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INTEGRATED MEMBRANE SYSTEMS

Introduction

The time has arrived for users of Reverse Osmosis (RO) and Nanofilter (NF) systems to re-evaluate the cost and operating benefits of Microfiltration (MF) and Ultrafiltration (UF) as RO pretreatment. This is particularly true when confronted with having to process a high-fouling feed water source such as a surface water, a wastewater, or an open-intake seawater. A system designed with a MF/UF membrane system in front of a RO system has been referred to as an integrated membrane system. The IMS™ design approach to water treatment systems has some significant advantages over RO systems designed with conventional pretreatment:

- MF/UF filtrate quality is better. The colloidal fouling load to the RO is reduced with significantly lower SDI s (Silt Density Index) and turbidity.
- MF/UF filtrate quality remains much more constant since it is an absolute membrane barrier. This is true even for those surface or wastewater sources afflicted by rapidly fluctuating quality.
- RO cleaning frequencies due to colloidal fouling are reduced.
- MF/UF systems can require less time and be easier to operate than some conventional filtration processes, particularly those prone to system upsets.
- MF/UF concentrated waste streams are easier to dispose of relative to chemically-enhanced conventional pretreatment processes.
- Floor space requirement is less, sometimes by a factor of 5 for large systems {1}.
- Future expandability is easier to design in.
- Operating costs are competitive, and in some schemes, less.
- Capital costs are competitive, and in some schemes, less.

Yesterday s Perspective

The interest in using MF or UF as pretreatment to RO for processing high-fouling feed waters (e.g. surface waters and waste waters) dates back to the 1980 s. The MF/UF design was generally dismissed as a commercial alternative to conventional pretreatment for a number of reasons. The primary reasons were:

- **Capital Costs:** The MF/UF capital costs were too high for treatment of surface waters. A MF/UF sub-system had a capital cost that was close to 100% of the capital cost of the RO system. Capital costs for conventional pretreatment systems (e.g. clarifiers, gravity filters, lime-softeners, multimedia filters, carbon filters) were 20 to 50% of the capital cost of the RO system.
- **Operating Costs:** The MF/UF operating cost savings versus conventional pretreatment was difficult to substantiate and guaranty without the benefits of empirical data collection from a pilot plant.
- **Pilot Plant Required:** Small system and industrial users not bound by government regulations tend to refrain from the use of pilot plants for a number of reasons (e.g. cost and time limitations).
- **Performance Guaranties:** Historically, the Original Equipment Manufacturer (OEM) has borne the responsibility of the total system performance warranty when a bid is issued. Without the benefits of pilot plant data to substantiate the merits of a MF/UF system in a competitive bid situation, the default pretreatment system was comprised of conventional pretreatment components.
- **Aversion to Unproven Technology:** MF/UF was essentially new technology . Concerns included the risk of buying a system that was considered a serial number one , where there were only a few MF/UF membrane suppliers to choose from, or where the MF/UF membrane design was proprietary in nature and would limit future replacements or service to only one firm.

Today s Perspective

The use of MF/UF as a water filtration process has exploded over the last couple of years in the municipal market place. There are several hundreds of MF/UF systems in operation for municipal drinking water systems throughout the world, with capacities that are evenly distributed between MF and UF exceeding 200 mgd (million gallons per day) total {2}. The proliferation of MF/UF systems in the municipal market place is the result of increasingly stringent water quality requirements being mandated for potable water derived from surface water sources. In the United States, MF/UF technology has been readily accepted to achieve potable drinking water quality in terms of controlling pathogenic microorganisms and potentially carcinogenic Disinfection By-Products (DBP). These microbiological and DBP guidelines were generated by federal government actions by establishing present and future regulations as set in the Surface Water Treatment Rule (SWTR), the Enhanced Surface Water Treatment Rules (ESWTR), and the Disinfectant/Disinfection By-Products Rule (D/DBPR).

Significant benefits from the expanded use of MF/UF, both commercially and technically, are starting to spill over to other segments of the water treatment market. The vast size of the municipal market place has resulted in a resurgence of private investment into the enhancement of MF/UF technology. This level of MF/UF investment is reminiscent of the investment in new products and system developments spurred by the power industry in the 1970 s and the electronics industry in the 1980 s. The benefits resulting from the infusion of this investment money are numerous:

- Polymer chemists have developed improved MF/UF membranes in both capillary and spiral-wound configurations.
- MF/UF membrane manufacturers and OEM s have developed improved operational techniques that reduce the rate of fouling and chemical cleaning frequency to acceptable intervals.
- There are at least eight major suppliers of MF/UF membranes and systems.

- The large volume of actual MF/UF membranes sales have reduced the unit cost of these membranes to make MF/UF system costs competitive with conventional pretreatment.
- The requirements of the municipal market place have allowed for extensive on-site pilot testing to be conducted under well-defined test conditions and under the supervision of competent engineering firms and/or consultants. Extensive evaluation of the pilot data has enhanced the ability of membrane suppliers to better project expected operating parameters for varying feed water conditions, cleaning frequencies and filtrate quality.

MF/UF as RO Pretreatment

The activities in the municipal market in the development of MF/UF technology as a commercially viable filtration process has been primarily focused on producing filtrate water suitable for drinking water purposes. The next market focus for the MF/UF process is the development of integrated membrane systems (also known as IMS™), where MF/UF is used as pretreatment to RO. The demand for MF/UF systems as pretreatment to RO will be accentuated by the increasing scarcity of low-fouling feed water sources (e.g. well water) and the need to treat more difficult feed water sources (e.g. surface waters, industrial waste waters and municipal sewer waters).

MF/UF Membrane Characteristics

The available MF membranes have typical pore sizes of 0.1 to 0.35 micron. UF membranes designed for use as pretreatment to RO have nominal molecular weight cutoffs of 20,000 to 750,000 Dalton (0.002 to 0.05 microns).

Typical operational transmembrane pressures (TMP) range from 3 to 30 psi. The transmembrane pressure is defined as the pressure required to force water through the membrane and is the feed pressure less the filtrate pressure. Filtrate is the industry name for the MF/UF product water. TMP requirements will be higher for tighter membranes with smaller pore sizes, with higher flux rates, colder water temperatures, and most significantly when fouling occurs.

MF/UF membranes can be developed from inorganic materials (e.g. ceramic) or from organic polymers. Common polymeric membrane materials include polyolefin, polyether sulfone, polysulfone, polypropylene, cellulosic and other proprietary formulations. Most membrane materials typically have a wide pH tolerance range to accommodate for low and high pH cleaning chemicals. Most membranes also have a free chlorine tolerance that allows for periodic or continuous sanitization. Maximum operating temperatures for the polymeric membranes is in the area of 40⁰ C where the ceramic is much higher.

MF/UF membranes can come in a number of configurations: spiral wound flat sheet, hollow fiber, tubular and plate-and-frame. The prevalent configurations for pretreatment to RO are hollow fiber and spiral wound based on the combined attributes of capital cost, energy efficiency, fouling resistance and the ability to restore flux by a combination of flushing and chemical cleaning.

The service operation of hollow fiber MF/UF membranes can be performed two ways. The first approach is to use a pressurized feed that forces filtrate through the membrane. The second approach is to use a vacuum draw of the filtrate through the membrane.

MF/UF Operating Characteristics

MF/UF membranes are operated in two different service modes: dead-end flow and cross-flow. The dead-end flow mode of operation (also known as direct-flow) is similar to that of a cartridge filter where there is only a feed flow and filtrate flow (no concentrate flow). The dead-end flow approach typically allows for optimal recovery of feed water in the 95 to 98% range, but is typically limited to

feed streams of low suspended solids (e.g. < 10 NTU turbidity). The cross-flow mode of operation is similar to that of a RO where there is feed flow, filtrate flow, and concentrate flow. The cross-flow mode is typically used for feed waters with higher suspended solids (e.g. 10 to 100 NTU turbidity). The cross-flow mode of operation typically results in 90 to 95% recovery of the feed water.

In some cases, feed water recovery for MF/UF systems can be improved up to 99% by collecting and processing further the concentrated feed water and/or the water used for periodic backwashes. This secondary processing step can be accomplished by using conventional solid-settling systems or by the use of another MF/UF system.

A major reason for the re-emergence of MF/UF technology has been improvements in the control of fouling during the service operation and by improved foulant removal techniques. The basic premise to remember here is that a MF/UF system is specifically designed to effectively remove foulants and to be cleaned. RO systems are designed to remove salts, not foulants, and are not specifically designed to be fouled and cleaned frequently.

A variety of membrane and element designs and operational techniques have been developed by different MF/UF suppliers to control the rate of fouling during the service operation and minimize the frequency of full-scale off-line chemical cleanings. The common objective of all these designs, however, is to utilize a high velocity forward-flush or back-flush of the membrane to remove the foulant from the membrane surface and feed path. The back-flushing of the foulant typically uses filtrate, though it can be enhanced by use of air or a chemical (e.g. chlorine). Duration of the flushing sequence is normally held to less than a couple of minutes with the flushing frequency (normally every 15-60 minutes) set to obtain a long-term stabilized service.

If pretreatment is required by a MF/UF system, it is coarse filtration by the use of strainers rated at 100 to 150 micron. Occasionally the use of a coagulant aid like a ferrous salt is also considered to optimize suspended solids removal.

MF/UF membranes are typically operated in a flux range of 36 to 110 gfd (gallons per square foot per day) (60 to 183 l/m²/hr). Lower fluxes are used for feed waters with high suspended solids and fouling potential (e.g. tertiary waste waters) and higher fluxes for with lower suspended solids loading (e.g. 70 gfd for surface water sources). An important aspect in application development work will be the ability to correlate essential feed water fouling parameters (e.g. turbidity, suspended solids, etc.) to a design flux rate without the need for pilot work.

MF/UF filtrate quality in terms of turbidity and SDI (Silt Density Index) is significantly better and more consistent than conventionally pretreated water. Typical turbidity values for MF/UF filtrate is 0.04 to 0.1 NTU and does not increase with increases in feed water turbidity. Typical turbidity values for well-operated conventional pretreatment effluent on high turbidity waters is 0.2 to 1.0 NTU. Typical SDI values for MF/UF filtrate is 0.3 to 2. Typical SDI values for well-operated conventional pretreatment effluent on feed water sources with unmeasurable SDI is 2 to 6. Lower SDI values result in reduced fouling of the RO due to colloidal material deposition.

Improved RO Economics with MF/UF

The consistently high quality of MF/UF filtrate in terms of turbidity and SDI will allow for a higher design flux for downstream RO. A design flux of 12 to 20 gfd can be considered for RO systems with MF/UF pretreatment. Typical design fluxes for waste waters with conventional pretreatment is 8 to 12 gfd. Typical design fluxes for surface waters with conventional pretreatment is 10-15 gfd. An increased flux rate has the advantages of lower capital costs based on the need for fewer RO elements, pressure vessels and associated piping. Increased flux rates also have the advantage of higher RO permeate quality with 30 to 50% less salt passage.

LFC: A New RO Membrane for IMS™ Systems

A critical consideration in increasing the design RO flux rate when MF/UF pretreatment is used is a concern for organic fouling of the RO membrane. Reduction of organics is minimal with MF/UF at 20 to 30% maximum. Organic fouling of conventional negatively charged composite polyamide RO membranes results in dramatically higher feed pressure requirements. Field observations indicate that the use of low fouling membranes are important in the generation of a long-term stabilized flux and feed pressure requirement. The low fouling membranes have a neutral surface charge and are more hydrophilic in nature, which minimizes the absorption of charged, hydrophobic organic foulants to the membrane surface and is more efficiently removed by chemical cleaning {3}.

The LFC membrane offers significant advantages in long term and recoverable flux stability when compared to conventional PA membranes when the foulant is an organic. This capability makes removal of organics in the pretreatment less of an issue than in the past. Though no definitive level of acceptable organic content in a RO feed water exists, an alert level for the designer to consider LFC over a PA membrane could be considered to be:

- 3 ppm TOC (Total Organic Carbon as C)
- 6 ppm BOD (Biological Oxygen Demand as O₂)
- 8 ppm COD (Chemical Oxygen Demand as O₂).

A Lower Fouling RO Membrane

The best RO element to reduce the effects of organic fouling in an IMS™ system is one that has a surface that is neutrally charged and hydrophilic in nature to minimize the attachment of charged foulants. It can also be used with a biocide to control biological fouling and has a high surface area to decrease flux and increase cross-flow velocity. In the past, the cellulose acetate (CA) membrane with its neutral surface charge and a resistance to biocidal chlorine up to 1 ppm or 26,280 ppm-hours, exhibited the best fouling resistance for difficult water applications. However, the CA membrane had pH limitations, higher feed pressure requirements, and higher salt passage when compared to the popular negatively charged composite polyamide (PA) membranes. Today, a new generation of Low Fouling Composite polyamide (LFC) membrane is available. The LFC membrane has the unique advantages of equivalent rejection and feed pressure requirements of a durable PA membrane and the neutral surface charge of the CA membrane (see Figure 1). The LFC membrane, being a polyamide membrane material itself, has a chlorine tolerance level similar to conventional PA membranes of approximately 1,000 ppm-hours.

Figure 1: Comparison of RO Membranes

Membrane Type	LFC	PA	CA
Membrane polymer	Polyamide	Polyamide	Cellulose acetate
Surface charge	Neutral	Negative	Neutral
NaCl rejection	99%	99 to 99.7%	95 to 98%
Organic rejection	Similar	Similar	Lower
Test Pressure	225 psi	225 psi	420 psi
Specific flux (gfd per 100 psi of NDP)	13	13	5 to 6
Feed pH range	3 to 10	3 to 10	4 to 6
Temperature limit	113 F (45 C)	113 F (45 C)	104 F (40 C)
Chlorine tolerance	1000 ppm-hr	1000 ppm-hr	26,280 ppm-hr
Hydrophilicity	47° angle	62° angle	50° angle

Case Study of an IMS™ System

Figure 2 summarizes a water reclamation case study where the RO permeate specific flux (in gfd per psi) varied based on a combination of membrane types and pretreatment schemes. It is evident that the use of UF pretreatment for the removal of colloidal and suspended foulants, combined with the use of LFC1 (a low fouling composite polyamide membrane) which reduces the effect of organic fouling, resulted in the highest stabilized specific flux, lowest flux decline from initial to stabilized operation, and the lowest stabilized feed pressure requirements. The neutrally charged and hydrophilic CA (cellulose acetate) membrane has an organic fouling resistance similar to LFC1, but has a high flux decline due to colloidal fouling when conventional pretreatment is used and is burdened by a high feed pressure requirement due to a low initial specific flux. The ESPA1 test membrane is a conventional low-pressure, negatively charged composite polyamide membrane with lower hydrophilicity than CA or LFC1 membranes and the lowest initial feed pressure requirement due to the highest initial specific flux. The ESPA1 membrane has significant flux decline due to organic fouling even when UF pretreatment is used. The most significant flux decline scenario is for ESPA1 due to the combined effects of organic fouling by absorption to the membrane and when conventional pretreatment is used that aggravates colloidal fouling {4}.

Figure 2: Performance of RO Membranes with Conventional or UF Pretreatment

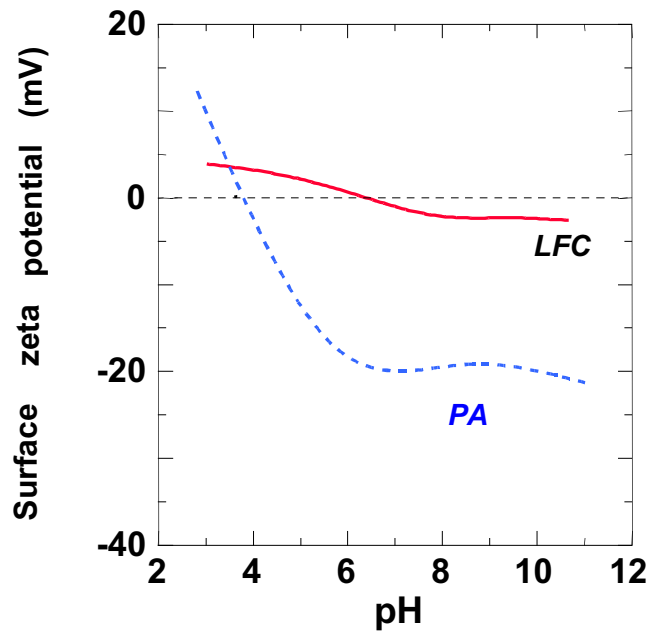
Membrane Type	CA	ESPA1	ESPA1	LFC1
Pretreatment	Conventional	Conventional	UF	UF
Specific flux, initial	0.07 gfd/psi	0.24 gfd/psi	0.24 gfd/psi	0.17 gfd/psi
Specific flux, stabilized	0.04 gfd/psi	0.04 gfd/psi	0.10 gfd/psi	0.15 gfd/psi
Flux decline	40%	85%	60%	12%
Stabilized Feed Pressure at 10 gfd	300-350 psi	300-350 psi	140-180 psi	100-150 psi

LFC Membrane Chemistry

The reduced fouling capability of the LFC membrane is the result of new membrane chemistry. The membrane is permanently modified during the casting process to produce a neutral surface charge and a more hydrophilic membrane surface. The combination of a neutral surface charge and increased hydrophilicity minimizes the adsorption of hydrophobic organic foulants (e.g. humic matter) onto the membrane surface. Flux degradation due to the build up of foulants that are organic in nature, hydrophobic metal gels (e.g. iron), and charged colloidal material is minimized. Just as important for long term operational stability is the enhanced ability to remove foulants and restore the system flux with periodic flushings and/or chemical cleanings.

The LFC membrane can operate with either acidic or basic feed waters and still maintain its neutral surface charge. The surface charge of three membranes over a pH range of 3 to 10 were analyzed quantitatively by measuring the Zeta Potential using Laser-Doppler electrophoresis equipment. The LFC membrane maintained a relatively neutral surface charge of -3 to $+5$ millivolts (mV). The conventional PA membrane has a negative charge of -5 to -21 mV between a pH of 4 to 10 due to the disassociation of the carboxylic groups in the polyamide chain. Interestingly, the PA membrane at a pH less than 4 actually exhibits a positive charge due to the disassociation state of the amine groups in the polyamide chain. [5] (See Figure 3).

Figure 3: pH Effects on Membrane Surface Charge

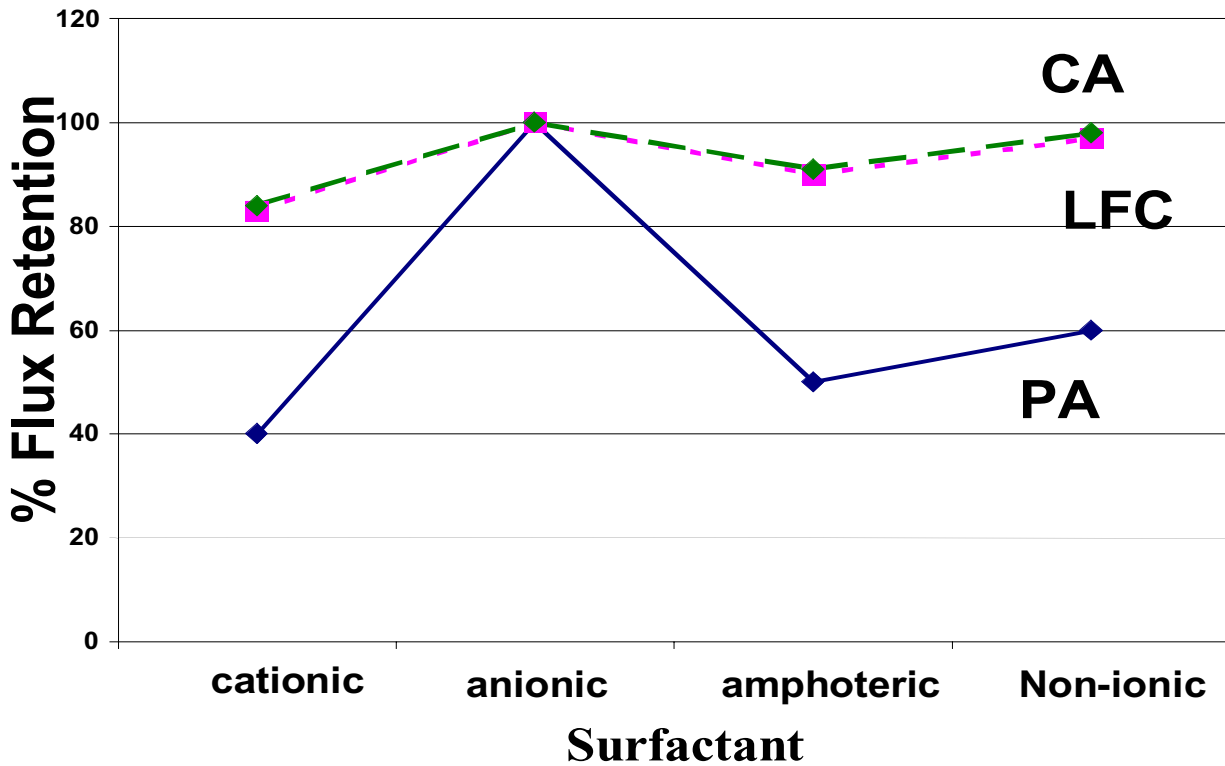


The LFC membrane, in the same fashion as the CA membrane, can operate with foulants of varying charges with minimal or no loss of flux. The conventional negatively charged (anionic) PA membranes are notorious for a dramatic irreversible loss of flux when exposed to cationic (positively charged), amphoteric (either positively or negatively charged based on pH conditions) and neutral polyelectrolytes which are so popular as potential pretreatment and cleaning chemicals (e.g. coagulants, flocculants, surfactants, detergents). Figure 4 depicts the excellent flux stability of the LFC membrane when challenged with cationic, anionic, amphoteric and neutral surfactants. [5]

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- 3 ppm TOC (Total Organic Carbon as C)
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Figure 4: Membrane Exposure to Surfactants



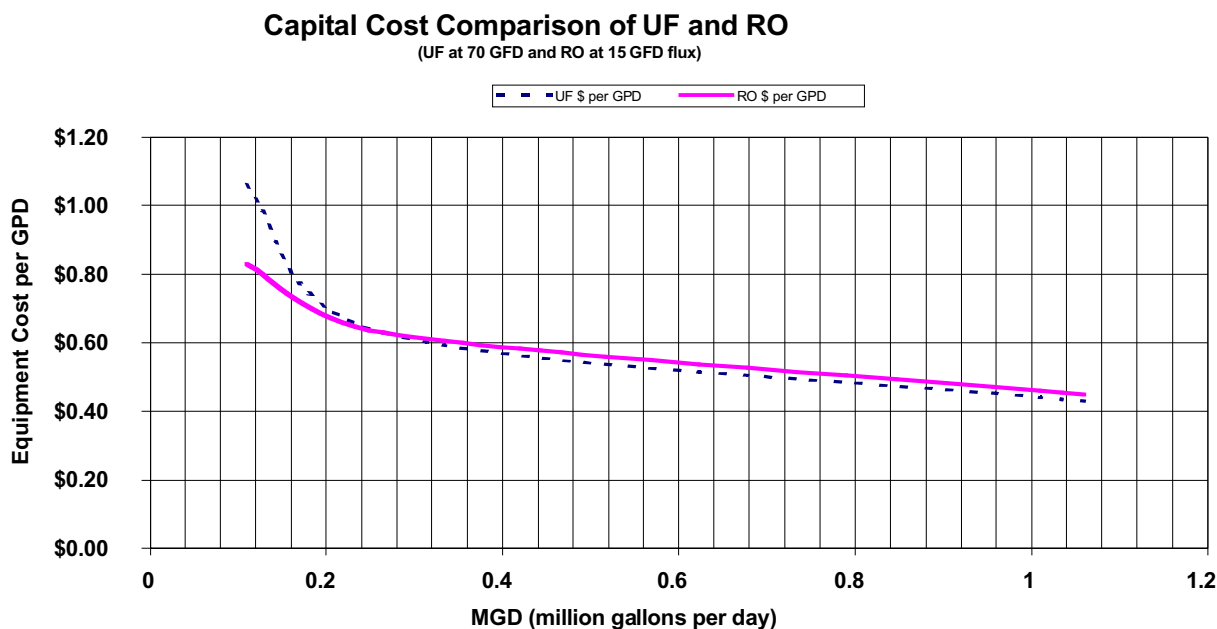
MF/UF Costs

The capital and operating costs of a MF/UF system can vary. Primarily the quality of the feed water to be treated and its impact on the design flux of the system affect capital cost variability. A higher fouling feed water which requires a lower design flux will result in higher capital costs due to the need for more MF/UF modules and a larger backwash and chemical cleaning system. Operating cost variability is primarily impacted by system design flux, recovery, and fouling potential of the feed water. A higher operating cost results when feed pressures are higher, feed flows are higher due to lower recovery, backwash and cleaning frequency is increased, and more modules have to be cleaned.

Capital cost estimates for MF/UF can range from about \$1.00 per gpd of filtrate for a 100,000 gpd (70 gpm) system, to \$0.50 per gpd of filtrate for a 700,000 gpd (486 gpm) system, to \$0.40 per gpd of filtrate for a 5,000,000 gpd (3472 gpm) system. The higher capital costs for smaller systems can be attributed to the cost impact of the auxiliary backwash and chemical cleaning skids. The capital costs noted above are based on a design system flux of 70 gfd. Figure 5 is a graphical comparison of capital costs of a MF/UF system at 70 gfd and a RO system at 15 gfd (with the cost of a cleaning system included for both). Increasing the flux rate from 70 to 100 gfd for a large MF/UF system can reduce system cost up to 30% and decreasing the flux rate from 70 to 40 gfd can increase system cost up to 50%

Operating cost estimates can range from \$0.10 to \$0.40 per 1000 gallons of filtrate, depending on how the operating cost is calculated. The lower value includes the cost of energy at \$0.05 per kilowatt-hr, cleaning chemicals and membrane replacement every 6 years.

Figure 5: Capital Cost Comparisons of UF and RO



Conclusion

The intent of this paper was to re-introduce the users of RO to the concept of using a MF/UF system as a technically and cost-effective pretreatment option. This is particularly true for high fouling feed water sources such as surface water, wastewater and water-for-reuse. The MF/UF system is particularly suited to reduce the rate of colloidal fouling of a RO system, but may not necessarily reduce the rate of organic fouling or biological fouling. The Integrated Membrane Solutions (or IMS™) design approach would recommend the use of a low fouling composite polyamide RO membrane with a resistance to organic fouling. This membrane is neutrally charged and hydrophilic in nature, which results in the reduced absorption of, charged, hydrophobic organic material that would pass through a MF/UF system. Biological fouling control if required may consist of the use of a periodic or continuous introduction of a biocide. A non-oxidizing biocide to consider is chloramine at dosing rate of 1 ppm (and up to 4-6 ppm for more difficult situations). A non-chemical consideration for biological fouling control is the use of an ultraviolet sterilizer sized for a minimum of 30,000 to 35,000 microwatt-seconds on the feed to the RO. An IMS™ system design approach that selectively addresses the range of RO foulants will allow the effective use of RO for the treatment of difficult waters.

References

1. Dawes, T.M., et.al., Meeting the Demand for Potable Water in Orange County in the 21st Century: The Role of Membrane Processes. AWWA Membrane Conference Proceedings March, 1999.
2. Bergman, R.A., Overview of Membrane Filtration for Municipal Water Treatment. AWWA Membrane Conference Proceedings March, 1999.
3. Hydranautics LFC1 Membrane Application Update, August 1999.
4. Wilf, M., Alt S., Reduction of Membrane Fouling and Improving Elements Integrity in Municipal Wastewater Reclamation. International Desalination Association Proceedings September, 1999.
5. R. Gerard, H. Hachisuka, M. Hirose. New Membrane Developments Expanding the Horizon for Application of Reverse Osmosis Technology, proceedings of 1997 Aquatech 98 in Amsterdam, the Netherlands.