EXECUTIVE SUMMARY

A Fully Integrated Membrane Bioreactor System for Wastewater Treatment in Remote Applications

SERDP Project ER-2237

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1.0 INTRODUCTION

An osmotic membrane bioreactor (OMBR) system with membrane distillation (MD) reconcentration was developed for potable reuse of wastewater at forward operating bases (FOBs). The three major processes of this system – biological reactor (BIO), forward osmosis (FO), and MD – were systematically investigated at bench scale before laboratory-scale versions were designed, fabricated, and integrated into a single system. The modular system was designed to operate using low-grade ("waste") heat existing at FOBs. The abundance of low-grade heat produced by power generators at FOBs make the OMBR-MD system ideal for such locations.

Membrane bioreactors (MBRs) have proven to be effective and beneficial for wastewater reclamation due to their ability to produce high quality effluent with a small footprint [6]. In a conventional MBR, microporous (microfiltration [MF] or ultrafiltration [UF]) membranes are immersed in a bioreactor and water is filtered through the membranes using vacuum. Suspended solids are then retained in the system and high levels of treatment can be achieved [7, 8]. MBRs replace the two stages of the conventional activated sludge process with a single, integrated process that has a much smaller footprint. MBR effluent may be suitable for use as irrigation water, process water, or potable water. For potable reuse, advanced treatment (e.g., reverse osmosis [RO] and chemical oxidation) is necessary after the MBR [9].

The advantages of MBRs over conventional treatment have been thoroughly reviewed [6] and include consistently high product quality, ease of operation/automation, a reduced footprint, reduced sludge production due to a high biomass concentration in the bioreactor, and complete suspended solids removal from the effluent [6]. The main problem associated with MBRs is membrane fouling. Membrane fouling can occur in the MBR itself and in the downstream RO system [10]. Specifically, high concentrations of dissolved organic compounds in the MBR effluent can cause severe fouling of RO membranes.

Membrane fouling lowers productivity, increases energy requirements, increases frequency of membrane cleaning and replacement, and may result in deterioration of treated water quality [11]. A novel MBR system that utilizes a submerged FO membrane in the bioreactor and a MD membrane in a separate reconcentration process was investigated. Prior to describing the integrated system, the basic FO and MD processes are outlined below.

Osmosis is the net diffusive transport of water through a selectively permeable membrane from a solution of low solute concentration (low osmotic pressure) to a solution of high solute concentration (high osmotic pressure). In osmosis, the membrane allows passage of water, but rejects almost all solute molecules and ions. FO is an engineered application of osmosis [12, 13].

In FO, impaired water is in contact with the dense side of a semipermeable membrane and a highly concentrated draw solution is in contact with the support side of the membrane. The draw solution is typically an aqueous solution of a low molecular weight salt [14]. In the FO process, relatively pure water is transported from the impaired water into the draw solution; the impaired water becomes concentrated and the draw solution becomes diluted. A desalination process (e.g., RO or distillation) can be used to reconcentrate the draw solution and simultaneously produce high-quality product water (Figure 1). Thus, in most water treatment applications, FO is not the ultimate process but rather a high-level pretreatment process before an ultimate reconcentration/ desalination process [14].

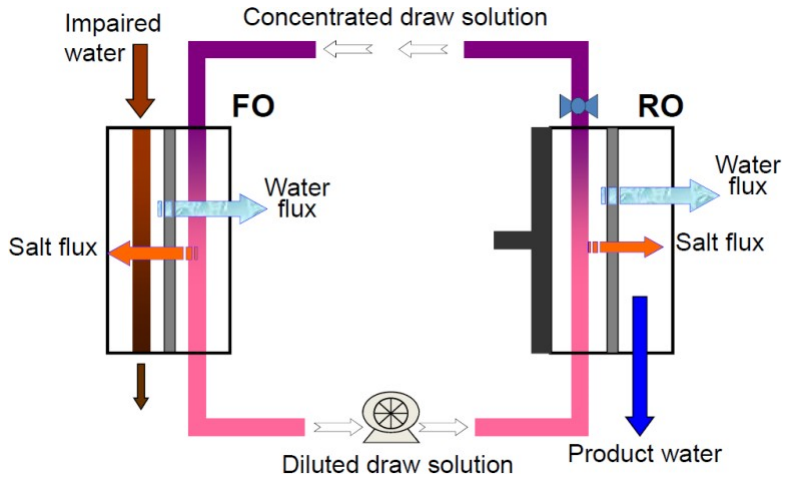

Figure 1. Forward Osmosis (FO) System Schematic

FO has been evaluated for seawater and brackish water desalination [12, 13, 15], wastewater concentration and reclamation [16-19], and food concentration [20-22]. FO can also be employed in conjunction with biological processes for wastewater reuse in OMBRs [23, 24]. The main advantage of FO is that it operates at very low hydraulic pressure, it has high rejection of a broad range of contaminants [23, 25], and may have lower fouling propensity and/or fouling that is more reversible than in RO processes [18, 26].

MD is a non-pressure-driven membrane separation process that has the potential to become a viable tool for water desalination and brine (or draw solution) concentration. MD involves the transport of mass and heat through a hydrophobic, microporous membrane. In direct contact MD (DCMD) (Figure 2), a feed solution at elevated temperature is in contact with one side of the membrane and a permeate solution at lower temperature is in direct contact with the opposite side of the membrane, and it is mainly the temperature difference between the liquids that induces the vapor pressure gradient for mass transfer. DCMD is one of the simplest configurations of MD; it requires only a membrane module and two low-pressure pumps to move the liquids over the membrane. The simplicity of this configuration makes it highly suitable for implementation in large-scale applications.

Compared to conventional distillation methods, MD requires only small temperature differences, which are achievable through the use of low-grade heat/renewable energy. Compared to RO, the driving force in MD is essentially not reduced by osmotic pressure and thus, MD operates at hydrostatic pressure and can be used to treat high salinity solutions or provide enhanced recovery through brine desalination. Also, compared to RO, MD produces an even higher quality distillate; it does not allow the passage of small non-volatile compounds and provides high removal of emerging contaminants. Specifically, MD has been shown to reject greater than 97% of low molecular weight organics [27]. For these reasons, MD may have substantial energy, recovery, maintenance, and water quality advantages over conventional methods.

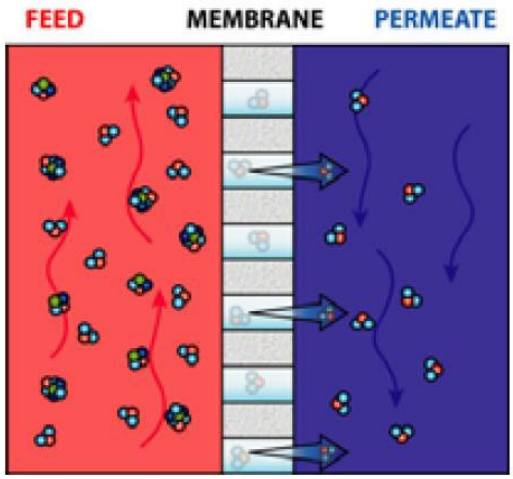

Figure 2. DCMD Feed and Permeate Solutions Are Both in Direct Contact With the Membrane

One very important aspect of the MD process is the membrane cannot become hydrophilized. Hydrophilization can occur due to fouling of the MD membrane if proper pretreatment is not used. FO provides outstanding pretreatment of feed streams to MD membranes because it provides high rejection of dissolved species [18, 23].

In an OMBR system (Figure 3), wastewater is fed into a reactor which is aerated as needed to supply oxygen for the biomass and to minimally scour the membrane. Through osmosis, water diffuses from the bioreactor, across a semipermeable membrane, and into the draw solution. The FO membrane acts as a barrier to solute transport and provides high rejection of contaminants in the wastewater stream [16, 19, 25]. The diluted draw solution is sent to a reconcentration process (e.g., RO or MD), which reconcentrates the draw solution and generates a high-quality product water. Thus, FO serves as a high-level pretreatment step before the RO or MD reconcentration. Compared to the MF or UF process in a conventional MBR, the FO process in the OMBR offers the advantages of much higher rejection (semi-permeable membrane versus microporous membrane) at lower hydraulic pressure. The FO membrane inside the bioreactor also has much lower fouling propensity than the MF/UF membranes, and thus, requires minimal scouring and much less frequent backwashing. When comparing an OMBR system (OMBR followed by RO or MD reconcentration) with a conventional MBR-followed-by-RO system, the high rejection of the FO membrane results in RO influent with lower fouling propensity and ultimately produces higher quality product water. Furthermore, the FO-followed-by-RO or -MD reconcentration treatment scheme represents a tight dual barrier system. This is particularly important when pharmaceutical or personal care products are present.

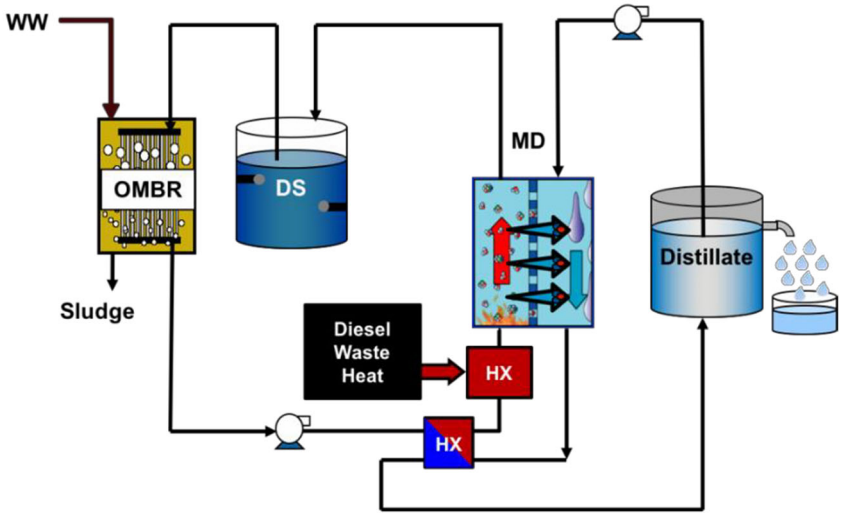

Figure 3. OMBR System

During the initial stages of development, bench-scale systems were used to demonstrate the feasibility of OMBR with MD reconcentration for wastewater treatment and reuse and to evaluate process performance and water quality with each process. Alternating aeration on/off cycles, or aeration cycling, was used to accomplish biological carbon and nitrogen removal in a single reactor with the OMBR. Bench-scale experiments were conducted to determine the effects of FO membrane module configuration (submerged in the bioreactor or as a sidestream process) on OMBR performance. Bench-scale results were used to guide development of the pilot-scale system and performance of a submerged FO membrane module and a spiral-wound membrane module were evaluated at pilot-scale. Process metrics were established to evaluate membrane performance and wastewater treatment; membranes and membrane modules were evaluated for water flux, reverse salt flux, and solute rejection and wastewater treatment was evaluated for contaminant (carbon and nitrogen) removal. Results from long-term testing using a high-strength wastewater showed essentially 100% removal of chemical oxygen demand (COD) and 96.2% removal of NH_4^+-N could be achieved with aeration cycling in a single reactor. The research results from this SERDP project have been published in several peer-reviewed journal articles [1-5], with more in preparation.

2.0 OBJECTIVES

The main objective of this research was to design, fabricate, and test a pilot-scale OMBR-MD system consisting of BIO, FO, and MD processes and driven by on-site waste heat. As part of this, the operating parameters necessary for highly efficient wastewater treatment and water reuse were determined. The biologically-catalyzed processes of the OMBR provide a combined treatment process for organic matter degradation and nutrient removal. When coupled to the osmotically driven mass transport mechanisms of FO, the system offers a low-fouling alternative to traditional MBR applications while also decreasing the electrical energy demands required for the process. The FO membranes also serve as a selective barrier system capable of rejecting recalcitrant trace organic compounds in order to prevent passage into the draw solution stream, thereby decreasing fouling in the downstream MD process.

The steps toward achieving the goals of the proposed research were to systematically investigate the three major processes of the OMBR system (BIO, FO, and MD) and determine the energy requirements for each process. Subsequently, the processes were integrated into a single system through the design and construction of a modular pilot-scale OMBR system that operates using renewable energy or low-grade ("waste") heat existing at FOBs. At each stage of the research, process performance was evaluated based on specific indicators (e.g., water quality and quantity, energy and resource consumption, and frequency of hands-on modification to the system).

To carry out the objectives, several tasks were performed including fundamental studies aimed at researching specific issues for each of the processes (BIO, FO, and MD) and the heat exchangers. Investigation of the critical phenomena for each process enabled answering of fundamental questions about microbial ecology, FO membrane fouling, and MD membrane flooding, which are essential for scale-up and integration of these technologies. The research team collaborated with Hydration Technologies, Inc. (HTI) (now Fluid Technology Solutions, Inc.), GE Water & Process Technologies, and Parker Performance Materials to acquire the most appropriate FO and MD membranes to characterize and test. Then, process models and controls for each of the subsystems were developed. Finally, OMBR and MD processes were integrated and a system-level model was developed and tested for water quality and membrane performance.

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3.0 TECHNOLOGY DESCRIPTION

To design, fabricate, and test a pilot-scale OMBR-MD system consisting of a BIO, FO, and MD process and driven by on-site waste heat several tasks were performed, as outlined below. The project started by investigating fundamental aspects of the main components of the OMBR-MD system in Task 1: bioreactor design, instrumentation, and control, Task 2: bench-scale FO membrane fouling, Task 3: bench-scale MD process modeling, and Task 4: heat exchanger design and computational fluid dynamics (CFD) modeling for MD. Then, it progressed toward the assembly of the integrated pilot-scale system in Task 5: lab-scale BIO+FO sub-system testing, Task 6: design of lab-scale MD + heat exchanger sub-system, Task 7: lab-scale FO-BIO and MD-heat exchanger sub-system assembly, Task 8: lab-scale OMBR system testing, and Task 9: OMBR system scale-up testing.

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4.0 RESULTS AND DISCUSSION

4.1 DEVELOPMENT OF AN OXIC-ANOXIC BIOREACTOR FOR TREATING FOB WASTEWATER

A simultaneous nitrification-denitrification process was tested and optimized. The bioreactor used for these experiments was inoculated with a combination of activated sludge, bacteria from a nitrifying trickling filter biomass, and microbes from a denitrifying system at a regional wastewater treatment facility. Bench-scale experiments were conducted in a pair of bioreactors specially designed to accommodate a flat sheet membrane. The two process waste streams were tested in parallel using the two systems: (1) graywater influent [28] and (2) combined gray+black wastewater (based on typical domestic wastewater composition). For purposes of this subtask, the unit mimicked a traditional MBR system in that it had an MF membrane solely for the purpose of retaining biomass. In subsequent research tasks, the biological system was combined with the FO system, which served to retain more than biomass. Since the FO system in subsequent tasks contains a semi-permeable membrane that is similar to a RO membrane, both suspended and dissolved constituents can be retained within the system. In the current task, which utilizes an MF membrane (pore opening size ~0.1-0.2 micrometers [µm]), only suspended constituents larger than this size were retained. The bioreactor was operated in alternating aerobic/anoxic reaction modes to allow for carbon and nitrogen removal. The following general, non-balanced chemical reactions (Eqs. 1-3) describe the major reactions occurring in each mode. Operation in this fashion allowed simultaneous urea hydrolysis (Eq. 1), nitrification (Eq. 2), and denitrification (Eq. 3), which ultimately removed dissolved contaminants.

Aerobic & Anoxic: $\text{Urea} + \text{Microorganisms} \rightarrow NH_3 + CO_2$ *(Equation 1)*

Aerobic only: $NH_3 + O_2 \rightarrow NO_2^-/-/NO_3^- + H_2O$ *(Equation 2)*

Anoxic only: $\text{Organic Carbon} + NO_2^-/NO_3^- \rightarrow N_2 + CO_2 + H_2O$ *(Equation 3)*

Biochemical experimental variables monitored during the experiments included DOC, ammonia-nitrogen (NH_3-N), nitrite-nitrogen (NO_2^--N), nitrate-nitrogen (NO_3^--N), and biomass (measured as volatile suspended solids). Chemical parameters were measured in the feed stream, within the bioreactor during reaction cycles, and in the final effluent stream using accepted analytical methods.

Microbial populations and ecology (see below) were monitored at reasonable long-term intervals (i.e., 15-30 days). To be effective at achieving complete treatment, cycle duration and switching between aerobic and anoxic conditions were optimized. While these processes were relatively easy to optimize individually, it was technically challenging to engineer these processes such that they occurred in a single system and consumed a minimum of resources (specifically, oxygen and exogenous resources such as energy for mixing, internal recirculation, and membrane cleaning).

Shortcut denitrification is a process where the nitrogen in ammonia is only oxidized to nitrite (NO_2^-) which is subsequently reduced to nitrogen gas during denitrification. This process relies on less oxygen (and consequently less energy) than traditional nitrification-denitrification sequences. As such, a major thrust was to perform fundamental research on a single-sludge process and study the microbiological community shifts that occurred throughout the system.

The central hypothesis that was addressed during this research phase was: a balanced nitrification-denitrification treatment process can be achieved with carefully tailored oxic and anoxic reaction times based on chemical signatures in the bioreactor. Thus, a major component of this task was to identify proper chemical cues (including water quality and biochemical indicators) that were required to switch between operational modes. Specifically, oxidation-reduction potential (ORP), dissolved oxygen (DO), and pH were continuously analyzed along with water quality parameters (i.e., ammonia, nitrite, nitrate, total organic carbon [TOC], and COD) to determine appropriate switching times between aerobic and anaerobic reaction sequences.

Simulated FOB wastewater was developed to challenge the microbial reactor system and enable researchers to rigorously identify the switching requirements for achieving maximum nutrient removal. The key FOB wastewater constituents that were mimicked in the laboratory study were organic carbon load (measured as TOC and COD), nitrogen speciation and respective constituent concentrations (urea and ammonia), typical nutrients and micronutrients (phosphorus, iron, sulfur), and suspended solid concentrations (measured as total suspended solids and volatile suspended solids).

4.2 DEVELOPMENT OF AN INTEGRATED SENSOR-BASED CONTROL SYSTEM FOR THE BIOREACTOR

A control and operating program for the biological process of the OMBR system was developed; specifically, the program allowed biological nutrient removal in a single reactor. The program relied on DO, ORP, pH, and time to switch between oxic and anoxic phases to achieve nitrification and denitrification, respectively. This subtask was built upon data collected in the previous task, which describe changes in biochemical reaction during wastewater treatment.

A LabVIEW-controlled system was added to the bioreactor system used during the previous task and a computer-controlled system was developed to allow the aeration system to automatically turn on and off as a function of bioreactor probe outputs. Briefly, experimental data provide a suite of ORP, DO, and pH performance curves that were correlated with the biochemical environment, specifically, dissolved nitrogen speciation and organic carbon concentrations. Based on these data, real time switching criteria were incorporated into the LabVIEW-controlled system to control and maintain proper air delivery to the bioreactor.

4.3 FO MEMBRANE FOULING, PROCESS MODELING, AND CONTROL

FO uses an osmotic pressure differential across a semipermeable membrane, rather than hydraulic pressure differential (as in RO), as the driving force for water transport through the membrane. The FO process results in concentration of a feed stream and dilution of an osmotic agent (i.e., a draw solution).

There is evidence that in some cases, fouling occurs to a lesser extent in FO than in RO [18]. In other cases, it has been found that fouling occurs at similar rates in FO and RO; however, the fouling is more reversible in FO – possibly due to the absence of hydraulic pressure in the FO feed stream [27]. Although there is indication in the literature that the presence of salt at the membrane surface may play a role in membrane cleaning [29], the basis for the more reversible fouling has not been proven. This was explored to provide critical insight into FO transport phenomena and fouling reduction.

FO membranes were used to conduct bench-scale fouling and cleaning experiments. A range of foulants (organic, inorganic, colloidal, and biological) were used to assess the ability to remove these foulants using rinsing and osmotic backwashing. During osmotic backwashing, water passed from the support side of the membrane to the active side, thereby reversing the direction of flow through the FO membrane and likely removing foulants. Foulant removal was determined by hydraulic testing and scanning electron microscopy imaging.

4.4 MD MEMBRANE HYDROPHILIZATION, PROCESS MODELING, AND CONTROL

Maintaining membrane hydrophobicity is absolutely essential to ensure that the pores of MD membranes do not become wet. If hydrophobicity is lost, the pores can wet with water and salts and other contaminants can diffuse into the distillate [30]. Pore wetting is considered irreversible, in other words, after the pressure that caused pore wetting is removed, the pores remain wet. In order to restore permeate quality, removal of fluids and drying is required. Pore wetting results in a decline in distillate quality; downtime and prevention of this phenomenon is absolutely critical for successful MD scale-up.

The interrelationship between hydrophobicity and pore size distribution is described in terms of the liquid entry pressure (LEP). LEP is given by the difference between liquid pressure and vapor pressure:

$$P_1 - P_V \leq -\frac{2\gamma_L \cos\theta}{r} \qquad \text{(Equation 4)}$$

Where, P_l is the pressure of the liquid in the channels of the MD module, P_v is the pressure of the vapor inside the membrane pores, γ is liquid surface tension, θ is contact angle, and r is pore radius (for non-cylindrical pores, a geometry correction factor needs to be introduced). Theoretically, the membrane pores remain dry as long as the LEP is not exceeded.

4.5 ENERGY CONSIDERATIONS FOR MD

In this task, the MD system was analyzed using commercial CFD tools to determine energy and flow requirements and to optimize MD system design. The thermal and hydraulic characteristics of the MD system and its heat exchanger system were numerically analyzed using ANSYS Fluent, a commercial CFD package. The operational conditions and dimensions based on the small flat-sheet MD system under testing were considered for the CFD analysis to calculate energy and flow requirements of the MD system.

4.6 TESTING OF THE FO + MD SUB-SYSTEM

Integration of the FO and MD processes centered on selection of an appropriate draw solution. The draw solution for the FO process was the feed solution for the MD process; the ideal draw solution was one that optimized flux in FO and was easily reconcentrated by MD. It was also one that did not reduce MD membrane hydrophobicity after exposure to high temperatures over longer time periods. After screening the draw solutions, bench-scale FO experiments were carried out to test the water flux produced by each draw solution and quantify the ability to reconcentrate the selected draw solutions.

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5.0 IMPLICATIONS FOR FUTURE RESEARCH AND BENEFITS

An integrated OMBR-MD system was designed, fabricated, and tested at the pilot-scale for wastewater treatment and production of high-quality reuse water. There were very clear synergies in using MD to reconcentrate the draw solution used in the OMBR process, in using low-grade heat/renewable energy to drive the MD process, and in recovering heat from the distillate solution to reduce the overall heat consumption of the system. The abundance of low-grade heat produced by diesel generators at FOBs make the OMBR-MD system an ideal system for such locations. During long-term OMBR operation, carbon and nitrogen removal was achieved in a single reactor by alternating between aerobic and anoxic bioreactor conditions. Results from long-term testing using a high-strength wastewater showed 98.4% removal of COD and 90.2% removal of $NH4^{+}$-N was achieved.

Physical design aspects and process controls for system integration as well as procedures for wastewater treatment with the single-stage OMBR were established. Several areas for improvement were also identified throughout the development and testing phases of this system. Namely, passage of some dissolved, low molecular-weight contaminants into the closed-loop FO-MD solution led to MD membrane fouling and reduced MD water flux, and over time, the ammonium concentration in the distillate solution tended to increase. A suggested improved design will draw upon this and other lessons learned during the current project. Ammonium passage into the distillate solution is mitigated by improved biological ammonium removal (i.e., nitrification/denitrification) in the OMBR. To accomplish this, it is suggested to configure the OMBR with a dual-chamber tank with separate anoxic and oxic zones. Aeration in the oxic zone would be optimized for nitrification and membrane cleaning; the anoxic zone would be optimized for denitrification. The revised OBMR-MD system operates in a semi-batch configuration defined by two separate process loops. One loop operates the OMBR and the other operates multiple MD modules. Each loop operates individually until two triggering conditions are met; the draw solution reaches a target dilution in the OMBR loop and the MD feed solution reaches a target concentration in the MD loop. Once the triggering conditions are met, the solutions in each loop are exchanged; the diluted FO draw solution is then treated by MD and the concentrated MD feed solution is then diluted by FO. This semi-batch process scheme allows for independent and "continuous" operation of the FO and MD loops, reducing the complexity of the necessary control mechanisms to maintain steady-state operation of a dynamic, continuous process and thus implementing a simplified operation that maximizes the performance of each process. Once the tank switch occurs, the diluted draw solution volume will pass to the MD loop and undergo further treatment through a fine screen, a cartridge filter, a carbon filter, and an ultraviolet reactor to reduce the possibility of membrane fouling, membrane damage, or passage through the MD membrane by small organic compounds or disinfection residual.

The suggested revised system will be programmed to periodically backwash the FO membranes to extend membrane life and improve performance. The backwash cycle will utilize a portion of the distillate stored in a standalone tank. When FO performance degrades, or after a predetermined time period has elapsed, the system will cease normal operation and change to the necessary valve configuration to route the distillate to the FO module. Backwashing in a counter-flow regime will reverse the impact of fouling and scaling that occurs over time on the FO membrane. The used backwash solution will be further treated with MD to maximize water recovery.

Regarding the MD subsystem and based on testing subsequent to development of the current OMBR-MD system, it is suggested to considered use of an FO module in the revised system instead of the DCMD module that is used in the current system. In AGMD, heat recovery can be integrated into the module design.