MEMBRANES

MEMBRANE PROCESS OFFERS IMPROVED WATER RECOVERY

In the United States, technical and economic challenges associated with the disposal of high-salinity effluents and reverse osmosis (RO) membrane plant reject water continues to undermine the viability of commercial and industrial projects in arid southern states, which suffer from depleting water reserves (1). As the dissolved ions and other constituents become more concentrated, the reduced volume of RO reject streams become more difficult to dispose of in an economical manner since these rejects cannot be discharged to receiving waters, or comply with local municipality permit requirements.

Dependence on costly and energy-intensive brine concentrators and crystallizers to dispose of these rejects (i.e., in some cases up to 25% to 30% of the influent raw water volume) and to achieve zero liquid discharge (ZLD) is technically challenging, and in most cases not economically viable (2). There is a need to address water shortages, maximize fresh water utilization, and minimize the volume of waste (reject) streams to enable their economical disposal via evaporation ponds or low-cost spray dryers.

Limitations of Existing Processes

While RO and other membrane desalination processes, including electrodeionization (EDI) achieve effective water purification, the extent of purified water

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recovery is in most cases limited by the concentration of scale-precursors and the concentration of fouling and colloidal material in the raw water. These compounds deposit on the membrane surface and undermine the rate of permeation (i.e., the flux) and result in premature cleaning and subsequent failure, and membrane replacement. Calcium and magnesium are common scale precursors, which form temporary (or permanent) insoluble hardness compounds as they concentrate over the RO membrane surface.

While temporary, calcium and magnesium hardness (i.e., due to carbonate) can be addressed by acidifying the water, permanent (sulfate) hardness is relatively independent of the pH. Silica also has a limited solubility, which increases by increasing the pH (e.g., by addition of caustic soda). Unfortunately, increasing the pH converts the relatively soluble calcium bicarbonate to the rather insoluble calcium carbonate, resulting in scale deposition on the membrane surface.

To complicate the matter further, most water sources from aquifers contain a mixture of calcium, carbonate alkalinity, sulfate and silica, thus undermining the pretreatment system's ability to achieve high RO membrane system recoveries (i.e., greater than 80%) by simple pre-conditioning with acid and/or antiscalant. Typical low-pressure, first-stage RO membrane systems can achieve recoveries in the range 60% to 80%, depending on the influent composition and pretreatment method. The treatment of 20% to 40% reject volume using thermal (evaporative) processes, even with energy recycling as in multi-stage flash distillation systems, or vapor recompression evaporators and crystallizers, is very costly both in terms of capital and energy costs.

Alternatively, there are processes where the reject stream from an existing RO system is processed further using a second, high-pressure RO stage, after additional conditioning (e.g., by pH adjustment and/or antiscalant addition), or by using inter-stage chemical softening. This approach will enable additional water recovery, perhaps up to 85% to 90%, depending on the water quality and inter-stage softening method used. However, this method is not cost-effective since it treats a relatively large first-stage RO concentrate stream. The chemical precipitation equilibrium will leave a significant concentration of sparingly soluble calcium compounds and silica that will limit the overall re-
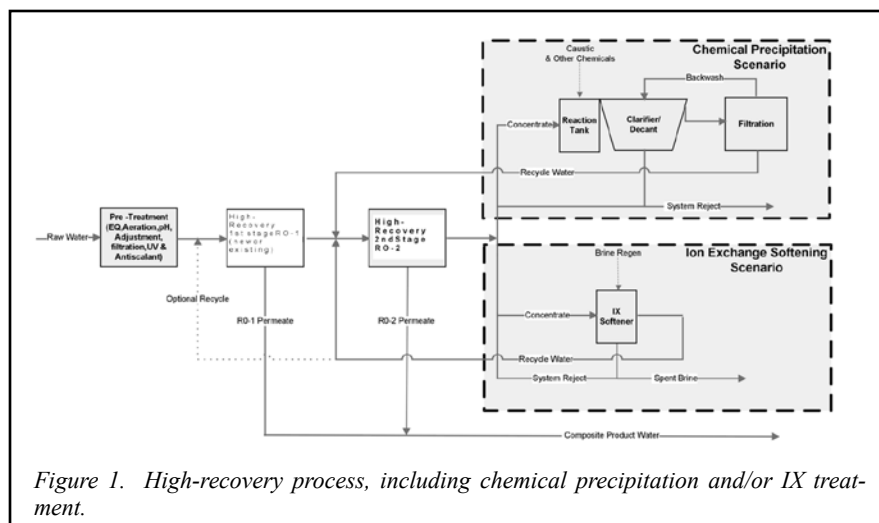

Figure 1. High-recovery process, including chemical precipitation and/or IX treatment.

covery due to precipitation upon further concentration over the second-stage RO membranes.

In order to alleviate scaling limitations because of silica, other patented processes teach effective pretreatment that may include lime softening, followed by complete removal of calcium and magnesium hardness ions using weakly acidic ion exchange (WAC-IX) resins. Additional pretreatment steps include acidification and/or degasification to remove carbonate alkalinity, followed by the addition of caustic soda to raise the pH to greater than 10, thereby increasing the solubility of silica and preventing its precipitation on the surface of the RO membranes. The high pH also mitigates biological fouling, increases the ionic separation efficiencies and improves the RO membrane flux.

Although the above method has been reported to achieve recoveries of greater than 90% and is ideally suited to "high silica" influents, it includes the costly lime softening and weakly acidic ion exchange (IX) treatment of the entire influent water volume in order to achieve complete calcium and magnesium ion removal in the first pretreatment step. Presence of even low-parts-per-million (ppm) concentrations of calcium will result in scale formation on the RO membranes at the high operating pH. This process also requires the addition of substantial quantities of caustic soda to raise the pH of the full influent stream to a pH greater than 10.

These processes are successful in treating power station blowdown effluents having moderate hardness and silica concentrations. However, for influent streams containing relatively high silica concentrations (e.g., 50 to 100 milligrams per liter [mg/L]), the maximum allowable operating pH of approximately 10 will increase the silica solubility, but only to the extent of achieving 80% to 90% permeate recoveries, in addition to the significant WAC-IX spent regenerant volume produced.

Increasing the pH above 10, while further increasing the silica solubility, will also add more sodium ions to the water, increase the total dissolved solids (TDS) and osmotic pressure of the influent, and undermine the RO permeate recovery. Furthermore, operation of RO membranes at elevated pH greater than 10 could limit their useful life since the chemical stability of the materials used in the manufacture of RO elements will be challenged at this pH range. So is it possible to achieve much higher water recoveries (i.e., in the range 95% to 99%) in an economical manner without undermining the service life of the RO membranes, and regardless of the calcium hardness and silica concentrations?

High Recovery Process

A membrane system[a] has been developed (6) that achieves very high permeate recoveries by addressing the following two fundamental problems:

Mitigation of surface fouling. To address the fouling problem, raw water or industrial effluents must first be pretreated using effective low-cost filtration and fouling mitigation principles. These pretreatment methods are based on the simple concept of aeration to address the biological fouling and iron and manganese fouling problems, followed by charge neutralization and reversal to mitigate colloidal fouling, if required. These patented processes (3) apply to a wide range of surface water, groundwater, and organically (and inorganically) contaminated wastewater streams, enabling implementation of low-cost RO solutions to challenging water purification and wastewater treatment applications.

Control of scale formation. Sparingly soluble compounds include calcium carbonate, silica, barium sulfate, and calcium fluoride. As these inorganic compounds concentrate because of permeation of pure water through RO membranes, they will eventually become saturated and deposit on the membrane surface as crystalline scale compounds that are difficult to remove, thus undermining the efficient permeation of pure water. Formation of these compounds is one of the main reasons why RO systems can normally treat only about 70% to 75% of the raw water.

Process Description

The high-recovery process concept comprises a number of configurations whose design depends on the influent water flowrate, its hardness, the concentration of silica relative to other hardness precursors, and its total dissolved solids (TDS). The process minimizes the scale potential of the water by continuously removing hardness ions (i.e., barium, calcium, and silica) from the RO concentrate stream as they form (4). Hardness and silica are removed efficiently by a process of chemical precipitation, IX softening, or a combination of these processes, depending on the raw water composition. As shown in Figure 1, the high-recovery process includes the following fundamental steps:

1. Pretreatment, as described above, including fine media or membrane (i.e., microfiltration [MF] or ultrafiltration [UF]) filtration, antiscalant addition, and if necessary, pH adjustment with acid to minimize the scale potential. Pretreatment should target an achievable silt density index $(SDI_{15}) < 4$ and preferably < 3.

2. First stage RO membrane purification step (RO-1) intended to treat a large portion of the water at low pressure (typically 50% to 75% of the influent). In many cases, this step could be an existing RO system, which produces a large reject stream of up to 30% of the influent volume, due to fouling and high scale formation potential. If this is an existing RO, it is necessary to review the existing pre-treatment system design and the RO reject quality, which might require enhancements to ensure compatibility with the high-recovery process.

3. Second-stage RO step (RO-2) that treats concentrate from the first stage (RO-1), combined with an appropriate flowrate of recycled RO-2 concentrate. The flowrate of the treated, high-TDS recycled RO-2 concentrate is computed based on mass balance and scale prediction calculations to ensure low scale formation potential over the second-stage RO membranes at the targeted overall process permeate recovery. The overall process permeate recovery is the total permeate from RO-1 and RO-2 membranes, expressed as a percentage of the influent water, which is typically greater than 95%.

TABLE A
Power Station High-Recovery Wastewater Treatment Process

Parameter	Raw Water Feed to RO-1	Permeate from RO-1	Permeate from RO-2	High-Recovery Permeate	High Recovery Reject
Temperature (°C)	25	25	25	25	25
Water flowrate (gpm)	280.0	210.0	64.8	274.8	5.25
SDI (SDI_{15})	<4	<2	<2	<2	>6
pH	6.5	6.0	6.5	6.0	9.0
TDS (mg/L)	939.4	17.2	114.9	41.2	48,201.7
TSS (mg/L)	<2	<1	<1	<1	8,366.0
Conductivity (µS/cm)	4,409.1	25.9	172.3	61.8	56,878
Total hardness (as $CaCO_3$) (mg/L)	139.2	2.55	1.86	5.46	7,136.6
Silica (as SiO_2) (mg/L)	50.2	0.92	1.36	1.02	2,623.2
Bicarbonate (mg/L)	82.6	1.52	3.31	1.9	0.0
Carbonate (mg/L)	0.0	0.0	0.0	0.0	4,305.0
Chloride (mg/L)	64.5	1.18	6.89	2.53	3,308.4
Fluoride (mg/L)	0.6	0.01	0.0	0.02	30.9
Sulfate (mg/L)	446.5	8.20	65.7	21.7	28,369.2
Barium (mg/L)	0.1	0.001	0.0	0.0	2.70
Calcium (mg/L)	22.3	0.41	0.30	0.87	1,142.7
Iron (mg/L)	0.1	0.003	0.0	0.01	7.1
Magnesium (mg/L)	20.3	0.37	0.27	0.79	1,040.0
Potassium (mg/L)	0.2	0.003	0.02	0.006	7.8
Sodium (mg/L)	251.7	4.62	37.0	12.3	1,612.2
Strontium (mg/L)	0.4	0.008	0.05	0.02	22.6
Recommended design pressure (psig)		185	725		

4. The fourth key step is the RO-2 concentrate-softening step. Softening of the volume-reduced high TDS RO-2 concentrate can be achieved cost-effectively by chemical precipitation of calcium, magnesium, and silica hardness, or, if silica is not limiting, by IX softening using strongly acidic cation exchange resins, weakly acidic IX resins, or metal-chelating resins. The chemical precipitation step uses caustic soda and/or soda ash, depending on the ratio of alkalinity to calcium hardness. Magnesium may also be added, depending on the relative concentration of silica in the RO-2 concentrate. Effective solids contacting is necessary to ensure efficient hardness and silica removal, reduced chemical consumption, and a minimum RO-2 concentrate recycle volume.

5. A small RO-2 concentrate reject stream is removed in order to control the TDS (and osmotic pressure) of the second stage RO concentrate, since the second stage is typically operated at a maximum pressure of 900 to 1,000 pounds per square inch gauge (psig). A small slurry reject stream is also removed from the bottom of the small clarifier and pumped out directly to a solar evaporation pond where it is disposed of, along with the small RO-2 concentrate bleed stream. Alternatively, the concentrate reject stream can be sent to a small thermal evaporator or spray dryer to enable close to 100% water recovery in an economical manner. The slurry reject stream typically has a suspended solids concentration of less than 1%, which enable its direct transfer as a slurry, thus alleviating the need for costly thickening and solids filtration equipment. The combined volume of the two small reject streams is less than 5%, giving an overall process recovery of greater than 95%.

6. In the IX softening configuration, the overall reject stream includes the RO-2 concentrate bleed and a small waste stream consisting of IX column back-wash and the spent brine used to regenerate the resin. As in the chemical precipitation softening scenario, the combined reject stream will be less than 5% of the influent, resulting in greater than 95% overall product water recovery.

7. The high-recovery process is designed with flexibility to control the relative recoveries obtained from the first and second RO stages in order to prevent scale formation, while achieving the maximum system recovery.

8. Depending on the influent water flowrate and concentration of silica and calcium, the high-recovery process will have a number of different configurations, including: RO-RO-CP*, RO-RO-IX, or RO-IX-RO.

*Where the term "CP" identifies the concentrate softening method, in this case "Chemical Precipitation", or IX for ion exchange softening.

9. The achievable overall process permeate recovery is determined by the feedwater TDS and the RO system design pressure. For influents with TDS less

Parameter High-recovery process configuration	Case 1 RO-1 + RO-2 + Precipitation	Case 2 RO-1 + RO-2 + IX Softening
Raw water flowrate (gpm)	1,388.90	694.4
Raw water flowrate (mgd)	2.00	1.00
Product water flowrate (gpm)	1,361.1	632.6
Product water flowrate (mgd)	1.96	0.911
Raw water TDS (mg/L)	703	8,000
Raw water hardness (mg/L as $CaCO_3$)	339	335.2
Raw water silica (mg/L)	25	0.0
RO-1 feedwater flowrate (gpm)	1,388.9	694.4
RO-1 permeate flowrate (gpm)	1,041.6	347.2
RO-1 permeate recovery (%)	75%	50%
RO-1 permeate TDS (mg/L)	12.9	110.5
RO-1 system operating pressure (psig)	175	305
RO-2 feedwater flowrate (gpm)	666.6	555.6
RO-2 permeate flowrate (gpm)	319.4	295.1
RO-2 permeate recovery (%)	47.9%	53.1%
RO-2 permeate TDS (mg/L)	186.5	687.4
RO-2 system operating pressure (psig)	500	1,015
Membrane system recovery (maximum) (%)	98.0	92.5
High-recovery process recovery (maximum) (%)	98.0	91.1
High-recovery system product TDS (mg/L)	54	376
Reject stream flowrate (gpm)	28	61.6
Reject stream TDS (%)	2.35	10.2
Reject stream TSS (mg/L)	7,500	<50
High-Recovery System Parameters		
Capital cost ($1,000s)	2,950	1,550
*Installed cost ($1,000s)	5,000	2,650
**Installed cost ($1,000s)	8,850	4,650
!Annual process operating cost ($1,000s)	1,110	440
Product water cost ($/m3)	0.65	0.55
Process product water cost ($/1,000 gal)	2.5	2.1

NOTES:
[a]See Endnote
* Installation inside existing building (scale-up factor of 1.7 used).
** Installation of high-recovery system skids in a "green site" (scale-up factor of 3.0 used).
! Annual operating costs include labor, chemicals, power, and membrane replacement costs.
Product water cost includes depreciation (10% straight line), labor, chemicals, power, and membrane replacement costs)
All costs are "conceptual engineering estimates", with accuracy of ± 30%.
Cost figures are in U.S. dollars.

than 1,000 mg/L, it would be possible to achieve overall permeate recoveries in the range 97% to 98%. For influents with TDS less than 500 mg/L, a recovery of 99% has been demonstrated.

Advantages

The following features are unique for the high-recovery process[a] referred to in this article.

1. The process maximizes the permeate recovery by continuously reducing the scale potential, rather than using a staged approach.

2. The RO membrane permeate recovery is limited only by the osmotic pressure and the applied operating pressure, enabling up to 99% permeate recovery on high hardness low-TDS influents, and reducing water demand especially in regions involving depleted water resources.

3. Control of scale formation potential by balancing the first-stage and second-stage RO system recoveries.

4. Reduces the capital and operating costs of the overall process since treatment is applied to a relatively small RO concentrate stream, equivalent to 30% to 50% of the raw water flowrate. By achieving very high recoveries, substantial cost savings are realized with respect to raw water and wastewater disposal costs. The process can be used to further recover water from large RO reject streams produced in applications involving wastewater recovery and potable water from brackish water. The cost of high quality water is reduced substantially.

5. For low-TDS and large influent flow applications, the process deploys a two-stage system to minimize pressure energy cost.

6. The process can be integrated with existing RO systems to enhance the water recovery by achieving 95% to 99% overall product water recovery.

7. Produces product water that is as pure as needed, ranging from desalinated potable water or treated industrial effluents to high-purity water for steam generators or semiconductor wafer manufacturing.

8. By reducing the reject volume to less than 5%, and even to less than 2%, the high-recovery process obviates the need for costly brine concentrators, and enables the option of zero liquid discharge, by deploying a solar pond or a small spray dryer, depending on the influent flowrate, and the site meteorological conditions.

9. The process minimizes adverse impact of the water treatment process on the environment by enabling complete water recovery.

Process Application Case Study

TABLE C — Comparison of Annual Costs*						
RO Recovery (%)	Evaporator (MVC)	RO + MVC Evaporator (different RO recoveries)			High-Recovery RO + MVC	
80%	95%	RO Rec. 70%	99%		RO Rec. 75%	RO Rec.
Annual cost ($/annum)	$3,500,000	$1,700,000	$1,600,000	$1,450,000	$1,200,000	$1,100,000
Cost savings over MVC (%)	NA	51.4%	54.3%	58.6%	65.7%	68.6%
Cost savings over RO + MVC (%)	NA	NA	NA	NA	24.2%	30.5%

*($1,000s) to process 1,000 gpm to ZLD mechanical vapor compression evaporator (MVC) versus (RO + MVC) versus (high-recovery system + MVC). NA = not applicable

The performance of the high recovery process can best be illustrated by the power station example displayed in Figure 1 and Table A.

In this case, approximately 280 gallons per minute (gpm) of effluent, consisting of cooling tower blowdown, RO membrane system reject water, and sand filter backwash were equalized, aerated, and filtered. Following pretreatment, a scale inhibitor was added prior to the high recovery membrane process. The process includes a first-stage, low-pressure RO membrane system (RO-1) operating at approximately 185 psig, and delivering 210 gpm of RO-1 permeate, which corresponds to 75% permeate recovery in the first stage. The concentrate from the first stage is combined with the treated portion and introduced into the high-pressure second stage system (RO-2) to achieve further water recovery.

This "seawater" RO membrane system operates at 700 to 800 psig, and produces up to 64.7 gpm of RO-2 permeate. In applications where RO-1 is an existing system, rejects from this system might require additional treatment before introducing them into RO-2. Also, the first-stage RO system operating conditions may need to be optimized through modeling, including adjustment to its permeate recovery, to enable efficient integration with the second stage RO system. The combined high-recovery process (RO-1 and RO-2) permeate flow-rate is up to 274.7 gpm, corresponding to an overall product water recovery as high as 98.1%.

As shown in Figure 1, most of the RO-2 membrane system concentrate is softened using a high-pH chemical precipitation step where caustic soda and other chemicals are added to precipitate the calcium, magnesium, and silica. A small RO-2 concentrate reject "bleed stream" is removed to control the TDS and the second-stage osmotic pressure. The reaction mixture is clarified, filtered, recycled, and blended with the feed to the second-stage RO system to enable further water recovery, while mitigating scale formation. By implementing recycle of this high-TDS softened water stream, it is possible to achieve the 98.1% overall water recovery.

A small slurry reject stream containing approximately 0.8% W/V of total suspended solids (TSS), (predominantly calcium carbonate, silica, iron and magnesium hydroxides, calcium fluoride, and barium sulfate), is also withdrawn from the bottom of the clarifier and disposed of along with the high-TDS RO-2 reject bleed, to an existing solar pond via the common reject line. The volume of the combined rejects is in the range 5 to 10 gpm. The filter backwash can be recycled to the clarifier. By performing the softening on the relatively small volume of the RO-2 concentrate, the size, footprint, and capital cost of the softening equipment train is reduced substantially. The improved chemical softening efficiency also translates into a smaller quantity of chemicals and reduced annual operating costs.

While the RO-1 permeate TDS is 17.2 mg/L, the RO-2 permeate TDS is substantially higher (i.e., 114.9 mg/L), as to be expected, yielding a composite product water TDS of 41.2 mg/L, composed predominantly of sodium sulfate. This water can be readily reused as make-up water for the cooling tower, or it can be blended with pre-filtered raw water as feed to the water treatment plant (WTP), thereby reducing demand on the filtered water supply. Because of its very low hardness and low TDS, this product water will also result in a higher achievable number of cooling tower cycles, as well as a substantial reduction in the ionic load on the WTP's IX resin, thereby reducing the frequency of the regeneration cycle and the volume of spent regenerant produced.

By using this high-recovery process, the power station is able to use an existing "solar" pond for the disposal of the small volume of composite rejects, averaging approximately 5 to 10 gpm, depending on the ambient temperature, thus obviating the need for a costly thermal evaporator. As seen in Table A, the concentration of total dissolved solids in this final process reject stream is approximately 4.8% and the total suspended solids approximately 0.84%, thereby facilitating direct pump-out of a relatively small slurry stream, without the need for thickening and other costly sludge processing equipment.

Other Examples
Table B shows a summary of computed high-recovery system process performance parameters and estimated capital and operating costs for two other high-flow, high water recovery cases.

As in the previous example (Table A, Figure 1), Case 1 in Table B represents a relatively low influent TDS of

703 mg/L, thus enabling a high overall process recovery of 98%. Because of the relatively high influent water hardness (339 mg/L as CaCO$_3$) and silica (25 mg/L) and relatively low TDS, a two-stage high-recovery process is used in this case, including post-softening with chemical precipitation. Starting with a raw water flowrate of 2 million gallons per day (mgd), a purified (product) water flowrate of 1.96 mgd is obtained, with a TDS of 54 mg/L, and producing a small reject stream with a TDS of 2.35% and a TSS of 0.75%. The operating pressures of the first stage and second stage (i.e., RO-1 and RO-2) are estimated at 175 psig and 500 psig, respectively. The installed capital cost of the system is estimated at U.S. $5 million (± 30%), the annual operating cost approximately $1.1 million and the product water cost approximately $2.50 per 1,000 U.S. gallons, including a 10-year straight-line depreciation component (5).

Case 2 in Table B represents a different high-recovery process configuration involving use of IX softening of the blended RO-1 concentrate and second-stage RO-2 concentrate (Figure 1). Use of simple IX softening treatment is made possible in this case primarily because of the absence of silica. However, the overall process recovery of 91% is limited by the relatively high influent TDS of 8,000 mg/L. Similarly, the product water TDS is relatively high (376 mg/L). However, if a more pure product water is desired, a relatively "low-cost" second-pass RO membrane system can be used to further decrease the TDS to less than 10 mg/L, without adversely affecting the overall process water recovery.

In contrast to the chemical precipitation scenario in Case 1, the installed capital cost of treating a flowrate of 1 mgd of high-TDS raw water in Case 2 is substantially lower (i.e., $2.65 million [± 30%]), the operating cost $0.44 million per annum, and the product water cost approximately $2.10 per 1,000 U.S. gallons, reflecting the substantially reduced process complexity.

Economics

Until relatively recently, thermal energy was used to distill water with high salinity, or chemical and biological contaminants, to produce potable water or industrial service water. We can now use a simple low-temperature membrane processes (e.g., RO) to achieve the same, but at substantially lower cost (i.e., almost one third the cost to evaporate water), needless to mention there are also environmental benefits.

Let us compare a ZLD scenario where we need to treat 1,000 gpm of water or wastewater, and produce high-quality water for municipal use or for industry. Table C shows the approximate annual costs and cost savings when the high-recovery process is used to achieve up to 99% recovery of high quality water (5). The annual cost includes depreciated capital and operating costs (i.e., labor, power, chemicals, and membrane replacement). These costs are compared to efficient evaporative processes (e.g., mechanical vapor compression [MVC]). Energy savings may total 80% to 90%.

While the second column represents the annual cost of a high-efficiency MVC evaporator operating exclusively to process 1,000 gpm, the third, fourth, and fifth columns represent the annual costs of integrated RO-MVC process scenarios which are typical of industry's current practice. The cost advantage of increasing the RO permeate recovery upstream of the MVC is illustrated by considering three RO system recoveries (i.e., 70%, 75% ,and 80%, respectively). Conventional RO systems typically operate at 70% to 75% recovery. By treating the water with RO membranes first, an average annual cost saving of 50% to 55% is realized, mostly in the form of energy cost savings, when compared to using MVC exclusively.

The last two columns of Table C represent two high-recovery RO-MVC scenarios, with membrane system recoveries of 95% and 99%, respectively. Overall, a high-recovery RO-MVC process operating at 95% to 99% product water recovery will save 65% to 70% in annual costs when compared to the cost of MVC evaporators exclusively (i.e., column 2), and up to 30% when compared to using an integrated process involving "conventional" RO with limited permeate recovery, followed by MVC evaporators (i.e., columns 3, 4, and 5). These profound cost savings render the high recovery process economically viable and, in many cases, there is no need for mechanical evaporators downstream. Because of the very small volume of reject stream from high-recovery RO, a small solar pond, a small spray dryer, or other low-cost disposal options become viable.❏

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Endnote
aThe membrane recovery method referred to in the text as the "high-recovery process" is known as the ARROW™ process. This patented process has been licensed exclusively to O'Brien & Gere, which is based in Cincinnati, Ohio.