h} \cdot \text{bar}$$

2. Calculate the specific flux in July.
 - a. Calculate the flux in July at a standard temperature of 20°C using Eq. 12-8:

$$\begin{aligned} J_s &= J_m(1.03)^{T_s - T_m} = (85 \text{ L/m}^2 \cdot \text{h})(1.03)^{20^\circ\text{C} - 19^\circ\text{C}} \\ &= 87.6 \text{ L/m}^2 \cdot \text{h} \end{aligned}$$

- b. Calculate the specific flux in July using Eq. 12-9:

$$J_{sp} = \frac{J_s}{\Delta P} = \frac{87.6 \text{ L/m}^2 \cdot \text{h}}{0.52 \text{ bar}} = 168 \text{ L/m}^2 \cdot \text{h} \cdot \text{bar}$$

3. Calculate the percent loss of performance due to fouling:

$$\begin{aligned} &\frac{175 \text{ L/m}^2 \cdot \text{h} \cdot \text{bar} - 168 \text{ L/m}^2 \cdot \text{h} \cdot \text{bar}}{175 \text{ L/m}^2 \cdot \text{h} \cdot \text{bar}} \times 100 \\ &= 4\% \text{ loss of flux due to fouling} \end{aligned}$$

Comment

The specific flux at 20°C has declined from 175 to $168 \text{ L/m}^2 \cdot \text{h} \cdot \text{bar}$. Thus, although the plant is operating at a higher flux with a lower pressure in July than it was in March, there has been a 4 percent loss of performance due to fouling.

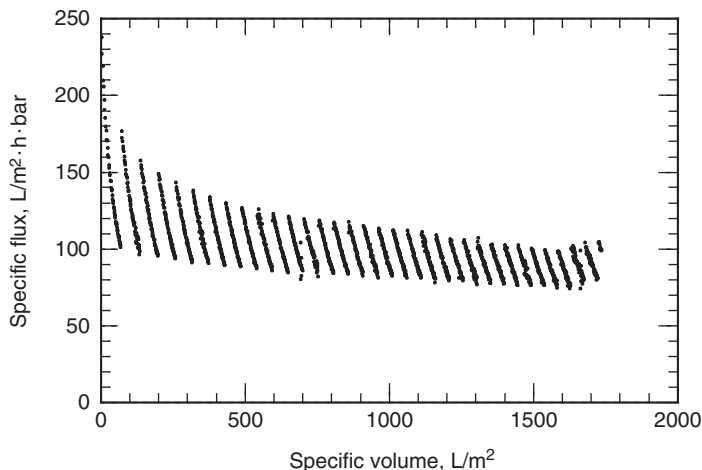


Figure 12-16
Fouling of a membrane filter during
filtration of natural water.

Mechanisms of Fouling

Membrane fouling is traditionally visualized as occurring through three mechanisms—pore blocking, pore constriction, and cake formation—as shown on Fig. 12-17. The similarity between these mechanisms and the mechanisms for particle retention on Fig 12-15 should be evident. Pore blocking occurs when the entrance to a pore is completely sealed by a particle. For this mechanism, the membrane is viewed as a plate with orifices in it, and hydraulic resistance to flow is proportional to the net area of open pores. As a portion of the pore area is blocked at the surface, flow declines by a commensurate amount. While this mechanism may

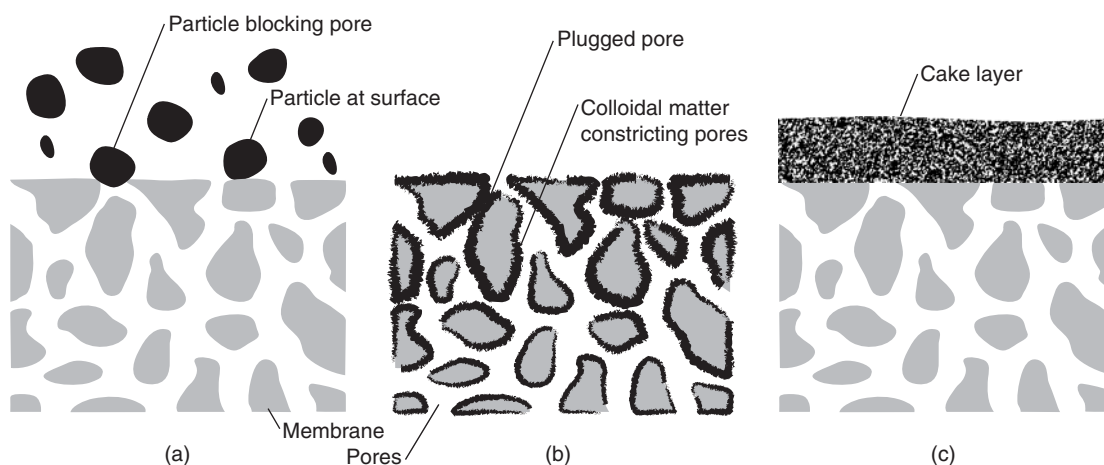


Figure 12-17
Mechanisms for fouling in membrane filtration: (a) Pore blocking, (b) pore constriction, and (c) cake layer formation.

be relevant on a track-etched membrane (see Table 12-3), it might have minimal significance in the fouling of commercial membranes for water treatment. As shown on Fig. 12-3, commercial membranes have a depth of tortuous interconnecting voids with a flow path much longer than the pore width. Since typical membrane thickness ranges from 0.1 to 0.6 mm and the pore width is 0.01 to 0.1 μm , the pores are 1000 to 10,000 times longer than they are wide. Hydraulic resistance occurs throughout this depth, and the interconnectedness of the pores would allow flow to redistribute within the membrane matrix if a portion of the pores were plugged at the surface.

Pore constriction is the reduction of the void volume within a membrane due to adsorption of materials within the pores. Several essential elements must take place for pore constriction to occur. First, the materials must be smaller than the pore size of the membrane so they can penetrate into the membrane matrix instead of being sieved at the surface. Second, they must be transported to the pore walls either by diffusion or hydrodynamic conditions. Although the residence time through the membrane is short (less than 0.5 s), the characteristic diffusion time of a colloid to a pore wall is about 4 orders of magnitude shorter, so sufficient opportunity for diffusion exists (Howe, 2001). Third, materials must have an affinity for attaching to the pore walls, without which they would pass right through the membrane. Research has demonstrated that hydrophobic membranes foul more than hydrophilic ones, and hydrophobic materials in the feed water can cause greater fouling. Concepts of particle stability presented in Chap. 9 are also relevant here. Finally, the attached material must be sufficiently large to constrict the pore dimensions. Research has shown that high-MW and colloidal organics cause more fouling than low-MW dissolved materials. Low-MW dissolved materials would not have as large of an impact on pore dimensions as colloidal materials.

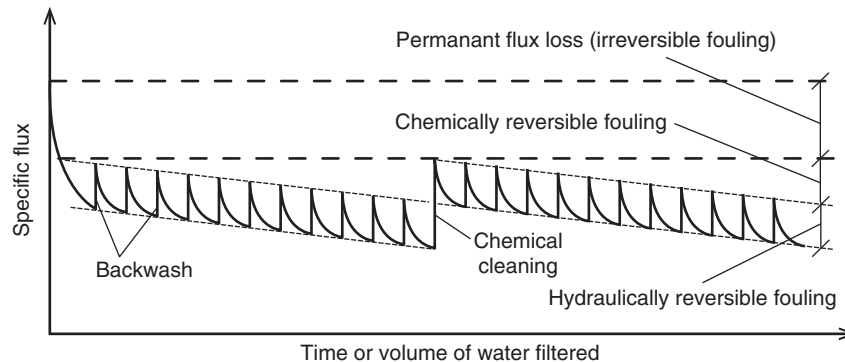
Particles that are too large to enter the pores collect on the membrane surface in a porous mat called a filter cake. Sediment larger than 0.2 mm is typically prefiltered with cartridge filters to protect the membrane, so the cake is initially composed of material between 0.2 mm and the membrane retention rating. The cake layer acts as a “dynamic” filter and can retain additional smaller material, but also generates hydraulic resistance to flow as it does so. The cake layer can prevent particles smaller than the retention rating from reaching the membrane, improving filtration effectiveness and possibly minimizing fouling from pore constriction.

Fouling can be characterized as irreversible or reversible. An idealized graph of specific flux over time is shown on Fig. 12-18. Full-scale facilities generally operate in a constant-flux mode; therefore, a decline in specific flux is caused by an increase in transmembrane pressure while flux remains constant. A decline in specific flux occurs during initial operation, and a portion of this flux loss cannot be recovered during backwashing and

Reversibility of Fouling

Figure 12-18

Variation in specific flux during filtration of natural waters. The loss of specific flux from the initial clean membrane permeability, which cannot be recovered by backwashing or cleaning, is called irreversible fouling; that which can be recovered is called reversible fouling.



cleaning operations. Permanent flux loss is called irreversible fouling and depends on the source water quality as well as the type of membrane used. The specific flux declines further during each filter run (normally recorded as an increase in transmembrane pressure) but a significant portion can be recovered during backwashing. This loss of flux that can be recovered during backwashing is called hydraulically reversible fouling. Fouling due to cake formation is largely reversible during backwash. The longer-term, slower decline in specific flux over multiple filter runs is due to the slow adsorption and clogging of materials within the membrane matrix (pore constriction), which can be dissolved and removed during chemical cleaning. The loss of flux that can be recovered during cleaning is called chemically reversible fouling.

Resistance-in-Series Model

As noted in the previous sections, several factors may contribute to resistance to flow. The resistance-in-series model applies a resistance value to each component of membrane fouling, assuming that each contributes to hydraulic resistance and that they act independently from one another. Typical forms of the resistance-in-series model are

$$J = \frac{\Delta P}{\mu (\kappa_m + \kappa_{ir} + \kappa_{hr} + \kappa_{cr})} \quad (12-10)$$

$$= \frac{\Delta P}{\mu (\kappa_m + \kappa_c + \kappa_p)} \quad (12-11)$$

where κ_m = membrane resistance coefficient, m^{-1}
 κ_{ir} = irreversible fouling resistance coefficient, m^{-1}
 κ_{hr} = hydraulically reversible fouling resistance coefficient, m^{-1}
 κ_{cr} = chemically reversible fouling resistance coefficient, m^{-1}
 κ_c = cake layer resistance coefficient, m^{-1}
 κ_p = pore constriction resistance coefficient, m^{-1}

The resistance-in-series equation can be applied to any number of individual resistances, which may be due to irreversible and reversible components, specific fouling materials (organic fouling resistance, biological fouling resistance, etc.), or fouling mechanisms (cake fouling resistance, pore constriction fouling resistance, etc.).

Individual resistance coefficients can be calculated by selecting operating conditions in which individual forms of fouling can be isolated. The procedure for calculating individual resistance coefficients is demonstrated in Example 12-5.

Example 12-5 Calculation of resistance coefficients

The MF membrane in Example 12-3 is used under full-scale conditions in a water treatment facility, producing a flux of $84 \text{ L/m}^2 \cdot \text{h}$ at 1.1 bar just before cleaning and $106 \text{ L/m}^2 \cdot \text{h}$ at 0.52 bar immediately after cleaning, both at a standard temperature of 20°C . Calculate values for the membrane resistance coefficient, irreversible fouling resistance coefficient, and chemically reversible fouling resistance coefficient.

Solution

1. The membrane resistance coefficient was calculated in Example 12-3 (under conditions when $\kappa_{ir} = 0$ and $\kappa_{cr} = 0$) and found to be $3.81 \times 10^{11} \text{ m}^{-1}$.
2. Determine the irreversible fouling resistance coefficient.
 - a. The viscosity of water at 20°C from Table C-1 in App. C is $1.00 \times 10^{-3} \text{ kg/m} \cdot \text{s}$. Also recall that $1 \text{ bar} = 100 \text{ kPa} = 10^5 \text{ N/m}^2 = 10^5 \text{ kg/s}^2 \cdot \text{m}$.
 - b. The reversible component of fouling is removed by cleaning, so immediately after cleaning the chemically reversible fouling resistance coefficient is zero ($\kappa_{cr} = 0$). The only factors that cause resistance to flow are the membrane resistance and the irreversible fouling resistance, so the resistance-in-series equation can be written

$$J = \frac{\Delta P}{\mu (\kappa_m + \kappa_{ir})} \quad (1)$$

- c. Rearrange Eq. 1 to solve for κ_{ir} :

$$\begin{aligned} \kappa_{ir} &= \frac{\Delta P}{\mu J} - \kappa_m = \frac{(0.52 \times 10^5 \text{ kg/s}^2 \cdot \text{m})(3600 \text{ s/h})(1 \times 10^3 \text{ L/m}^3)}{(1.00 \times 10^{-3} \text{ kg/m} \cdot \text{s})(106 \text{ L/m}^2 \cdot \text{h})} \\ &\quad - 3.81 \times 10^{11} \text{ m}^{-1} \\ &= 1.39 \times 10^{12} \text{ m}^{-1} \end{aligned}$$

3. Determine the chemically reversible fouling resistance coefficient.
 a. Prior to cleaning, three components of resistance are present:

$$J = \frac{\Delta P}{\mu (\kappa_m + \kappa_{ir} + \kappa_{cr})}$$

- b. Rearrange the above equation to solve for κ_{cr} :

$$\begin{aligned} \kappa_{cr} &= \frac{\Delta P}{\mu J} - \kappa_m - \kappa_{ir} \\ &= \frac{(1.1 \times 10^5 \text{ kg/s}^2 \cdot \text{m}) (3600 \text{ s/h}) (1 \times 10^3 \text{ L/m}^3)}{(1.00 \times 10^{-3} \text{ kg/m} \cdot \text{s}) (84 \text{ L/m}^2 \cdot \text{h})} \\ &\quad - 3.81 \times 10^{11} \text{ m}^{-1} - 1.39 \times 10^{12} \text{ m}^{-1} \\ &= 2.94 \times 10^{12} \text{ m}^{-1} \end{aligned}$$

Comment

In this example, the chemically reversible fouling resistance coefficient is the largest resistance and nearly an order of magnitude larger than the membrane resistance coefficient, which demonstrates the importance of fouling on overall membrane performance.

Fouling by Particles

Large particles form a cake on the membrane surface. Fouling due to cake formation is often modeled using the resistance-in-series model. In laboratory studies using well-defined synthetic suspensions such as monodisperse spherical latex particles, the cake layer resistance coefficient in Eq. 12-11 can be calculated using the Kozeny equation for flow through a granular medium (see Chap. 11):

$$\kappa_C = \frac{36\kappa_K (1 - \varepsilon)^2 \delta_C}{\varepsilon^3 d_P^2} \quad (12-12)$$

where κ_C = cake layer resistance coefficient, m^{-1}
 κ_K = Kozeny coefficient, unitless (typically 5)
 ε = cake porosity, dimensionless
 δ_C = thickness of cake layer, m
 d_P = diameter of retained particles, m

In dead-end filtration, the thickness of the cake layer as a function of time can be calculated from the mass flux of particles toward the membrane

surface, assuming that the backward migration of particles due to diffusion can be neglected:

$$\delta_C(t) = \frac{CV}{\rho_P a (1 - \varepsilon)} \quad (12-13)$$

where $\delta_C(t)$ = thickness of cake layer at time t , m

C = concentration of particles, mg/L

V = volume of feed water filtered, m³

ρ_P = density of particles, kg/m³

a = membrane area, m²

In natural systems, the cake layer is an amorphous mat of polydisperse solids, so the cake layer resistance cannot be related easily to parameters such as particle diameter, porosity, and cake thickness. In this case, the cake resistance coefficient can be defined in terms of a specific cake resistance, or resistance per unit of mass loading. The mass loading is the mass of dry solids retained by the membrane per unit of membrane area. Because the influent solids concentration, volume filtered, and membrane area can be readily measured, the cake resistance as a function of time (via volume filtered) can be determined:

$$\kappa_C = \alpha_C \frac{CV}{a} \quad (12-14)$$

where α_C = specific cake resistance, m/g

During cross-flow filtration, cake layer formation is more complex because additional phenomena contribute to the transport of particles away from the membrane surface. The convective flow of particles toward the membrane is opposed by at least three mechanisms (Kim and DiGiano, 2009; Cheryan, 1998). First, the flow of water parallel to the membrane surface causes surface shear forces that drag particles downstream and minimize the formation of the cake. Second, the cross-flow velocity decreases the thickness of the concentration boundary layer, which increases the concentration gradient and enhances diffusion of particles away from the membrane surface. Finally, a velocity gradient exists close to the membrane surface such that particles in this velocity field are exposed to a higher velocity on the side opposite the membrane.

The differential velocity between the near and far sides of particles leads to inertial lift that draws them away from the membrane surface. The random movement of particles in this velocity field also enhances their migration toward streamlines moving at a higher velocity, a phenomenon that has been called shear-enhanced diffusion (Zydney and Colton, 1986). Shear-enhanced diffusion coefficients can be more than two orders of magnitude greater than Brownian diffusion coefficients (Belfort et al., 1994). Additional phenomena, such as electrostatic interactions (McDonogh et al.,

1988; Welsch et al., 1995), boundary layer separation (Treybal, 1980), gravitational forces, or van der Waals attractions (Wiesner and Chellam, 1992), may contribute to the movement of particles toward or away from the membrane surface.

A substantial number of models have been developed to predict the flux of membranes during cross-flow filtration of particle suspensions (Kim and DiGiano, 2009; Bacchin et al., 1995; Cheryan, 1998; Field et al., 1995; Koltuniewicz, 1992; Lee and Clark, 1997; Wiesner and Chellam, 1992). The trajectory of particles is evaluated in these models by considering all forces on individual particles. An important prediction of many of these models is that under certain conditions the net forces pushing particles away from the membrane are greater than the convective forces drawing particles to the membrane surface. As a result, no particle deposition and no fouling should occur. Wiesner et al. (1989) suggested that particles above a certain size should not deposit on the membrane, and others (Field et al., 1995) extended this to define the concept of a “critical flux,” the flux at which no particles deposit on the membrane surface and no fouling occurs. In addition, many of the models suggest that particles between about 0.1 and 1 μm in diameter exhibit the least backmigration from the membrane surface and will cause the greatest fouling during cross-flow filtration.

Biofouling

Biofouling is the loss of system performance due to the formation of a biofilm (Ridgway and Flemming, 1996). During filtration, microorganisms are transported to the membrane surface, where they can attach with sufficient force as to prevent removal during backwashing. Once attached, they can excrete extracellular material that causes additional fouling. Biofouling is particularly relevant in membrane filtration systems operated in wastewater applications, such as membrane bioreactors. In water treatment, biofouling is addressed by the use of chemical disinfectants such as chlorine in the feed water, backwash water, or both. The recent trend by manufacturers to use membrane materials with good chlorine resistance has helped reduce the importance of biofouling.

Natural Organic Matter Fouling

Fouling by particles can be managed by proper backwashing (as long as the particles are significantly larger than the membrane pores) and biofouling can be managed with proper disinfection. The most problematic and least controllable membrane fouling is due to the adsorption of natural organic matter (NOM) to the membrane surface. Fouling by NOM (or the dissolved fraction, DOM) has been confirmed with laboratory experiments with solutions of commercially available dissolved organic matter. The ability of DOM to adsorb to membranes has been demonstrated with traditional adsorption isotherms (Crozes et al., 1993; Jucker and Clark, 1994). Filtration of commercially available humic, fulvic, and tannic acid

solutions has been shown to lead to rapid membrane fouling (Crozes et al., 1993; Lahoussine-Turcaud et al., 1990; Yuan and Zydney, 1999, 2000). Surface cake formation and pore constriction have both been proposed as mechanisms for fouling (Combe et al., 1999; Kim et al., 1992; Yuan and Zydney, 1999; Yuan et al., 2002). The relationship between DOM adsorption and flux has not been successfully described mathematically, and there are currently no models that can predict the extent of DOM fouling as a function of water quality measurements. Fouling depends on characteristics of the DOM, the membrane material, and the solution properties, although the size and stability of the DOM appear to be the most important factors. The effects of several important factors on DOM fouling are identified in Table 12-8.

Some studies have found that removing a large fraction of DOM (up to 85 percent) by carbon adsorption (Lainé et al., 1990; Carroll et al., 2000; Lin et al., 2000) can have little or no reduction of fouling or, alternatively, removing a small fraction of DOM (less than 10 percent) by prefiltration through a 3-kD membrane (Howe, 2001) can completely eliminate fouling. Collectively, this research suggests that only a fraction of DOM causes the majority of fouling in membrane filtration, and this and other research suggests that the high-MW and colloidal fractions are the necessary components because they have the necessary dimensions to constrict membrane pores. Chemical properties and particle stability are also important (Huang et al., 2008a; Huang and O'Melia, 2008) because fouling will not occur unless the colloids have an affinity for attachment to the membrane pore walls.

Significant fouling by a small amount of DOM is possible when the relationship between transmembrane pressure and pore diameter under laminar flow conditions is considered. Poiseuille's law for laminar flow (see Eq. 11-10 in Chap. 11) indicates that pressure drop varies as the inverse fourth power of diameter under constant flow conditions (i.e., transmembrane pressure could double following a mere 16 percent decrease in pore diameter).

Strategies to minimize fouling by NOM, such as coagulation pretreatment, are discussed in Sec. 12-8.

Models that simulate fouling mechanisms have been developed for filtration under specific laboratory operating conditions and have come to be known as the blocking laws. The analytical solution for each fouling mechanism is shown in Table 12-9. The equations predict a different rate of flux decline for each type of fouling.

Hermia (1982) demonstrated that the equations in Table 12-9 can be written in a consistent format. By multiplying flux by membrane area and integrating with respect to time, the total volume of permeate produced can be determined. Rearranging the equations with time as the dependent

Blocking Filtration Laws for Membrane Fouling

Factors contributing to membrane fouling by dissolved organic matter (DOM)

Factor	Observed Effects
Hydrophobicity	Hydrophobic membranes adsorb more DOM and therefore foul more rapidly than hydrophilic membranes (Matthiasson, 1983; Laine et al., 1989; Cheryan, 1998). Hydrophobic fractions of DOM and hydrophobic sources of DOM are expected to cause greater fouling, which has been observed in some research (Crozes et al., 1993; Yuan and Zydney, 1999; Schäfer et al., 2000). However, researchers have also reported that hydrophilic fractions of DOM may be implicated in greater fouling (Amy and Cho, 1999; Carroll et al., 2000; Lin et al., 2000).
Electrostatic charge	Most DOM is negatively charged, and many MF and UF membranes acquire a slight negative charge during operation. Conditions that increase electrostatic repulsion might reduce fouling. The magnitude of the negative charge on membrane (Causserand et al., 1994; Nyström et al., 1994; Combe et al., 1999) and the negative charge on DOC both tend to increase at higher pH. As expected, low-pH conditions increase the adsorption of DOM to membranes (Jucker and Clark, 1994; Combe et al., 1999) and the fouling due to DOM adsorption (Kulovaara et al., 1999).
Size/molecular weight	Size may be an essential factor in determining which components of DOM cause fouling. Several studies suggest that high-MW and colloidal materials cause greater fouling (Lin et al., 1999, 2000; Yuan and Zydney, 1999, 2000; Habarou et al., 2001; Howe and Clark, 2002). Fouling by this colloidal fraction is consistent with the ability of larger material to constrict pores more efficiently than dissolved materials.
Colloidal stability	Since colloids must be smaller than the pore size to enter the membrane matrix, an additional mechanism must explain their attachment to the pore walls. A model developed by Huang et al. (2008a) and supported by experimental results indicated that colloids with low particle–membrane stability and high particle–particle stability caused the greatest fouling.
Ionic strength	High ionic strength reduces electrostatic repulsion (and particle stability) by compressing the double layer, and irreversible fouling has been shown to increase at high ionic strength (e.g., seawater) (Kulovaara et al., 1999).
Calcium concentration	Calcium ions may act as a positively charged bridge between DOM and membrane surfaces. Calcium has been shown to neutralize the negative charge on DOM and increase the adsorption of NOM on membranes (Jucker and Clark, 1994) and contribute to greater flux decline (Schäfer et al., 2000).

Table 12-9
Blocking filtration laws

Flux Equation	Equation Number	Filtration Coefficient, k	Filtration Exponent, n	Major Features and Assumptions
Complete Blocking Filtration Law (Pore Sealing)				
$J_t = J_0 \exp \left(-1.5 \frac{CJ_0 t}{\rho_P d_P} \right)$	(12-16)	$\frac{1.5CJ_0}{\rho_P d_P}$	2	<div> <input type="checkbox"/> Models blockage of the entrance to pores by particles retained at the membrane surface. <input type="checkbox"/> Each retained particle blocks an area of the membrane surface equal to the particle's cross-sectional area. <input type="checkbox"/> Flux declines in proportion to the membrane area that has been covered. <input type="checkbox"/> No superposition of particles occurs. Each particle lands on the membrane surface and not on other particles, so flux reaches zero when a monolayer of particles has been retained. </div>
Standard Blocking Filtration Law (Internal Pore Constriction)				
$J_t = \frac{J_0}{\left(1 + \frac{CJ_0 t}{L\rho_P} \right)^2}$	(12-17)	$\frac{2C}{L\rho_P} \left(\frac{J_0}{a} \right)^{0.5}$	1.5	<div> <input type="checkbox"/> Models the reduction of the void volume within the membrane. <input type="checkbox"/> Assumes the membrane is composed of cylindrical pores of constant and uniform diameter. <input type="checkbox"/> Particles deposit uniformly on the pore walls; pore volume decreases proportionally to the volume of particles deposited. <input type="checkbox"/> L = membrane thickness, m </div>

(continues)

Table 12-9 (Continued)

Flux Equation	Equation Number	Filtration Coefficient, k	Filtration Exponent, n	Major Features and Assumptions
Intermediate Blocking Filtration Law (Pore Sealing with Superposition)				
$J_t = \frac{J_0}{\left(1 + 1.5 \frac{C J_0 t}{\rho_P d_P}\right)}$	(12-18)	$\frac{1.5C}{\rho_P d_P a}$	1	<ul style="list-style-type: none">Models blockage of the entrance to pores by particles retained at the membrane surface.Extension of the complete blocking filtration law.Relaxes the “monolayer” assumption in the complete blocking filtration law by allowing particles to land on previously retained particles or on the membrane surface by evaluating the probability that a particle will block a pore.
Cake Filtration Law				
$J_t = \frac{J_0}{\left(1 + 2 \frac{\alpha_C C J_0 t}{\kappa_M}\right)^{0.5}}$	(12-19)	$\frac{\alpha_C C}{\kappa_M J_0 a^2}$	0	<ul style="list-style-type: none">Models the formation of a cake on the surface of a membrane using the resistance-in-series model.The retained particles have no impact on the membrane itself, i.e., no pore blocking or pore constriction.

variable and taking the first and second derivatives with respect to volume, all four filtration laws can be written in the form

$$\frac{d^2 t}{dV^2} = k \left(\frac{dt}{dV} \right)^n \quad (12-15)$$

where t = time, s

V = volume, L

k = blocking law filtration coefficient, units vary depending on n

n = blocking law filtration exponent, unitless

The parameters k and n are defined for each filtration law in Table 12-9. The form of the filtration laws shown in Eq. 12-15 can be used to analyze experimental data to determine which law most closely approximates the data. Permeate volume is measured at constant time intervals and tabulated in a spreadsheet. Using the spreadsheet, the derivatives $d^2 t/dV^2$ and dt/dV can be determined. The slope of the line is the coefficient n from Table 12-9.

Many researchers have fit laboratory flux data to these equations in an attempt to isolate which mechanism causes fouling under specific conditions. Despite their use in research studies, however, the equations have failed to have much predictive value to relate bench studies to full-scale applications for several reasons. First, membranes are modeled as perfectly flat and uniformly porous with vertical cylindrical pores, which does not match the membranes used in commercial applications. Second, filtration is modeled as being under constant-pressure, declining flux conditions, compared to the constant-flux, rising pressure situation normally used during water treatment. Third, the original models envision flux as declining due to a single mechanism and the shape of the flux decline curve reveals the fouling mechanism, whereas in full-scale operation several fouling mechanisms probably occur simultaneously. Finally, the original model equations have few or no parameters related to membrane properties so it is not possible to analyze, for instance, the impact of pore size on which mechanism might be most important for fouling by a particular size particle. More sophisticated models have been developed recently to address some of these limitations (Ho and Zydney, 2000; Chellam and Xu, 2006; Cogan and Chellam, 2009), but current models do not yet have predictive value for full-scale applications.

The filtration blocking laws apply only to constant-pressure, dead-end filtration. Models for flux decline have been developed for other operating conditions with varying levels of complexity (Belfort et al., 1994; Chang and Benjamin, 2003; Fane and Fell, 1987; Yuan et al., 2002; Kim and DiGiano, 2009). Theory regarding fouling of MF and UF membranes is evolving, and the current technical literature should be consulted if an in-depth understanding of flux modeling is required.

Membrane Fouling Index

In the absence of fundamental models that can predict full-scale performance, it is useful to have empirical models that can compare fouling under different conditions, such as with different source waters, different membrane products, or at different scales. A fouling index can be derived using the resistance-in-series model with two resistance terms: one for clean membrane resistance and another for fouling resistance:

$$J = \frac{\Delta P}{\mu (\kappa_m + \kappa_f)} \quad (12-20)$$

where κ_f = resistance due to all forms of fouling, m^{-1}

With the basic assumption that fouling resistance is directly proportional to the mass of foulants that have been transported to the membrane surface with the feed water, the fouling resistance can be related to the amount of water filtered per unit of membrane area, that is,

$$\kappa_f = k V_{sp} \quad (12-21)$$

where k = rate of increase in resistance, m^{-2}

V_{sp} = specific throughput, volume of water filtered per membrane area, m^3/m^2

After conversion to flux at a standard temperature using Eq. 12-8, Eq. 12-21 can be written in terms of specific flux:

$$J_{sp} = \frac{J_s}{\Delta P} = \frac{1}{\mu (\kappa_m + k V_{sp})} \quad (12-22)$$

For a new membrane, $V_{sp} = 0$ so $\kappa_f = 0$, so

$$J_{sp0} = \frac{1}{\mu \kappa_m} \quad (12-23)$$

Membrane filtration performance is typically evaluated by comparing the flux over time to the initial flux through the membrane when it was new. Clean membrane permeability can vary from one membrane sample to another due to slight variations in membrane pore dimensions, thickness, or porosity because of manufacturing variability. Normalizing against new membrane performance eliminates membrane sample variability when comparing experiments. Dividing by clean membrane specific flux yields

$$J'_{sp} = \frac{J_{sp}}{J_{sp0}} = \frac{1/[\mu (\kappa_m + k V_{sp})]}{1/(\mu \kappa_m)} = \frac{\kappa_m}{\kappa_m + k V_{sp}} \quad (12-24)$$

A fouling index can be defined as the slope of the line when the inverse of J'_{sp} is plotted as a function of the specific throughput:

$$\frac{1}{J'_{sp}} = 1 + (\text{MFI}) V_{sp} \quad (12-25)$$

where $\text{MFI} = k/\kappa_m = \text{membrane fouling index, m}^{-1}$

The MFI is an empirical fouling index that can be used to compare the rate of fouling between experiments or between bench and pilot-scale results. The MFI is valid for any form of fouling as long as the fouling resistance is directly proportional to the specific throughput. The use of specific flux allows filter runs at either constant pressure or constant flux to be compared, because J_{sp} declines as membranes foul regardless of whether filtration occurs at constant pressure (J declines at constant ΔP) or constant flux (ΔP increases at constant J). Specific throughput allows runs of different duration or systems with different membrane area (i.e., different scale) to be compared. Huang et al. (2008b) demonstrated that Eq. 12-25 could be derived as an approximation for standard blocking, intermediate blocking, or cake filtration mechanisms under either constant-flux or constant-pressure filtration scenarios. The MFI has been used to compare fouling between different membrane products and source waters, and studies have shown reasonably good agreement between MFI values using bench-scale and pilot-scale data with the same membrane and source water (Huang et al., 2008b, 2009b).

The MFI can be calculated using either a linear regression of flux data or as the slope of the line between two points, depending on the data available. Calculation of the MFI using both methods is demonstrated in Example 12-6.

Early bench-scale studies to explore membrane filtration and understand fouling were often done with flat-sheet membranes in a constant-pressure operating mode over a single filter run. A typical flat-sheet membrane experimental setup of this type is shown on Fig. 12-19a. While the research

Evaluating Fouling with Bench-Scale Studies

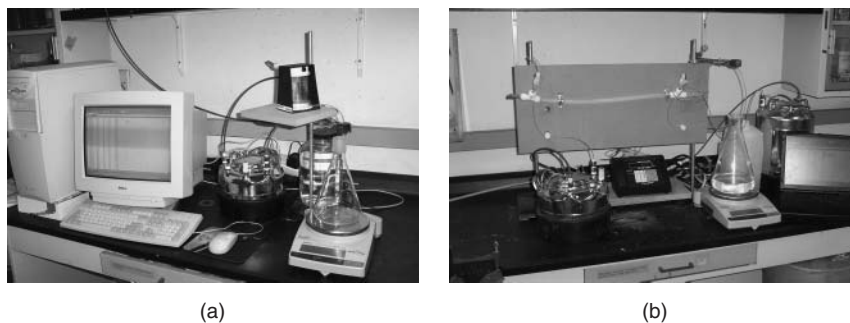


Figure 12-19
Equipment for bench-scale testing of membrane filtration: (a) flat-sheet membrane cell configuration and (b) backwashable hollow-fiber configuration.

Example 12-6 Calculation of the membrane fouling index

A laboratory membrane experiment using a backwashable single-fiber membrane module was carried out to collect the data in Fig. 12-16. The membrane had a total area of 23.0 cm^2 and the initial permeability of the new membrane was $225.0 \text{ L/m}^2 \cdot \text{h} \cdot \text{bar}$. The test was run at a constant pressure of 1.023 bar and temperature of 22°C . The membrane was backwashed every 30 min. Time and volume filtered were recorded at 2-min intervals and the data from filter run 6 is shown in the first two columns of Table 1 below. The flux at the beginning of each of the first 10 filter runs is also shown in Table 2 below. Calculate the fouling index during filter run 6 and the hydraulically irreversible fouling index (fouling that corresponds to the flux that could not be recovered by backwashing).

Solution

1. Divide the volume filtered by the membrane area to determine the specific throughput. Results are in column (3) in Table 1. For the second row,

$$V_{sp} = \frac{(743.92 \text{ mL}) (10^4 \text{ cm}^2/\text{m}^2)}{(23.0 \text{ cm}^2) (10^3 \text{ mL/L})} = 323.4 \text{ L/m}^2$$

2. Calculate the volume filtered in each time increment by subtracting the previous volume. Results are in column (4) in Table 1. For the second row,

$$\Delta V = 743.92 \text{ mL} - 732.63 \text{ mL} = 11.29 \text{ mL}$$

3. Divide the volume filtered in each increment by membrane area and time to determine flux. Then correct for temperature and pressure using Eqs. 12-8 and 12-9 to determine specific flux. Results are in column (5) in Table 1. For the second row,

$$J_m = \frac{(11.29 \text{ mL}) (10^4 \text{ cm}^2/\text{m}^2) (60 \text{ min/h})}{(23.0 \text{ cm}^2) (2 \text{ min}) (10^3 \text{ mL/L})} = 147.3 \text{ L/m}^2 \cdot \text{h}$$

$$J_{sp} = \frac{J_m (1.03)^{T_s - T_m}}{\Delta P} = \frac{147.3 \text{ L/m}^2 \cdot \text{h} (1.03)^{20 - 22}}{1.023 \text{ bar}}$$

$$= 135.7 \text{ L/m}^2 \cdot \text{h} \cdot \text{bar}$$

4. Divide the specific flux (J_{sp}) by the initial specific flux (J_{sp0}). Results are in column (6) in Table 1. For the second row,

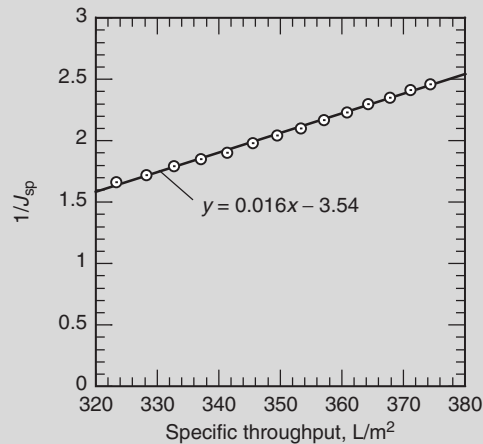
$$J'_{sp} = \frac{135.7}{225.0} = 0.60$$

5. Invert the normalized flux from column 6. Results are in column (7) in Table 1.

Example 12-6 Table 1

(1)	(2)	(3)	(4)	(5)	(6)	(7)
Filtration Time, min	Volume Filtered, mL	Specific throughput, L/m ²	Delta volume, mL	Specific flux, L/m ² · h	Normalized specific flux, J'_{sp}	Inverse normalized specific flux, $1/J'_{sp}$
0	732.63	—	—	—	—	—
2	743.92	323.4	11.29	135.7	0.60	1.66
4	754.79	328.2	10.87	130.6	0.58	1.72
6	765.26	332.7	10.47	125.8	0.56	1.79
8	775.40	337.1	10.14	121.9	0.54	1.85
10	785.17	341.4	9.77	118.4	0.53	1.90
12	794.63	345.5	9.46	113.7	0.51	1.98
14	803.79	349.5	9.16	110.1	0.49	2.04
16	812.70	353.3	8.91	107.1	0.48	2.10
18	821.34	357.1	8.64	103.8	0.46	2.17
20	829.73	360.8	8.39	100.8	0.45	2.23
22	837.88	364.3	8.15	97.9	0.44	2.30
24	845.85	367.8	7.97	95.8	0.43	2.35
26	853.62	371.1	7.77	93.4	0.42	2.41
28	861.22	374.4	7.60	91.3	0.41	2.46

6. Plot the inverse of the normalized specific flux ($1/J'_{sp}$) as a function of the specific throughput (V_{sp}), as shown in the following figure:



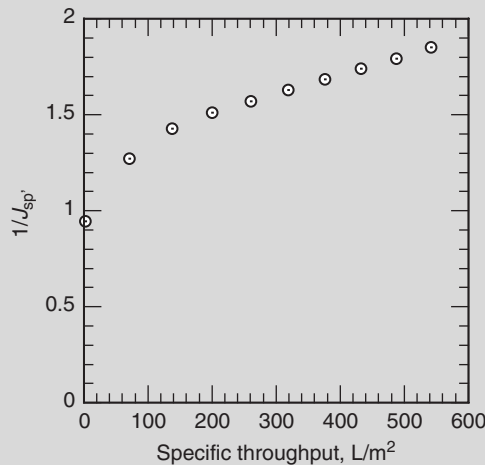
The slope of the line is the membrane fouling index for filter run 6, $MFI_6 = 0.016 \text{ m}^2/\text{L} = 16 \text{ m}^{-1}$. Note that the intercept of the graph is not 1.0 as is suggested by Eq. 12-25. This result is because backwashes remove foulants and reset membrane performance to a higher flux, whereas the specific volume progresses continuously. For an initial filter run (i.e., before any backwashes or cleanings), the intercept is very close to 1.0.

7. Determine the hydraulically irreversible membrane fouling index (MFI_{hi}). The MFI_{hi} represents the flux that cannot be recovered by backwashing and can be evaluated by considering the net reduction in flux at the beginning of each filter run (immediately after backwashing). Data from the first 10 filter runs of the experiment shown in Fig. 12-16 is shown in Table 2 below. Column (1) is the filter run number, Column (2) is the specific throughput at the beginning of each filter run, and Column (3) is the average specific flux over the first 30 of each filter run.

Example 12-6 Table 2

(1)	(2)	(3)	(4)	(5)
Filter Run	Specific throughput, L/m^2	Specific flux, $\text{L}/\text{m}^2 \cdot \text{h}$	Normalized specific flux, J'_{sp}	Inverse normalized specific flux, $1/J'_{sp}$
1	2.2	238.0	1.06	0.95
2	71.3	176.9	0.79	1.27
3	137.6	157.7	0.70	1.43
4	200.0	149.0	0.66	1.51
5	260.5	143.3	0.64	1.57
6	319.0	138.0	0.61	1.63
7	376.4	133.6	0.59	1.68
8	432.6	129.3	0.57	1.74
9	487.9	125.5	0.56	1.79
10	542.4	121.6	0.54	1.85

8. A graph of the inverse of the normalized flux ($1/J'_{sp}$) as a function of the specific throughput is shown in the following figure:



The graph indicates more rapid fouling during the first two filter runs (i.e., the first two runs are not linear with the rest of the data), and a linear regression through all of the data would not reflect the longer-term fouling index. The long-term hydraulically irreversible membrane fouling index can be calculated as a straight line between runs 3 and 10:

$$\text{MFI}_{\text{hi}} = \frac{(1/J'_{\text{sp}})_{10} - (1/J'_{\text{sp}})_3}{(V_{\text{sp}})_{10} - (V_{\text{sp}})_3} = \frac{1.850 - 1.427}{542.4 \text{ L/m}^2 - 137.6 \text{ L/m}^2}$$

$$= 0.00104 \text{ m}^2/\text{L}$$

$$\text{MFI}_{\text{hi}} = (0.00104 \text{ m}^2/\text{L})(10^3 \text{ L/m}^3) = 1.04 \text{ m}^{-1}$$

has helped improved the industry's understanding of membrane filtration, researchers have gradually realized that performance with this system did not effectively simulate full-scale commercial membrane filtration systems. New membranes tend to experience an initial period of irreversible fouling that might mask longer-term fouling trends. Constant-pressure filtration through a flat-sheet membrane can cause very high initial flux that leads to rapid fouling in the first moments of operation. Studies have shown different fouling with constant-pressure operation than with constant-flux operation (Tarabara et al., 2002). Commercial membrane manufacturers often have proprietary chemical formulations or fabrication methods to optimize performance, and use of a laboratory-grade flat-sheet membrane may not adequately represent the commercial product, even if nominally

of the same material. Studies have demonstrated that rates of fouling of hollow-fiber and flat-sheet membranes are not the same even when made of the same material and filtering the same water (Howe et al., 2007).

To rectify these limitations, researchers have developed small-scale hollow-fiber modules (often with only one or two fibers) that can be used at bench scale but more effectively simulate pilot- or full-scale systems (Chang and Benjamin, 1996; Kim and DiGiano, 2006; Huang et al., 2009b). A typical hollow-fiber experimental system is shown on Fig. 12-19b. The hollow-fiber membranes are exactly the same product as used in commercial membrane modules. The bench-scale modules can be operated in either inside-out or outside-in flow configurations to match full-scale operation and can be backwashed to allow operation over multiple filter runs. Constant flux operation can be simulated, although pumps must be selected carefully because low-flow positive-displacement pumps often used in laboratories may produce pulsations that are not characteristic of centrifugal pumps used in pilot- or full-scale applications. While the hydraulics of small-scale systems are not identical to commercial modules, they are more representative of larger systems than earlier flat-sheet bench-scale systems were. The membrane fouling index presented in the previous section offers the potential to relate bench-scale membrane performance to pilot- or full-scale performance, although more experimental validation of this approach is needed.

12-8 Process Design

Membrane filtration design concepts are changing rapidly, so the application of this technology presents unique challenges for the design engineer. Design based on previous projects or “tried-and-true” rules of thumb may fail to capitalize on recent technological advancements. On the other hand, undue reliance on manufacturers’ claims about unproven technologies may lead to failure. A critical role for the design engineer is to stay abreast of technical advancements and provide appropriate guidance to facility owners. An understanding of the fundamentals of membrane materials, modules, fouling, and performance is necessary to evaluate new technologies with the objective of allowing the design to capitalize on valuable technological advancements while avoiding unproven alternatives that have an unreasonable chance of failure. Detailed guidance and design manuals for membrane filtration systems have recently been published by EPA and AWWA (U.S. EPA, 2005; AWWA, 2005b, 2008).

The primary tasks for the design engineer during preliminary design of a membrane filtration installation include:

1. Set performance criteria, such as retention criteria, plant capacity, and recovery, based on raw-water resources, finished-water quality goals, and projected water demand.

2. Evaluate the need for supplementary unit processes such as pretreatment or disinfection in the overall treatment facility, based on raw and finished-water quality and the capabilities of membrane filtration.
3. Evaluate process alternatives, including new developments and technological advancements. Frequently, this evaluation will include pilot studies and evaluation of the construction and O&M costs of the appropriate alternatives.
4. Establish process design criteria and develop specifications for major system components. Pilot studies contribute to the basis of design.
5. Predesign ancillary and support facilities, such as transfer piping, pumping facilities, chemical storage facilities, laboratory space, buildings, and electrical systems. Evaluate hydraulic-grade line and waste washwater disposal options.

Important performance criteria include retention capabilities, capacity requirements, and recovery. Performance criteria also provide a basis for determining whether membrane filtration must be coupled with other treatment technologies.

Performance Criteria

RETENTION CAPABILITIES

As noted in Sec. 12-5, MF membranes achieve excellent rejection of most microorganisms but are not a complete barrier to viruses. Some UF products do provide excellent virus rejection, suggesting that there might be an advantage to specifying UF membrane systems exclusively. However, viruses are readily inactivated by free chlorine, and most regulatory agencies, if not all, will require primary disinfection as part of a multibarrier approach to water treatment. Thus, while some UF membranes may provide a more robust treatment train with respect to virus removal, as a practical matter there is generally no regulatory or cost advantage to selecting UF membranes over MF membranes when specifying acceptable membrane technologies.

Because membrane filtration is still a relatively new water treatment process, the regulatory system in the United States does not have uniform criteria for the rejection capabilities of membrane filters. Criteria are applied at the state level and vary between states. In the United States, most states with guidelines for membrane filtration grant between 2 and 4 log rejection credits for *Giardia* and *Cryptosporidium* and between 0 and 4 log rejection credits for viruses. Numerous states grant the same log rejection credit regardless of whether the system is marketed as an MF or UF system (Herschell, 2007). The *Cryptosporidium* challenge testing requirements in the LT2ESWTR (U.S. EPA, 2005) are becoming the de facto standard for specifying rejection requirements for membrane systems.

Effluent turbidity is an important performance criterion in the design of granular filtration facilities. For membrane filtration design, turbidity

provides little guidance because all properly operating membrane filters will reduce turbidity to very low levels.

PLANT CAPACITY AND RECOVERY

Plant capacity is governed by the anticipated water demand at the end of the design life. Summer and winter demand must be considered separately because of the effect of temperature on permeate flux. In most locales, summer water demand is higher than winter demand, which fortunately corresponds to the seasonal variation in water temperatures. For each season, required plant size should be determined for the peak-day demand and minimum water temperature, which are worst-case conditions.

Recovery is the ratio of net water production to gross water production over a filter run:

$$r = \frac{Q_p}{Q_f} = \frac{V_f - V_{bw}}{V_f} \quad (12-26)$$

where r = recovery

Q_p = permeate flow rate, m^3/d

Q_f = feed flow rate, m^3/d

V_f = volume of water fed to membrane over filter run, m^3

V_{bw} = volume of water used during backwash, m^3

The calculation for recovery is identical to that for granular filters, except that common terminology in granular filtration (UFRV and UBWV) currently is not used in membrane filtration. Recovery in membrane filtration is typically 95 to 98 percent, which is comparable to rapid granular filters. If waste washwater is recovered, processed, and recycled to the feed stream, even higher recovery (greater than 99 percent) can be achieved.

Integration with Other Treatment Processes

Application of membrane filtration for water treatment started with small facilities with straightforward treatment requirements, so the membranes were a stand-alone process with no significant pretreatment or integration with other processes (except postmembrane disinfection). Now membrane filtration is an alternative to granular filtration regardless of source water quality. Integration of membrane filters as part of a larger process train is now the norm (Adham et al., 2005). Membrane filtration is used as pretreatment for other processes, such as granular activated carbon (GAC) adsorption or reverse osmosis. Processes that precede membrane filtration include coagulation and flocculation, sedimentation, granular filtration, adsorption, oxidation, and softening. A substantial amount of technical literature about pretreatment for membrane filtration is available, including reviews by Farahbakhsh et al. (2004) and Huang et al. (2009a).

Pretreatment is employed for two major reasons. First, additional processes can remove contaminants that are not removed by membrane

filtration alone, such as dissolved constituents. For example, coagulation is used for removal of arsenic; GAC is used for removal of taste, odor, and synthetic organic chemicals; oxidants are used for the removal of iron and manganese; and lime is used for the removal of hardness. Other technologies are being explored for other treatment objectives.

Second, pretreatment can enhance membrane performance by reducing the solids loading on the membrane process or by reducing membrane fouling. High turbidity sources may require excessive backwashing unless coagulation, flocculation, and sedimentation are used to remove a significant fraction of the solids, a strategy identical to conventional treatment using granular filtration. Processes can also remove membrane foulants, allowing the membrane system to operate at higher flux or with decreased backwashing and cleaning requirements. If the flux enhancement is significant, the cost of pretreatment may be offset by decreased membrane system costs.

Regardless of the reason for applying pretreatment, the impact on the membrane process must be considered. While some pretreatment can substantially improve membrane performance, other pretreatment can foul or even damage the membranes. Bench or pilot testing is recommended to evaluate the impact of chemicals on membranes. The two most common pretreatment strategies, coagulation and adsorption, are considered in more detail in the following sections.

COAGULATION PRETREATMENT

Coagulation and flocculation, sometimes with settling, is the most common pretreatment for membrane filtration. Coagulation can remove 15 to 50 percent of the NOM in natural waters and may be appropriate for utilities that need some NOM removal to meet DBP regulations. Coagulation with settling allows membrane filtration to be applied to source waters with higher and more variable turbidity.

Coagulation frequently reduces membrane fouling, and many studies and full-scale facilities have reported higher flux or reduced cleaning requirements when coagulation is used (Huang et al., 2009a). As noted earlier, high-MW and colloidal constituents may be a primary factor in fouling by NOM, and coagulation is effective at removing the higher-MW and more hydrophobic components of NOM. In some cases, however, coagulation has made fouling worse (Adham et al., 2006; Schäfer et al., 2001; Shorney et al., 2001; Shrive et al., 1999). The effect of coagulation on membrane fouling is site specific and is due to specific interactions between coagulants, feed water components, and membrane materials. One factor in the differences between the studies reported above is the coagulant dose: When the dose is sufficient to remove a significant fraction of the NOM, membrane performance may be improved due to the reduction of fouling (Howe and Clark, 2006). Until further research clearly identifies the interactions between coagulants, foulants, and membranes,

pilot studies are necessary to assess the effect of coagulation on membrane performance.

Experience with polymeric coagulant aids is mixed. Some studies have reported improved membrane performance when coagulant aids were used during coagulation, but others have reported that the polymers irreversibly fouled the membranes. Some manufacturers discourage the use of cationic polymers as pretreatment to their membrane filter systems.

POWDERED ACTIVATED CARBON–MEMBRANE REACTORS

Powdered activated carbon has been applied in the raw water of conventional treatment facilities for the removal of taste and odors as well as a variety of synthetic organic compounds. In conventional facilities PAC use can be complicated by competition from other treatment chemicals, limited contact time due to settling of the PAC in sedimentation basins, or breakthrough of the PAC through the filters and into the distribution system. The coupling of PAC treatment with membrane filtration provides a unique process without these disadvantages. Additional details of adsorption by PAC are provided in Chap. 15.

In the typical PAC–membrane reactor, PAC is added to the feed water upstream of the membranes. By using pressure-vessel membranes in a cross-flow mode or by using submerged membranes, the PAC is recycled to the feed water, increasing contact time and maximizing carbon utilization. Powdered activated carbon is considerably larger than the membrane pores, so passage of PAC into the distribution system is avoided. A recirculating or submerged membrane filtration system provides an ideal reactor for PAC adsorption, and this process is in use at full-scale water treatment plants (Anselme et al., 1999; Petry et al., 2001).

Powdered activated carbon adsorption is affected by the operating mode. The PAC can be added and withdrawn continuously during filtration in a feed-and-bleed configuration or the PAC can be added to an upstream reactor and allowed to accumulate over a filtration cycle. Each operating mode provides a different contact time distribution between the PAC and the contaminants. Campos et al. (2000a,b) determined that dosing carbon at the beginning (i.e., pulse input) of the filtration run rather than continuously resulted in lower average concentrations of contaminants.

While PAC is generally effective at removing synthetic organics, its ability to improve membrane performance is less evident. Although some studies have concluded that PAC can improve membrane performance, others have shown only minor improvement or worse performance. The primary reason PAC is less effective than coagulation at improving membrane performance is that activated carbon tends to remove the lower-MW components of NOM. The high-MW components of NOM that are implicated in membrane fouling tend to be excluded from the small pores of activated carbon.

**System
Components**

Currently, membrane filtration systems are available as modular systems from several manufacturers. Current vendors of membrane filtration equipment are listed in AWWA (2005b). Other suppliers are expected to enter the market as the technology evolves, including suppliers experienced in the reverse osmosis (see Chap. 17) market. Systems are often offered as a package containing all necessary filtration and ancillary equipment. Integral parts of membrane filtration facilities are the pretreatment processes, backwashing facilities, and cleaning facilities. These components are discussed in the following sections.

PRETREATMENT

When processes for nonparticulate treatment goals are not present, the pretreatment requirements for membrane filtration are minimal. Pretreatment is necessary to protect the filter fibers from damage or clogging of the lumen (in the case of inside-out membranes). Microscreening or prefiltration to remove coarse sediment larger in diameter than 0.1 to 0.5 mm, depending on the manufacturer, is required. Prefiltration is accomplished with self-cleaning screens, cartridge filters, or bag filters.

Because the primary removal mechanism is straining, chemical conditioning to destabilize particles is not required, although coagulation is now often applied as pretreatment for other reasons. The lack of a requirement for particle destabilization can be an advantage over granular filtration because the elimination of coagulation and flocculation facilities reduces chemical handling and storage facilities and residual management requirements.

BACKWASHING

The objective of backwashing is to remove the surface cake that develops during the filtration cycle. Backwashing occurs automatically at timed intervals ranging from 30 to 90 min. The increase in transmembrane pressure during the filtration cycle is typically 0.01 to 0.07 bar (0.2 to 1 psi). Most systems also initiate the backwash cycle early if the increase in transmembrane pressure during the filter run exceeds a preset limit. The backwash cycle lasts 1 to 3 min, and the sequence is run entirely by the control system. All modules in a rack are backwashed simultaneously. Backwashing of MF membranes involves forcing either air or permeate water through the fiber wall in the reverse direction at a pressure higher than the normal filtration pressure. Ultrafiltration membranes are backwashed with permeate water because the air pressure required to force water from the small pores in UF membranes can be excessive. In some pressure-vessel systems, the backwash flow is supplemented by a high-velocity flush in the feed channels to assist with removing the surface cake, and the wastewater is piped to a washwater handling facility. The backwash water in submerged systems flows directly into the feed tank.

Many membrane systems periodically add chemicals to backwash water to improve the backwash process, a sequence called chemically enhanced backwash (CEB). CEB chemicals can include hypochlorite or other cleaning chemicals. Chemically enhanced backwash reduces the rate of membrane fouling and can decrease the required frequency for the more extensive clean-in-place (CIP) cycle, which is discussed below.

Many membrane systems use a single backwash system to service multiple racks. The system is sized to backwash a single rack at a time, so backwashing of multiple racks must be staggered. The automated backwash sequence is a complex operation that involves the sequencing of numerous valves, which may take several minutes. The design engineer must consider the time requirements to complete one backwash cycle and ensure that there is sufficient time for all units to be washed within the allowable time between backwashes, including a factor of safety. If sufficient time is not available, more than one backwash system may be necessary.

CLEANING

Despite frequent backwashing, membrane filters gradually lose filtration capacity due to clogging or adsorption of material that cannot be removed during the backwash cycle. When the transmembrane pressure increases to a preset maximum limit or when a preset time interval has elapsed, the membranes are chemically cleaned. Chemical cleaning frequency ranges from a few days to several months depending on the membrane system characteristics and source water quality. The cleaning procedure typically takes several hours and involves circulating cleaning solutions that have been heated to 30 to 40°C. Cleaning solutions are proprietary mixtures provided by membrane manufacturers but are often high-pH solutions containing detergents or surfactants, which are effective for removing organic foulants. Low-pH solutions such as citric acid can be used for removing inorganic foulants. For some membrane materials (e.g., cellulose acetate), the pH of the cleaning solution is limited by the pH compatibility of the material. The membranes in both pressure-vessel and submerged systems are typically cleaned without removing the membranes from the modules, so the process is typically called the clean-in-place cycle.

POSTTREATMENT

The membrane filtration process has no inherent posttreatment requirements. Fluoridation or pH adjustment may be added after membrane filtration to fulfill other treatment objectives. Although membrane filtration is capable of completely removing microorganisms, disinfection is normally practiced after filtration as part of the multibarrier concept and to provide a disinfectant residual in the distribution system. Most state regulatory agencies have specific regulations for chemical disinfection following filtration.

**Integrity Testing
and Monitoring**

Membrane integrity monitoring involves procedures to verify that membrane filters are meeting treatment objectives. Integrity monitoring is important because of the physical characteristics of the filtration barrier. In a granular filtration plant, water is cleaned gradually as it flows through a series of processes, ending with a thick bed of filter media; clean water and dirty water are separated in both time and space. In a membrane filtration plant, water is cleaned nearly instantaneously as it flows through a thin membrane; clean water and dirty water are separated by a distance less than 1 mm and time less than 1 s. In addition, broken fibers or leaking O-ring connectors may compromise the filtration system. The regulatory framework for integrity monitoring is provided as part of the LT2ESWTR (U.S. EPA, 2006).

Integrity monitoring for membrane filtration has both direct and indirect components. As defined in the LT2ESWTR, the direct integrity test is a physical test that is sensitive enough to detect a 3- μm breach in the membrane system, is conducted at least one per day, and can verify the log removal value awarded to the membrane process. Continuous indirect integrity monitoring is the measurement of a water quality parameter that is indicative of particle removal, such as turbidity or particle counts.

INDIRECT INTEGRITY MONITORING

Indirect integrity monitoring is the continuous (at least every 15 min) monitoring of a suitable effluent water quality parameter, such as turbidity or particle levels. Indirect integrity monitoring is not as sensitive as direct integrity testing, but it has the advantages that it can be applied continuously and uses commercially available equipment that can be used with any membrane system (whereas most direct integrity testing equipment is proprietary to the specific membrane system manufacturer). Therefore, it is complementary to direct integrity testing in an overall integrity verification program.

Membrane filter effluent may be monitored for turbidity or particle concentrations. Particle counters, in which particles are classified according to size, are in common use in water treatment facilities (see Chap. 2). Particle monitors are a less expensive alternative to particle counters and provide only a relative measurement of total particulate matter in water. Particle counting is generally more sensitive than turbidity monitoring.

The sensitivity of effluent monitors is dependent on the filtration area being monitored. A study performed by Adham et al. (1995) showed that turbidity monitoring was able to detect a single pinhole in 5 m² of membrane area, whereas a particle monitor and a batch particle counter were able to detect similar breaches in 12 and 720 m² of membrane area, respectively. However, individual membrane modules may have 40 to 80 m² of membrane area and racks may contain 2 to 100 modules. The limitation of turbidity for integrity monitoring, which drives the need for direct integrity testing, is illustrated in Example 12-7.

Example 12-7 Indirect integrity monitoring with turbidity

A membrane filtration plant treating a feed water with 10^6 microorganisms/L and turbidity of 10 NTU reduces both to below detection limits. For this example, assume the detection limits are 1 org/L for microorganisms and 0.03 NTU for turbidity. Using this information, calculate (1) the log rejection value for microorganisms under normal operation and (2) the effluent turbidity and microorganism concentration and the rejection of microorganisms assuming the membrane develops a hole that allows 0.01 percent of the water to pass through untreated.

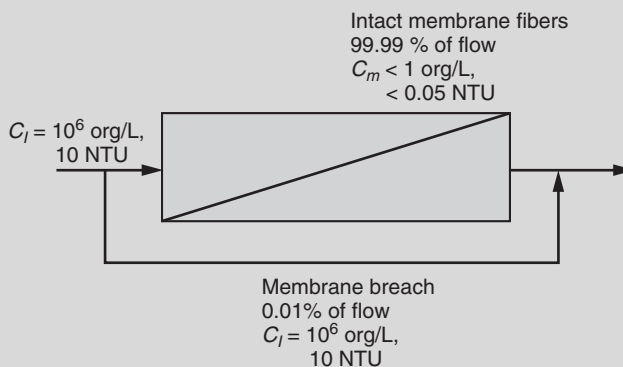
Solution

1. Calculate the log rejection value under normal operation. Use the detection limit for the effluent concentration and Eq. 12-4:

$$\text{LRV} = \log \left(\frac{C_f}{C_p} \right) = \log \left(\frac{10^6 \text{ org/L}}{1 \text{ org/L}} \right) = 6.0 \text{ for microorganisms}$$

Because the effluent concentration is below the detection limit, the log rejection is greater than calculated, that is, $\text{LRV} \geq 6.0$ for microorganisms.

2. Calculate log rejection under compromised operation.
 - a. Draw a mass balance diagram for membrane breach:



- b. Write the mass balance equation for microorganisms and solve:

$$\begin{aligned} C_e &= \frac{Q_m C_m + Q_b C_b}{Q_e} = \frac{(1 \text{ org/L})(0.9999) + (10^6 \text{ org/L})(0.0001)}{1} \\ &= 101 \text{ org/L} \end{aligned}$$

- c. Write the mass balance equation for turbidity and solve:

$$C_e = \frac{Q_m C_m + Q_b C_b}{Q_e} = \frac{(0.03 \text{ NTU})(0.9999) + (10 \text{ NTU})(0.0001)}{1} \\ = 0.031 \text{ NTU}$$

- d. Calculate the log rejection value under compromised operation using Eq. 12-4:

$$\text{LRV} = \log \left(\frac{C_p}{C_f} \right) = \log \left(\frac{10^6 \text{ org/L}}{101 \text{ org/L}} \right) = 4.0 \text{ for microorganisms}$$

Comment

As demonstrated in this example, effluent turbidity with a 0.01 percent breach would not be distinguishable from the turbidity with an intact membrane (turbidity = 0.031 NTU versus 0.03 NTU). For microorganisms, however, $C_E = 101 \text{ org/L}$ and $\text{LRV} = 4.0$ compared to $C_E < 1 \text{ org/L}$ and $\text{LRV} > 6.0$ with no breach—a dramatic reduction in rejection capabilities. In other words, a small breach has a substantial effect on microorganism log rejection value but is undetectable by turbidity measurements.

Most membrane filtration systems produce effluent water turbidity between 0.03 and 0.07 NTU, which is near the limit of detection. Indirect integrity monitoring involves establishing a control limit, typically between 0.10 and 0.15 NTU. Readings greater than the control limit for more than 15 min may indicate an integrity problem and would trigger the need to perform a direct integrity test.

DIRECT INTEGRITY TESTS

Direct integrity tests can be either pressure-based or marker-based tests. Pressure-based tests involve applying pressurizing one side of the membrane with air and monitoring the change in air pressure, flow of air, or volume of displaced water, based on the concept that passage of air through a hole in the membrane is much faster than the diffusion of air through the water-filled pores in the membrane. Marker-based tests involve spiking the influent with particles or molecular markers and measuring the concentration of the marker in the effluent, which is similar to the challenge test that manufacturers have performed to get systems approved for use under the LT2ESWTR. Pressure-based integrity tests are most common for commercial membrane filtration systems, and the equipment, instrumentation, and procedures for conducting the test are built into the skid and implemented automatically.

The general principle of pressure-based integrity testing is to isolate a group of modules being tested (typically, one rack), drain the water from one side of the membrane, and pressurize the system with air. In the pressure-hold test, the rate of decay in the pressure is monitored. In a membrane with no breaches, air will diffuse through the water in the membrane pores, and pressure will decay slowly. Air can flow more rapidly through pinholes. The pressure required must be high enough to detect a 3- μm breach but below the bubble point of the membrane material. This pressure range depends on membrane properties and the procedure to determine the appropriate pressure is defined in the *Membrane Filtration Guidance Manual* (U.S. EPA, 2005). The *Guidance Manual* also describes procedures necessary to calculate the sensitivity of the test, which is related to the log removal value that the system would achieve if a portion of the water was flowing through a hole in the membrane untreated and the rest of the water was flowing through membrane pores and achieving complete removal of microorganisms. Acceptable rates of pressure decay vary with the system being monitored according to calculations in the *Membrane Filtration Guidance Manual*. Decay rates of 0.007 to 0.03 bar/min (0.1 to 0.5 psi/min) are typical limits (U.S. EPA, 2001).

Pressure-hold tests have been reported to be capable of detecting one broken fiber in a module containing 20,000 fibers. Entire racks of membranes can be monitored simultaneously, but these tests are sensitive to the size of the system being monitored because the air diffusion through the pore water may exceed the airflow through a breach if the filter area being monitored is too large. Breakage of several fibers in a large pressure-vessel rack, containing 90 modules with 20,000 fibers each, cannot be detected with pressure-hold tests (Landsness, 2001).

The pressure-hold test is the most common method and most manufacturers include all necessary equipment as part of the skid. Less common variations of this test involve measuring the rate of airflow through the membrane at constant pressure or measuring the volume of water displaced by the flowing air.

Current direct integrity testing methods require that the plant be taken out of service, thus reducing the available time for water production. The required frequency of pressure-hold test requirements in state regulations vary from once every 4 h to weekly (Allgeier, 2001).

SONIC TESTING

Sonic testing is a method of identifying leaks in individual pressure vessels. The test involves manually placing an acoustic sensor against a module and listening for changes in the sounds emanating from within the module. During a pressure-hold test, air bubbling through a damaged fiber will make enough noise to be detected by the sensor. The sonic test is highly sensitive when performed by a skilled operator but is subject to background equipment noise. A single broken fiber in a module containing 20,000

fibers can be detected (Landsness, 2001). The sonic test is typically used in conjunction with other integrity monitoring techniques. Effluent monitoring or pressure-hold tests are typically used to identify possible problems in a group of modules, and sonic testing can then be used to confirm the presence of a damaged fiber and identify the individual module containing the breach.

REPAIR OF MODULES

Modules can be repaired by isolating damaged fibers in a process called pinning. The module is typically removed from the rack and placed in a special test casing. The module can be pressurized with air while the ends of the fibers are exposed and covered with water. In this setting, bubbles will emit from the end of the broken fiber, so that the damaged fiber can be identified. A pin is glued into the end of the fiber, effectively taking the individual fiber out of service. The repaired module can then be placed back in service.

Typical operating criteria for membrane filtration facilities are given in Table 12-10. As demonstrated previously, steady-state membrane performance is controlled not by intrinsic membrane properties but by the fouled state of the membrane after it has been in contact with natural water. Fouling depends on interactions between the source water and membrane material, but current understanding of fouling is not sufficient for predicting basic design criteria from measurable water quality parameters and membrane properties. Thus, pilot testing is typically part of the process evaluation procedure. AWWA's *Manual M53* (AWWA, 2005b) describes pilot testing efforts for two different water supplies where the same two membrane products were tested. For one water supply, one membrane performed well and the other fouled rapidly; for the other water supply, the results were exactly reversed. Such information is critical for selecting the most appropriate membrane product and operating conditions for a particular water supply. In addition, nearly all states require pilot testing as part of the permitting process (Herschell, 2007).

Design Criteria Development Based on Pilot Testing

PILOT TESTING

When done properly, pilot testing can be used to demonstrate the effectiveness of innovative technologies or to provide a basis for comparing alternative systems. Pilot testing should incorporate all pretreatment processes that are being considered for the full-scale facility. For most MF and UF membrane studies the following parameters should be studied:

1. Feed and permeate water quality, including pH, turbidity, particle counts, TOC or DOC, UV₂₅₄ absorbance, and other parameters relevant to the specific site
2. Feed water temperature

3. Feed water and permeate flow rates
4. Transmembrane pressure
5. Backwash requirements (frequency, duration, flow rate, pressure)
6. Cleaning requirements (frequency, duration, chemical dosages, procedures)
7. Other constituents of concern in specific applications, such as NOM

PILOT TESTING PERIOD

Pilot testing should be performed over an extended time, ranging from several months to a year, depending on seasonal variations in water quality and temperature. For instance, spring runoff or lake turnover may lead to water quality conditions that cause considerably greater fouling than other times of the year. When complete, pilot testing will provide data on each of the design criteria shown in Table 12-10, with the exception of membrane life. For comparisons between alternatives to be meaningful, data for permeate flux, pressure, and temperature must be combined

Table 12-10

Typical operating characteristics of membrane filtration facilities

Parameter	Units	Range of Typical Values
Permeate flux		
Pressurized systems	L/m ² · h gal/ft ² · d	30–170 18–100
Submerged systems	L/m ² · h gal/ft ² · d	25–75 15–45
Normal transmembrane pressure		
Pressurized systems	bar psi	0.4–1 6–15
Submerged systems	bar psi	0.2–0.4 3–6
Maximum transmembrane pressure		
Pressurized systems	bar psi	2 30
Submerged systems	bar psi	0.5 7.4
Recovery	%	>95
Filter run duration	min	30–90
Backwash duration	min	1–3
Time between chemical cleaning	d	5–180
Duration of chemical cleaning	h	1–6
Membrane life	yr	5–10

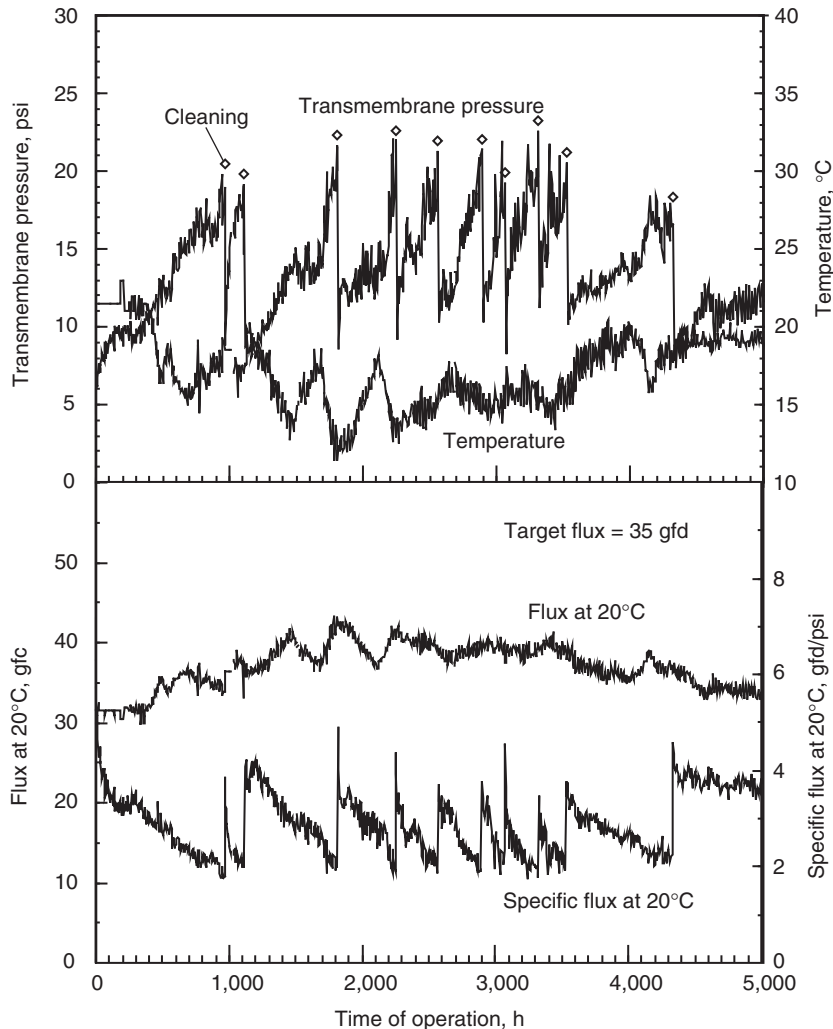


Figure 12-20
Variation in flux, pressure, temperature, and specific flux during pilot testing.

to determine specific flux at a standard temperature. Variations in flux, pressure, temperature, and specific flux from a 7-month pilot study are shown on Fig. 12-20. The effect of chemical cleaning (manifested as lower pressure or higher specific flux) can be observed on Fig. 12-20.

OPERATING PARAMETERS

A basic operating parameter that should be varied during pilot testing is permeate flux. Pilot facilities and full-scale facilities normally operate with constant-flux, rising-pressure conditions. Flux and recovery determine the filtration area required to provide the required capacity, which is a

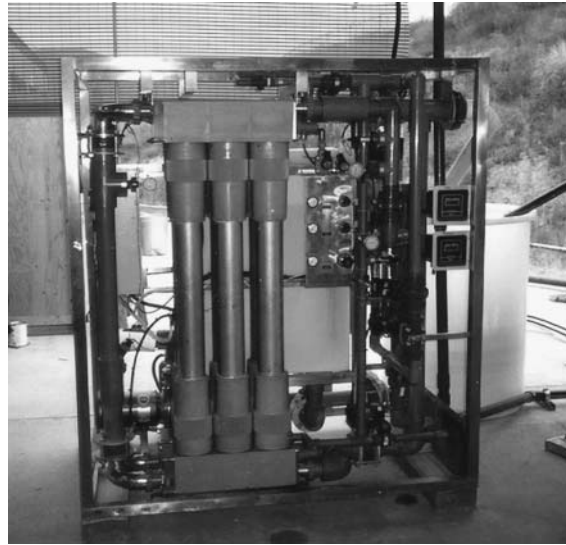
significant factor in determining the capital cost of a facility. Often, pilot testing demonstrates the existence of a critical limit to permeate flux, below which long-term operation is successful and above which pressure rise, backwash frequency, and cleaning frequency are unacceptable. Backwash and cleaning strategies should also be evaluated during the pilot study.

PILOT PLANT UNITS

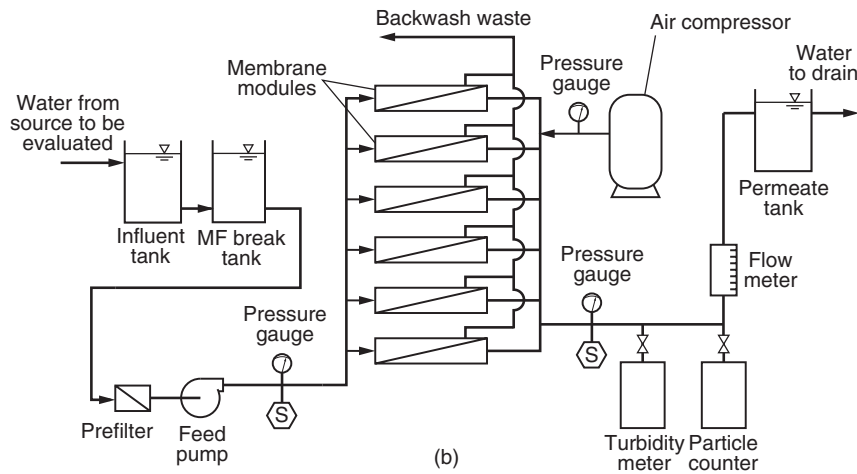
Most membrane manufacturers provide self-contained pilot plant units for use in evaluating performance. A typical skid-mounted membrane filtration pilot plant is shown on Fig. 12-21. Manufacturer-provided pilot plants typically contain all necessary equipment for their membrane system, including membrane modules, a feed tank, a feed pump, a backwash system with either an air compressor or liquid feed backwash pump, a clean-in-place system, permeate storage tank, all piping, valves, and instrumentation, and a programmable logic controller (PLC). The membrane modules are standard full-size modules identical to what would be provided on a full-scale system; the only difference is that a pilot unit typically will contain only 1 to 6 modules whereas a full-scale system may have 50 to 100 modules. Since the modules are identical and are tested with operating conditions that are identical to full-scale operation, the performance and fouling of the membranes can be expected to be very similar to that which would occur at full scale. Pilot plants may be designed with more instrumentation and operational flexibility than full-scale units to allow a range of testing conditions. Manufacturers typically supply specifications for pilot plant systems so they can be operated properly.

PILOT TESTING EXPECTATIONS

Pilot testing establishes the minimum performance requirements that can be accomplished by specific systems. For instance, the flux observed in pilot testing should be achievable in a full-scale facility by the same manufacturer, and the pilot testing can be used to set the minimum performance requirements for each manufacturer that will submit a bid for the project. Pilot testing provides a basis for comparing the effectiveness of alternative systems or new technologies. Individual design parameters, however, should not be compared directly when evaluating alternative systems. For instance, it would be inappropriate to use pilot testing to establish a minimum flux value as a requirement for all systems. A system operating at a low flux may be more cost effective if it operates at a lower pressure with less frequent backwash and clean sequences and has a lower cost per unit of filter area. Physical dimensions, capacity, and filtration area of individual modules, permeate flux, operating pressure, and backwashing and cleaning requirements, taken individually, generally are not a basis for comparing systems. Many parameters are interrelated and can only be compared on the basis of total system performance and cost.



(a)



(b)

Figure 12-21
Typical skid-mounted
pressure-vessel
configuration membrane
filtration pilot plant:
(a) photograph and
(b) schematic of the pilot
plant.

SYSTEM DESIGN FROM PILOT DATA

The data generated during pilot testing can be used to design the full-scale facility. Membrane systems are routinely taken off-line for backwashing, integrity testing, and cleaning, which reduces the time available for permeate production. The percent of time that permeate is produced, or online production factor (AWWA, 2005b), is expressed as

$$\eta = \frac{1440 - t_{bw} - t_{dit} - t_{cip}}{1440} \quad (12-27)$$

where η = online production factor

t_{bw} = time per day for backwashing, min

t_{dit} = time per day for direct integrity testing, min

t_{cip} = time for cleaning, prorated per day, min

The water produced during each filter run can be determined from the flux, membrane area, and run duration:

$$V_f = J a t_f \quad (12-28)$$

where V_f = volume of water filtered per filter run, L

t_f = duration of filter run (excluding backwash time), min

The water consumed during backwashing should be recorded during the pilot testing. With that information and the volume of water filtered from Eq. 12-28, the recovery and the required feed flow rate can be calculated with Eq. 12-26. The amount of time that the system is not producing permeate and the quantity of water that must be used for backwashing both increase the required total membrane area:

$$a_t = \frac{Q_p}{f \eta r} = \frac{Q_f}{f \eta} \quad (12-29)$$

where a_t = total membrane area, m²

r = recovery, unitless

Once the total membrane area is determined, the number of racks and modules per rack can be determined by relating the total required membrane area to the capabilities of the system. An example of the sizing of a full-scale membrane system from pilot data is demonstrated in Example 12-8.

Residual-Handling Requirements

Residual handling from membrane filters is similar in many respects to residual handling from granular filters. However, the reduced or eliminated use of coagulants reduces the generation of sludge and simplifies sludge disposal in some cases. Some utilities discharge the waste washwater to the wastewater collection system and allow the sludge to be handled at the wastewater treatment plant rather than have separate sludge-handling facilities at the water treatment plant. Waste washwater can be clarified and returned to the plant influent or the source water, depending on regulatory constraints. The sludge can be thickened and dewatered similar to sludge from granular filters, and when coagulants are not used, the sludge is generally easier to thicken and dewater. Residual management is discussed further in Chap. 21.

Example 12-8 Determining system size from pilot data

A treatment plant is to be designed to produce 75,700 m³/d (20 mgd) of treated water at 20°C. Pilot testing demonstrates that it can operate effectively at a flux of 65 L/m² · h at 20°C with a 2-min backwash cycle every 45 min and cleaning once per month. The membrane modules have 50 m² of membrane area. The pilot unit contained 3 membrane modules and the full-scale racks can contain up to 100 modules. Backwashes for the pilot unit consumed 300 L of treated water. Cleaning takes 4 h. Regulations require direct integrity testing, which takes 10 min, once per day.

Determine the following: (a) the online production factor, (b) system recovery, (c) feed flow rate, (d) total membrane area, (e) number of racks, and (f) number of modules per rack.

Solution

1. Determine the fraction of time the system is producing permeate.

$$t_{bw} = (2 \text{ min}) \left(\frac{1440 \text{ min/d}}{45 \text{ min}} \right) = 64 \text{ min/d}$$

$$t_{dit} = 10 \text{ min/d}$$

$$t_{cip} = \frac{(4 \text{ h})(60 \text{ min/h})}{30 \text{ d}} = 8 \text{ min/d}$$

$$\eta = \frac{1440 - t_{bw} - t_{dit} - t_{cip}}{1440} = \frac{1440 - 64 - 10 - 8 \text{ min/d}}{1440 \text{ min/d}} = 0.943$$

2. Determine the system recovery. The system recovery is the same for one element as for all elements and can be calculated using Eq. 12-26. For one element that filters for 43 min per cycle (2 min out of every cycle is backwash),

$$V_f = J a t_f = \frac{(65 \text{ L/m}^2 \cdot \text{h})(50.0 \text{ m}^2)(43 \text{ min})}{60 \text{ min/h}} = 2330 \text{ L}$$

$$V_{bw} = \frac{300 \text{ L}}{3 \text{ modules}} = 100 \text{ L}$$

$$r = \frac{V_f - V_{bw}}{V_f} = \frac{2330 \text{ L} - 100 \text{ L}}{2330 \text{ L}} = 0.957$$

3. Calculate required feed flow:

$$Q_f = \frac{Q_p}{r} = \frac{75,700 \text{ m}^3/\text{d}}{0.957} = 79,100 \text{ m}^3/\text{d}$$

4. Calculate the total membrane area required:

$$a = \frac{Q_f}{J_\eta} = \frac{(79,100 \text{ m}^3/\text{d})(10^3 \text{ L/m}^3)}{(65 \text{ L/m}^2 \cdot \text{h})(24 \text{ h/d})(0.943)} = 53,800 \text{ m}^2$$

5. Calculate the total number of modules required:

$$N_{\text{MOD}} = \frac{53,800 \text{ m}^2}{50 \text{ m}^2} = 1076$$

6. Determine the number of racks and modules/rack. Since the racks can accommodate up to 100 modules, at least 11 racks will be required. Dividing the required modules evenly among racks is preferred. In addition, leaving space in the racks is recommended as an inexpensive way to provide flexibility to reduce flux or increase capacity by adding additional modules in the future. Twelve racks are chosen in this example.

$$N_{\text{Racks}} = 12$$

$$N_{\text{MOD/Rack}} = \frac{1076}{12} = 90$$

The system will have 12 racks that each have 90 modules.

Problems and Discussion Topics

- 12-1 Discuss the differences between MF and UF membranes. What impact do these differences have on their use in water treatment?
- 12-2 Discuss the similarities, differences, advantages, and disadvantages between membrane filtration and rapid granular filtration. This is an essay question.
- 12-3 How effective do you think membrane filtration is for each of the following treatment issues? Explain your reasoning.
- Arsenic
 - Anthrax spores
 - Hardness
 - Taste and odor
- 12-4 Calculate rejection and log removal value for the following filtration process (to be selected by instructor). Use the number of significant figures necessary to correctly illustrate the removal being obtained.

	A	B	C	D	E
Influent concentration (#/mL)	10^6	6.85×10^5	7.1×10^5	1.65×10^7	2.8×10^6
Effluent concentration (#/mL)	10	136	0.16	65	96

- 12-5 An inside-out hollow-fiber membrane system is operated with a cross-flow configuration. Each module contains 10,200 fibers that have an inside diameter of 0.9 mm and a length of 1.75 m. Calculate the following for one module:
- Feed flow necessary to achieve a cross-flow velocity of 1 m/s at the entrance to the module.
 - Permeate flow rate if the system maintains an average permeate flux of $80 \text{ L/m}^2 \cdot \text{h}$.
 - Cross-flow velocity at the exit to the module.
 - Ratio of the cross-flow velocity at the entrance of the module to the flow velocity toward the membrane surface. Given the magnitude of this ratio, what effect would you expect cross-flow velocity to have on fouling in cross-flow versus dead-end filtration?
 - Ratio of permeate flow rate to feed flow rate (known as the single-pass recovery). What impact does this ratio have on operational costs in cross-flow versus dead-end filtration?
- 12-6 Hollow-fiber membranes with a membrane area of 23.3 cm^2 were tested in a laboratory and found to have the clean-water flow shown in the table below, at the given temperature and pressure.

	A	B	C	D	E
Flow (mL/min)	4.47	4.22	2.87	6.05	1.22
Temperature ($^{\circ}\text{C}$)	16	22	23	25	22
Pressure (bar)	0.67	0.80	0.71	1.25	0.21

For the data set selected by your professor,

- Calculate the specific flux at 20°C .
 - Calculate the membrane resistance coefficient.
 - Does membrane resistance coefficient depend on the pressure and temperature used for the tests? Why or why not?
- 12-7 The $0.2 \text{ }\mu\text{m}$ polyethersulfone microfiltration membrane shown on Fig. 12-3 was tested in the laboratory and found to have a clean-water flux of $6500 \text{ L/m}^2 \cdot \text{h}$ at 23°C and 0.69 bar. Assume that the flow through a microfiltration membrane can be modeled using the Kozeny equation for flow through porous media (Eq. 11-11 in Chap. 11).

- a. Calculate the specific surface area of the membrane assuming a porosity of 0.6, thickness of 0.10 mm, and Kozeny coefficient of 5.0.
 - b. What would the theoretical grain diameter be if the membrane were composed of spherical granular media with the same specific surface area (see Eq. 11-6)?
 - c. How does the theoretical grain diameter compare to the retention rating for the membrane? Using concepts of particle retention through granular media from Chap. 11, what does this comparison suggest about the mechanisms for particle removal in microfiltration?
 - d. Using the theoretical grain diameter as the characteristic dimension, calculate the Reynolds number for flow through a microfiltration membrane. Is the flow laminar or turbulent?
- 12-8 A membrane plant is operated at a volumetric flux of $75 \text{ L/m}^2 \cdot \text{h}$ at 17°C and 0.85 bar. Calculate the specific flux at 20°C .
- 12-9 Feed water pressure and temperature and permeate flux at a membrane filtration plant are reported on two dates below. For the plant selected by your instructor, calculate the specific flux on each date, and indicate whether fouling has occurred between the first and second dates.

	A	B	C	D	E
Day 1					
Flux ($\text{L/m}^2 \cdot \text{h}$)	72	26	31	86	112
Temperature ($^\circ\text{C}$)	21	17	17	22	19
Pressure (bar)	0.62	0.24	0.24	0.72	0.66
Day 2					
Flux ($\text{L/m}^2 \cdot \text{h}$)	56	26	27	90	120
Temperature ($^\circ\text{C}$)	4	15	10	25	11
Pressure (bar)	0.80	0.29	0.26	0.77	1.05

- 12-10 A new membrane plant is being designed. Pilot testing indicates that the membrane will be able to operate at a specific flux of $120 \text{ L/m}^2 \cdot \text{h} \cdot \text{bar}$ at 20°C . The full-scale plant will operate at 0.8 bar, online production factor of 95 percent, and recovery of 97 percent. Water demand projections predict a summer peak-day demand of $90,000 \text{ m}^3/\text{d}$ and a winter peak-day demand of $60,000 \text{ m}^3/\text{d}$. Historical records indicate that the source water has a minimum temperature of 3°C in winter and 18°C in summer.
- a. Which season will govern the size (membrane area) of the plant?
 - b. What is the required membrane area?

- 12-11 An ultrafiltration membrane with a membrane resistance coefficient of $2.7 \times 10^{12} \text{ m}^{-1}$ is used to filter a 150-mg/L suspension of 0.5- μm latex particles in a laboratory unstirred dead-end filtration cell. The experiment is operated at a constant flux of $120 \text{ L/m}^2 \cdot \text{h}$ and temperature of 20°C , and the membrane has an area of 28.2 cm^2 . Assume that fouling is due to cake formation, the particle density is 1050 kg/m^3 , the cake porosity is 0.38, and the Kozeny coefficient is 5. Neglecting the backmigration of particles due to diffusion, calculate and plot the transmembrane pressure and specific flux over the first 90 min of the filter run.
- 12-12 Show how the cake layer resistance coefficient (Eq. 12-12) can be derived from the Kozeny equation (Eq. 11-11 in Chap. 11) when the membrane feed water is a suspension of monodisperse, well-characterized particles.
- 12-13 A membrane plant containing 1200 m^2 of membrane area operates at a constant permeate flux of $45 \text{ L/m}^2 \cdot \text{h}$ at a temperature of 15°C and pressure of 0.25 bar immediately after backwash. The feed water contains 12 mg/L of suspended solids. After 40 min of operation, the pressure rises to 0.30 bar.
- Assuming that pressure rise between backwashes is due to formation of a cake layer, calculate the specific cake resistance.
 - If permeate flux is increased to $50 \text{ L/m}^2 \cdot \text{h}$, calculate the pressure immediately after backwash and the pressure after 40 min of operation.
- 12-14 Calculate the membrane fouling index for the following data, for the data set specified by your instructor.
- Experimental flat-sheet laboratory filter, membrane area = 30 cm^2 , initial flux = $3560 \text{ L/m}^2 \cdot \text{h}$, test pressure = 0.69 bar, test temperature = 23.9°C .

Time, min	Permeate Volume, mL	Time, min	Permeate Volume, mL
0	0	6	458.3
1	108.8	7	506.8
2	199.8	8	552.1
3	277.4	9	594.1
4	345.0	10	634.1
5	404.2	11	670.8

- Full-scale plant operating at constant permeate flow of 15,000 m^3/day , temperature = 20°C , 5800 m^2 of membrane area, pressure each day as shown below. Use day 0 as the initial flux.

Time, Day	Transmemb. Pressure, Bar	Time, Day	Transmemb. Pressure, Bar	Time, Day	Transmemb. Pressure, Bar
0	0.704				
2	0.712	12	0.747	22	0.786
4	0.721	14	0.754	24	0.794
6	0.726	16	0.765	26	0.801
8	0.735	18	0.770	28	0.812
10	0.740	20	0.777	30	0.812

- c. Data from a 30-min filter run in the middle of a day of laboratory testing of coagulated feed water, membrane area = 23 cm^2 , initial flux = $238 \text{ L/m}^2 \cdot \text{h} \cdot \text{bar}$, test pressure = 2.07 bar, test temperature = 21.5°C .

Time, min	Permeate Volume, mL	Time, min	Permeate Volume, mL	Time, min	Permeate Volume, mL
0	2276.64				
2	2292.62	12	2370.17	22	2444.76
4	2308.41	14	2385.31	24	2459.35
6	2324.05	16	2400.33	26	2473.88
8	2339.53	18	2415.24	28	2488.26
10	2354.92	20	2430.04		

- 12-15 A membrane filtration plant is to be designed using results from a pilot study. Treatment plant requirements and pilot results are given in the table below. For the selected system (to be specified by the instructor), determine (a) the online production factor, (b) system recovery, (c) feed flow rate, (d) total membrane area, (e) number of skids, and (f) number of modules per skid. The pilot system contained two membrane elements that had 45 m^2 of membrane area each. In the full-scale plant, integrity testing will be required by regulations once per day and will take 15 min. Chemical cleaning (CIP) will take 4 h.

	A	B	C	D	E
Design capacity (m^3/d)	56,000	115,000	38,000	76,000	227,000
Memb. area in full-scale modules (m^2)	45	55	45	45	80
Max. modules in skid	80	90	80	80	100

	A	B	C	D	E
Pilot results					
Flux ($\text{L}/\text{m}^2 \cdot \text{h}$)	80	125	40	80	110
Backwash frequency (min)	30	25	25	22	30
Backwash duration (min)	1.5	0.5	1	2	1
Backwash volume (L)	270	100	200	240	240
Cleaning frequency (day)	45	30	60	30	30

References

- Adham, S., Chiu, K., Gramith, K., and Oppenheimer, J. (2005) *Development of a Microfiltration and Ultrafiltration Knowledge Base*, American Water Works Association Research Foundation, Denver, CO.
- Adham, S., Chiu, K., Lehman, G., Howe, K., Marwah, A., Mysore, C., Clouet, J., Do-Quang, Z., and Cagnard, O. (2006) *Optimization of Membrane Treatment for Direct and Clarified Water Filtration*, American Water Works Association Research Foundation, Denver, CO.
- Adham, S. S., Jacangelo, J. G., and Laíné, J.-M. (1995) "Low-Pressure Membranes: Assessing Integrity," *J. AWWA*, **87**, 3, 62–76.
- Allgeier, S. C. (2001) Overview of Regulatory Issues Facing Microfiltration and Ultrafiltration, paper presented at the American Water Works Association Membrane Technology Conference, San Antonio, TX.
- Amy, G. L., and Cho, J. (1999) "Interactions between Natural Organic Matter (NOM) and Membranes: Rejection and Fouling," *Water Sci. Technol.*, **40**, 9, 131–139.
- Anselme, C., Baudin, I., and Chevalier, M. R. (1999) Drinking Water Production by Ultrafiltration and PAC Adsorption, First Year of Operation for a Large Capacity Plant, paper presented at the American Water Works Association Membrane Technology Conference, Long Beach, CA.
- Anselme, C., and Jacobs, E. P. (1996) Ultrafiltration, Chap. 10, in J. Mallevialle, P. E. Odendaal, and M. R. Wiesner (eds.), *Water Treatment Membrane Processes*, McGraw-Hill, New York.
- ASTM (2001a) D5090-90 Standard Practice for Standardizing Ultrafiltration Permeate Flow Performance Data, in *Annual Book of Standards*, Vol. 11.01, American Society for Testing and Materials, Philadelphia, PA.
- ASTM (2001b) D6161-98 Standard Terminology Used for Crossflow Microfiltration, Ultrafiltration, Nanofiltration and Reverse Osmosis Membrane Processes, in *Annual Book of Standards*, Vol. 11.02, American Society for Testing and Materials, Philadelphia, PA.
- ASTM (2001c) E1343-90 Standard Test Method for Molecular Weight Cutoff Evaluation of Flat Sheet Ultrafiltration Membranes, in *Annual Book of Standards*, Vol. 11.01, American Society for Testing and Materials, Philadelphia, PA.
- AWWA (1992) "Membrane Processes in Potable Water Treatment, AWWA Membrane Technology Research Committee Report," *J. AWWA*, **84**, 1, 59–67.

- AWWA (1998) "Membrane Processes, AWWA Membrane Technology Research Committee Report," *J. AWWA*, **90**, 6, 91–105.
- AWWA (2005a) "Committee Report: Recent Advances and Research Needs in Membrane Fouling," *J. AWWA*, **97**, 8, 79–89.
- AWWA (2005b) *Microfiltration and Ultrafiltration Membranes for Drinking Water: Manual of Water Supply Practices M53*, AWWA, Denver, CO.
- AWWA (2008) "Microfiltration and Ultrafiltration Membranes for Drinking Water," *J. AWWA*, **100**, 12, 84–97.
- Bacchin, P., Aimar, P., and Sanchez, V. (1995) "Model for Colloidal Fouling of Membranes," *AIChE J.*, **41**, 2, 368–376.
- Belfort, G., Davis, R. H., and Zydney, A. L. (1994) "Behavior of Suspensions and Macromolecular Solutions in Crossflow Microfiltration," *J. Memb. Sci.*, **96**, 1/2, 1–58.
- Campos, C., Mariñas, B. J., Snoeyink, V. L., Baudin, I., and Laine, J.-M. (2000a) "PAC–Membrane Filtration Process I: Model Development," *J. Environ. Eng.*, **126**, 2, 97–103.
- Campos, C., Mariñas, B. J., Snoeyink, V. L., Baudin, I., and Laine, J.-M. (2000b) "PAC–Membrane Filtration Process II: Model Application," *J. Environ. Eng.*, **126**, 2, 104–111.
- Carroll, T., King, S., Gray, S. R., Bolto, B. A., and Booker, N. A. (2000) "Fouling of Microfiltration Membranes by NOM after Coagulation Treatment," *Water Res.*, **34**, 11, 2861–2868.
- Causserand, C., Nyström, M., and Aimar, P. (1994) "Study of Streaming Potentials of Clean and Fouled Ultrafiltration Membranes," *J. Memb. Sci.*, **88**, 2/3, 211–222.
- Chang, Y.-J., and Benjamin, M. M. (1996) "Iron Oxide Adsorption and UF to Remove NOM and Control Fouling," *J. AWWA*, **88**, 12, 74–88.
- Chang, Y.-J., and Benjamin, M. M. (2003) "Modeling Formation of Natural Organic Matter Fouling Layers on Ultrafiltration Membranes," *J. Environ. Eng.*, **129**, 1, 25–32.
- Chellam, S., and Xu, W. (2006) "Blocking Laws Analysis of Dead-End Constant Flux Microfiltration of Compressible Cakes," *J. Colloid Interface Sci.*, **301**, 1, 248–257.
- Cheryan, M. (1998) *Ultrafiltration and Microfiltration Handbook*, Technomic, Lancaster, PA.
- Cogan, N. G., and Chellam, S. (2009) "Incorporating Pore Blocking, Cake Filtration, and EPS Production in a Model for Constant Pressure Bacterial Fouling During Dead-End Filtration," *J. Memb. Sci.*, **345**, 1–2, 81–89.
- Combe, C., Molis, E., Lucas, P., Riley, R., and Clark, M. M. (1999) "The Effect of CA Membrane Properties on Adsorptive Fouling by Humic Acid," *J. Memb. Sci.*, **154**, 1, 73–87.
- Craun, G. F., Hubbs, S. A., Frost, F., Calderon, R. L., and Via, S. H. (1998) "Waterborne Outbreaks of Cryptosporidiosis," *J. AWWA*, **90**, 9, 81–91.
- Crozes, G., Anselme, C., and Mallevalle, J. (1993) "Effect of Adsorption of Organic Matter on Fouling of Ultrafiltration Membranes," *J. Memb. Sci.*, **84**, 1/2, 61–77.
- Fane, A. G., and Fell, C. J. D. (1987) "A Review of Fouling and Fouling Control in Ultrafiltration," *Desalination*, **62**, 117–136.

- Farahbakhsh, K., Svrcek, C., Guest, R. K., and Smith, D. W. (2004) "A Review of the Impact of Chemical Pretreatment on Low-Pressure Water Treatment Membranes," *J. Env. Eng. Sci.*, **3**, 4, 237–253.
- Ferry, J. D. (1936) "Statistical Evaluation of Sieve Constants in Ultrafiltration," *J. Gen. Physiol.*, **20**, 95–104.
- Field, R. W., Wu, D., Howell, J. A., and Gupta, B. B. (1995) "Critical Flux Concept for Microfiltration Fouling," *J. Memb. Sci.*, **100**, 3, 259–272.
- Glucina, K., Lâiné, J.-M., and Durand-Bourlier, L. (1998) "Assessment of Filtration Mode for the Ultrafiltration Membrane Process," *Desalination*, **118**, 1/3, 205–211.
- Habarou, H., Makdissy, G., Croue, J.-P., Amy, G. L., Buisson, H., and Machinal, C. (2001) Toward an Understanding of NOM Fouling of UF Membranes, paper presented at the American Water Works Association Membrane Technology Conference, San Antonio, TX.
- Hermia, J. (1982) "Constant Pressure Blocking Filtration Laws—Application to Power-Law Non-Newtonian Fluids," *Trans. IChemE*, **60**, 183–187.
- Herschell, J. A. (2007) Survey of State Regulatory Approaches for Approval of Low Pressure Membrane Systems, presentation at the 2007 AWWA Annual Conference, Toronto, ON.
- Ho, C. C., and Zydney, A. L. (2000) "A Combined Pore Blockage and Cake Filtration Model for Protein Fouling During Microfiltration," *Journal of Colloid and Interface Science*, **232**, 2, 389–399.
- Howe, K. J. (2001) Effect of Coagulation Pretreatment on Membrane Filtration Performance, Ph.D. Thesis, University of Illinois at Urbana-Champaign, Urbana, IL.
- Howe, K. J., and Clark, M. M. (2002) "Fouling of Microfiltration and Ultrafiltration Membranes by Natural Waters," *Environ. Sci. Technol.*, **36**, 16, 3571–3576.
- Howe, K. J., and Clark, M. M. (2006) "Effect of Coagulation Pretreatment on Membrane Filtration Performance," *J. AWWA*, **98**, 4, 133–146.
- Howe, K. J., Marwah, A., Chiu, K. P., and Adham, S. S. (2007) "Effect of Membrane Configuration on Bench-Scale MF and UF Fouling Experiments," *Water Res.*, **41**, 17, 3842–3849.
- Huang, H., and O'Melia, C. R. (2008) "Direct-Flow Microfiltration of Aquasols II. On the Role of Colloidal Natural Organic Matter," *J. Memb. Sci.*, **325**, 2, 903–913.
- Huang, H., Spinette, R., and O'Melia, C. R. (2008a) "Direct-Flow Microfiltration of Aquasols I. Impacts of Particle Stabilities and Size," *J. Memb. Sci.*, **314**, 1–2, 90–100.
- Huang, H., Young, T. A., Jacangelo, J. G. (2008b) "Unified Membrane Fouling Index for Low Pressure Membrane Filtration of Natural Waters: Principles and Methodology," *Environ. Sci. Technol.*, **42**, 3, 714–720.
- Huang, H., Schwab, K., Jacangelo, J. G. (2009a) "Pretreatment for Low Pressure Membranes in Water Treatment: A Review," *Environ. Sci. Technol.*, **43**, 9, 3011–3019.
- Huang, H., Young, T. A., Jacangelo, J. G. (2009b) "Novel Approach for the Analysis of Bench-Scale, Low Pressure Membrane Fouling in Water Treatment," *J. Memb. Sci.*, **334**, 1–2, 1–8.

- Ioan, C. E., Aberle, T., and Burchard, W. (2000) "Structure Properties of Dextran 2. Dilute Solution," *Macromolecules*, **33**, 15, 5730–5739.
- Jacangelo, J. G., Adham, S. S., and Laine, J.-M. (1995) "Mechanism of *Cryptosporidium*, *Giardia*, and MS2 Virus Removal by MF and UF," *J. AWWA*, **87**, 9, 107–121.
- Jacangelo, J. G., Laine, J.-M., Carns, K. E., Cummings, E. W., and Mallevialle, J. (1991) "Low-Pressure Membrane Filtration for Removing *Giardia* and Microbial Indicators," *J. AWWA*, **83**, 9, 97–106.
- Jucker, C., and Clark, M. M. (1994) "Adsorption of Aquatic Humic Substances on Hydrophobic Ultrafiltration Membranes," *J. Memb. Sci.*, **97**, 37–52.
- Kim, J. and DiGiano, F. A. (2006) "A Two-Fiber, Bench-Scale Test of Ultrafiltration (UF) for Investigation of Fouling Rates and Characteristics," *J. Memb. Sci.*, **271**, 1–2, 196–204.
- Kim, J. and DiGiano, F. A. (2009) "Fouling Models for Low-Pressure Membrane Systems," *Separation Purification Technol.*, **68**, 3, 293–304.
- Kim, K. J., Fane, A. G., Ben Aim, R., Liu, M. G., Jonsson, G., Tessaro, I. C., Broek, A. P., and Bargeman, D. (1994) "Comparative Study of Techniques Used for Porous Membrane Characterization: Pore Characterization," *J. Memb. Sci.*, **87**, 1/2, 35–46.
- Kim, K. J., Fane, A. G., Fell, C. J. D., and Joy, D. C. (1992) "Fouling Mechanisms of Membranes During Protein Ultrafiltration," *J. Memb. Sci.*, **68**, 1/2, 79–91.
- Koltuniewicz, A. (1992) "Predicting Permeate Flux in Ultrafiltration on the Basis of Surface Renewal Concept," *J. Memb. Sci.*, **68**, 1/2, 107–118.
- Koros, W. J., Ma, Y. H., and Shimidzu, T. (1996) "Terminology for Membranes and Membrane Processes," *J. Memb. Sci.*, **120**, 2, 149–159.
- Kulovaara, M., Metsämuuronen, S., and Nyström, M. (1999) "Effects of Aquatic Humic Substances on a Hydrophobic Ultrafiltration Membrane," *Chemosphere*, **38**, 15, 3485–3496.
- Lahoussine-Turcaud, V., Wiesner, M. R., and Bottero, J.-Y. (1990) "Fouling in Tangential-Flow Ultrafiltration: The Effect of Colloid Size and Coagulation Pretreatment," *J. Memb. Sci.*, **52**, 2, 173–190.
- Laine, J.-M., Clark, M. M., and Mallevialle, J. (1990) "Ultrafiltration of Lake Water: Effect of Pretreatment on the Partitioning of Organics, THMFP, and Flux," *J. AWWA*, **82**, 12, 82–87.
- Laine, J.-M., Hagstrom, J. P., Clark, M. M., and Mallevialle, J. (1989) "Effects of Ultrafiltration Membrane Composition," *J. AWWA*, **81**, 11, 61–67.
- Landsness, L. B. (2001) Accepting MF/UF Technology, Making the Final Cut, paper presented at the American Water Works Association Membrane Technology Conference, San Antonio, TX.
- Lee, Y., and Clark, M. M. (1997) "Numerical Model of Steady-State Permeate Flux During Cross-Flow Ultrafiltration," *Desalination*, **109**, 3, 241–251.
- Lin, C.-F., Huang, Y.-J., and Hao, O. J. (1999) "Ultrafiltration Processes for Removing Humic Substances: Effect of Molecular Weight Fractions and PAC Treatment," *Water Res.*, **33**, 5, 1252–1264.
- Lin, C.-F., Lin, T.-Y., and Hao, O. J. (2000) "Effects of Humic Substance Characteristics on UF Performance," *Water Res.*, **34**, 4, 1097–1106.

- Madaeni, S. S., Fane, A. G., and Grohmann, G. S. (1995) "Virus Removal from Water and Wastewater Using Membranes," *J. Memb. Sci.*, **102**, 65–75.
- Madigan, M. T., Martinko, J. M., and Parker, J. (1997) *Brock Biology of Microorganisms*, Prentice-Hall, Upper Saddle River, NJ.
- Matthiasson, E. (1983) "The Role of Macromolecular Adsorption in Fouling of Ultrafiltration Membranes," *J. Memb. Sci.*, **16**, 23–26.
- McDonogh, R. M., Welsch, K., Fane, A. G., and Fell, C. J. D. (1988) "Flux and Rejection in the Ultrafiltration of Colloids," *Desalination*, **70**, 1/3, 251–264.
- Nyström, M., Pihlajamäki, A., and Ehsani, N. (1994) "Characterization of Ultrafiltration Membranes by Simultaneous Streaming Potential and Flux Measurements," *J. Memb. Sci.*, **87**, 3, 245–256.
- Petry, M., Thonney, D., Roux, J. P., Moulart, P., and Bonnelye, V. (2001) Lausanne: Specific Design for a Challenging Project, paper presented at the American Water Works Association Membrane Technology Conference, San Antonio, TX.
- Ridgway, H. F., and Flemming, H.-C. (1996) Membrane Biofouling, Chap. 6, in J. Mallevialle, P. E. Odendaal, and M. R. Wiesner (eds.), *Water Treatment Membrane Processes*, McGraw-Hill, New York.
- Schäfer, A. I., Fane, A. G., and Waite, T. D. (2001) "Cost Factors and Chemical Pretreatment Effects in the Membrane Filtration of Waters Containing Natural Organic Matter," *Water Res.*, **35**, 6, 1509–1517.
- Schäfer, A. I., Schwicker, U., Fischer, M. M., Fane, A. G., and Waite, T. D. (2000) "Microfiltration of Colloids and Natural Organic Matter," *J. Memb. Sci.*, **171**, 2, 151–172.
- Shorney, H. L., Vernon, W. A., Clune, J., and Bond, R. G. (2001) Performance of MF/UF Membranes with In-Line Ferric-Salt Coagulation for Removal of Arsenic from a Southwest Surface Water, paper presented at the American Water Works Association Membrane Technology Conference, San Antonio, TX.
- Shrive, C. A., DeMarco, J., Metz, D. H., Braghetta, A., and Jacangelo, J. G. (1999) Assessment of Microfiltration for Integration into a Granular Activated Carbon Facility, paper presented at the American Water Works Association Membrane Technology Conference, Long Beach, CA.
- Tarabara, V. V., Hovinga, R. M., and Wiesner, M. R. (2002) "Constant Transmembrane Pressure vs. Constant Permeate Flux: Effect of Particle Size on Crossflow Membrane Filtration," *Environ. Eng. Sci.*, **19**, 2, 343–355.
- Tchobanoglous, G., Burton, F. L. and Stensel, H. D. (2003) *Wastewater Engineering: Treatment and Reuse*, 4th ed., Metcalf and Eddy, McGraw-Hill, New York.
- Treybal, R. E. (1980) *Mass-Transfer Operations*, McGraw-Hill, New York.
- U.S. EPA (1989) "National Primary Drinking Water Regulations: Filtration and Disinfection; Turbidity, *Giardia lamblia*, Viruses, *Legionella*, and Heterotrophic Bacteria; Final Rule," *Fed. Reg.*, **54**, 124, 27486.
- U.S. EPA (1998) "National Primary Drinking Water Regulations: Interim Enhanced Surface Water Treatment: Final Rule," *Fed. Reg.*, **63**, 241, 69478–69521.
- U.S. EPA (2001) *Low-Pressure Membrane Filtration for Pathogen Removal: Application, Implementation, and Regulatory Issues*, U.S. Environmental Protection Agency, Cincinnati, OH.

- U.S. EPA (2005) *Membrane Filtration Guidance Manual*, EPA 815-R-06-009, U.S. Environmental Protection Agency, Cincinnati, OH.
- U.S. EPA (2006) "National Primary Drinking Water Regulations: Long Term 2 Enhanced Surface Water Treatment; Final Rule," *Fed. Reg.*, **71**, 3, 654–786.
- Welsch, K., McDonogh, R. M., Fane, A. G., and Fell, C. J. D. (1995) "Calculation of Limiting Fluxes in the Ultrafiltration of Colloids and Fine Particulates," *J. Memb. Sci.*, **99**, 3, 229–239.
- Wiesner, M. R., and Chellam, S. (1992) "Mass Transport Considerations for Pressure-Driven Membrane Processes," *J. AWWA*, **84**, 1, 88–95.
- Wiesner, M. R., Clark, M. M., and Mallevialle, J. (1989) "Membrane Filtration of Coagulated Suspensions," *J. Environ. Eng.*, **115**, 1, 20–40.
- Yuan, W., Kocic, A., and Zydney, A. L. (2002) "Analysis of Humic Acid Fouling During Microfiltration Using a Pore Blockage-Cake Filtration Model," *J. Memb. Sci.*, **198**, 1, 51–62.
- Yuan, W., and Zydney, A. L. (1999) "Humic Acid Fouling During Microfiltration," *J. Memb. Sci.*, **157**, 1, 1–12.
- Yuan, W., and Zydney, A. L. (2000) "Humic Acid Fouling During Ultrafiltration," *Environ. Sci. Technol.*, **34**, 23, 5043–5050.
- Zydney, A. L., and Colton, C. K. (1986) "A Concentration Polarization Model for the Filtration Flux in Cross-Flow Microfiltration of Particulate Suspensions," *Chem. Eng. Commun.*, **47**, 1/3, 1–21.