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Application of integrated chemical precipitation and ultrafiltration as pre-treatment in seawater desalination

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ABSTRACT

Chemical precipitation including lime softening, chlorination, clarification and filtration have been long recognized treatment for process water used in various industries because it provides consistent water quality, however these processes were rarely used in pre-treating feed to reverse osmosis desalination systems. The newly developed submerged spiral wound ultrafiltration membranes in conjunction with chemical precipitation or lime softening can be used to treat reverse osmosis feed. Previous studies have proven the feasibility of integrating immersed ultrafiltration (UF) membrane with chemical precipitation and lime softening. This paper describes the relationships of the operating parameters such as pH, membrane flux, trans-membrane pressure (TMP), membrane permeability; optimum dosage of coagulant; to be used as the basis for the design and construction of the full scale plant. A pilot unit consisting of chemical reactor and UF system was operated at various pH conditions, at membrane flux rates of 15, 30 and 45 gfd. The filtrate from the pilot unit when operated as lime softener achieved alkalinity reduction to 20-30 mg/L with lime dosage close to the calculated value. The filtrate turbidity during the test using ferric sulfate and lime was consistently within 0.04-0.05 NTU. The TMP values obtained during the test ranges from 0.1 to 2.5 psi, while the permeability values ranges from 18.19 to 29.6 gfd/psi. The results the pilot test can be used as the basis of designing and operating a full scale integrated chemical precipitation or investigation of other applications of the integrated lime softening and UF treatment process.

Keywords: Chemical precipitation; Ultrafiltration; Seawater desalination; Pre-treatment

1. Introduction

Various manufacturing and industrial processes require treatment of potable municipal water supply, as well as wastewater and seawater. The municipal water supply at minimum should comply with the primary and secondary National Drinking Water Standards as shown in Table 1. The municipal water supply however vary

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from one area to another and may not be able to provide consistent quality required for various manufacturing and industrial processes therefore additional treatment is necessary. The municipal water supply also contains impurities that may affect specific manufacturing and industrial processes.

Other source of water provided to many industries and power plants are seawater and treated domestic or municipal wastewater. A typical seawater analysis is shown in Table 2 and secondary treated municipal wastewater analysis is shown in Table 3. In some industrial plants or power stations, organic matter, chloride, hard-

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Table 1

Selected contaminants limits in the national primary and secondary drinking water standards (EPA, 2003)

Primary drinking water standards		
Turbidity, NTU	<1–0.3 in 95% of daily	
	sampling in a month	
Chlorine (as Cl2), mg/L	MRDL = 4.0	
Cu, mg/L	1.0	
Pb, mg/L	0.015	
As, mg/L	0.010	
F, mg/L	4.0	
Hg, mg/L	0.002	
NO ₂ (as N), mg/L	10	
NO3 (as N), mg/L	1.0	
TTHMs, mg/L	0.08	
Secondary drinking water standar	rds	
Aluminum, mg/L	0.05–0.2	
Chloride, mg/L	250	
Sulfate, mg/L	250	
Color, CU	15	
Foaming agents, mg/L	2.0	
Iron, mg/L	0.3	
Manganese, mg/L	0.05	
Odor, threshold number	3	
pH	6.5-8.5	
TDS, mg/L	500	

ness, alkalinity and dissolved silica in the water supply is a concern.

Lime softening is the most common chemical precipitative water treatment process used in industries. The typical water treatment process includes pre-chlorination, lime softening with ferric salt dosage, media filtration or manganese greensand filtration. The addition of coagulants, such as ferric salts in lime softening process promotes better sludge settling and also can reduce organic matter in the raw water.

Recently, many industrial and manufacturing plants are replacing the lime-soda softening with other processes such as reverse osmosis, microfiltration and/or ultrafiltration. These processes, in most cases provide treated water that meets the quality requirements of the plants. However, there are cases when lime softening can not be replaced by reverse osmosis, especially when the high concentration of hardness, and dissolved silica in the raw water limits the recovery in the RO system. RO is excellent in reducing total dissolved solids, hardness and alkalinity in raw water, but it requires pre-treatment such as media filter or membrane microfiltration or ultrafiltration. The majority reverse osmosis plants require the raw water feed to be chlorinated to prevent biological fouling of the RO membranes. The drawback of chlo-

Table 2 Typical seawater analysis. Source: Bermuda Types Bay

>5
421
1290
11611
383
167
20734
2610
<5
4.5
8.0

Table 3

Typical secondary treated wastewater analysis. Source: South San Francisco secondary treated sewage

Turbidity, NTU	>10
Chlorine (as Cl2), mg/L	0.5
BOD, mg/L	20
Suspended solids, mg/L	30
Ca + Mg, mg/L as CaCO ₃	629
Na, mg/L	777.2
NH4, mg/L	35
Cl, mg/L	1450
SO4, mg/L	1.0
Alkalinity, mg/L as CaCO3	210
pН	8.0

rination of RO feed water is the breakdown of organic matter into smaller molecules and the formation of trihalomethanes (THMs), which are not rejected by the RO membranes.

It is common for UF systems to dose coagulant, such as ferric sulfate or ferric chloride. A novel approach is to dose lime to achieve softening. The application of lime for softening is not the same as dosing ferric salts. By combining both the lime dosing and membrane treatment, it will be possible to reduce the lime dosage and sludge production, and achieve the desired product water quality at reduced cost. The membrane utrafiltration is a barrier that can physically prevent microorganism from passing through into the treated water. The existing lime softening facilities can be integrated with ultrafiltration. Additional minor modification will increase the existing plant's capacity.

The initial research [1] was conducted to demonstrate the feasibility of combining chemical precipitation specifically the addition of lime with membrane ultrafiltration to achieve the water quality required in various manufacturing and industrial processes with minimum usage of chemicals and eliminating continuous chlorination of the raw water.

2. Treatment processes

2.1. Precipitative lime treatment

Lime treatment or softening has been long recognized as an effective process to reduce calcium and magnesium hardness in water by adding CaO or Ca(OH), (lime) and/ or Na₂CO₃ (soda ash) to precipitate calcium as CaCO₃ and magnesium as Mg(OH)₂. It will also remove CO₂ in the water. Other impurities such as iron, manganese, fluoride, phosphates, heavy metals, silica, chloride and total dissolved solids in the water are also removed with the addition of lime alone or in combination with other chemicals such as alum, sodium silicate, ferric and ferrous salts, flocculant, etc. The elevated pH required in the process also inactivates many microorganisms. Lime softening has been known to remove natural organic matter (NOM) in water specifically trihalomethane (THM) precursors [2]. Lime softening was found to remove significant fraction of fulvic acid extracted from ground water [3], and the NOM removal was achieved by the adsorption onto calcium carbonate and magnesium hydroxide formed in the process. EPA [4] recommendation to enhanced total organic carbon (TOC) removal using precipitative softening is to provide the conditions that favor the formation of magnesium hydroxide and small calcium carbonate particles. This can be achieved by elevating the pH to 10.8 or higher, delaying carbonate addition and sludge recycling. The degree of precipitation of calcium, magnesium and other impurities depends on the operating pH. Soda ash is also added to precipitate non-carbonate hardness and to precipitate excess lime. Caustic soda is also added to adjust the operating pH and promote precipitation of calcium and magnesium. This process is applicable if there is enough calcium in the raw water to complete the softening reactions. The typical reactions in lime, or similar precipitative softening processes are:

Lime as CaO when water is added becomes Ca(OH)₂

 $CO_2 + Ca(CO)_2 = CaCO_3 + H_2O$

At pH 9.5 or above the following reaction will occur:

 $Ca(HCO_3)_2 + Ca(OH)_2 = 2CaCO_3 + 2H_2O$ $Mg(HCO_3)_2 + Ca(OH)_2 = CaCO_3 + MgCO_3 + 2H_2O$

At pH 11 or above

 $MgCO_3 + Ca(OH)_2 = CaCO_2 + Mg(OH)_2 + Ca(OH)_2$ (excess)

Reactions with soda ash

 $Ca(OH)_2 + Na_2CO_3 = CaCO_3 + 2NaOH$ $CaSO_4 + Na_2CO_3 = CaCO_3 + Na_2SO_4$ $\begin{array}{l} CaCl_{2} + Na_{2}CO_{3} = CaCO_{3} + 2NaCl \\ Ca(NO_{3})_{2} + Na_{2}CO_{3} = CaCO_{3} + 2NaNO_{3} \\ MgSO_{4} + Na_{2}CO_{3} + Ca(OH)_{2} = CaCO_{3} + Mg(OH)_{2} + Na_{2}SO_{4} \\ MgCl_{2} + Na_{2}CO_{3} + Ca(OH)_{2} = CaCO_{3} + Mg(OH)_{2} + 2NaCl \\ Mg(NO_{3})_{2} + Na_{2}CO_{3} + Ca(OH)_{2} = CaCO_{3} + Mg(OH)_{2} + 2NaNO_{3} \end{array}$

Other reactions when coagulant is added are:

 $\begin{array}{l} Al_{2}(SO_{4})_{3} + 3Ca(HCO_{3})_{2} = 2Al(OH)_{2} + 3CaSO_{4} + 6CO_{2} \\ Al_{2}(SO_{4})_{3} + 3Na_{2}CO_{3} + 3Ca(OH)_{2} = 2Al(OH)_{2} + 3Na_{2}SO_{4} + 3CaCO_{3} \\ Fe_{2}(SO_{4})_{3} + 3Ca(HCO_{3})_{2} = 2Fe(OH)_{2} + 3CaSO_{4} + 6CO_{2} \\ Fe_{2}(SO_{4})_{3} + 3Na_{2}CO_{3} + 3Ca(OH)_{2} = 2Fe(OH)_{2} + 3Na_{2}SO_{4} + 3CaCO_{3} \end{array}$

In typical lime softening, additional and/or excess chemicals are often added to increase the mass of sludge to promote settling.

In addition to the lime and/or soda ash reactions described above, addition of excess lime and operation at pH of 11.2 and above, will precipitate the dissolved silica in a process called ultra high lime (UHL) treatment [5].

$$Ca(OH)_2 + SiO_2 = CaH_2SiO_4$$
 at pH 11 to 12

The ultra high lime with aluminum (UHLA) process can also be used in reducing chloride and sulfate in water [6]. The ultra high lime treatment is also effective in treating cooling tower blowdown to minimize the makeup water supply and generation of wastewater [7]. The ultra-high lime treatment has been used successfully in treating RO brine reject in a zero liquid discharge (ZLD) plants [8].

2.2. Limitations/problems associated with lime treatment

The lime softening although reliable and being used in various industries, has its limitations and problems. Some of the limitations and problems associated with lime softening are:

- Disposal of large amount of sludge generated by the process
- Requires larger plant area for the lime reactor, as well as the sludge handling equipment, lime preparation and storage facilities.
- Additional chemicals are required to promote settling of the sludge and solids.
- Requires media filtration after clarification.
- The lime softening plant should be continuously running and requires longer time to stabilize after startup.
- The lime-soda softening is more expensive compared to other competing processes.
- There are very limited companies now specialized in the manufacture of lime softening systems.

2.3. Ultrafiltration

Ultrafiltration is a pressure driven membrane process, where source water is passed through a membrane with nominal pore size of 0.01–0.1 µm. Suspended solids, colloidal particles, bacteria and other particles are retained by the UF membrane. Ultrafiltration also removes high molecular weight organic matter. The typical ultrafiltration membranes have a typical molecular cut-off of 150,000 Daltons (1 Dalton or Da = 1/12 mass of one atom of Carbon-12), however through the addition of coagulants, it can effectively remove organic matter with molecular weight down to less than 20,000 Daltons. The addition of coagulant in the form of ferric salts, poly aluminum chloride or alum is common in ultrafiltration process. The addition of lime in the feed of ultrafiltration membrane was never been reported in the literature, but there were published reports integrating pellet softening with UF membrane treatment [9]. In most membrane processes especially in reverse osmosis and nanofiltration, CaCO₃ scaling is a common problem. In treating hard water using ultrafiltration, the precipitation of CaCO₃ can be a problem, especially occurring in capillary or small diameter tubular UF membranes. The development of the Spirasep UF membrane as shown in Fig. 1, which air-scoured immersed membranes in spiral configuration developed by Trisep, will minimized the build up of scale in the UF membrane surface. Compared to RO or NF, there is no change in salt concentration in the membrane surface, therefore formation of scale will be minimized. The Spirasep membrane is similar in appearance to 8" diameter × 40" length RO membrane, made of polyether-sulfone, and with effective membrane area of 178 ft². The operating pH is from 4 to 11 on continuous basis and pH of 2 to 12 for cleaning. The Spirasep membrane has chlorine tolerance of 2,000 mg/L.

In the research the manufacturer's operating guidelines [10–12] were strictly followed because the UF unit is a working commercial unit with single UF element. Among the operating conditions maintained were the following:



Fig. 1. Spiral wound membrane.

- Continuous aeration at the recommended aeration rate of 0.02–0.05 scfm per 1 ft² of membrane area. Continuous aeration was recommended for water with high suspended solids concentration.
- Backflushing was set every 15 min with 30 s duration at the rate of 45 gfd.
- Transmembrane pressure was defined and measured as per the membrane manufacturer's guidelines.

3. Materials and methods

3.1. Experimental plan

The objective of the experiment is to demonstrate the applicability of combining lime softening with ultrafiltration membrane to produce water that meet specific treatment objectives:

- Reduce hardness in the feed water and, at the same time, maintaining alkalinity of the treated water to less than 50 mg/L as CaCO₃
- Determine the relationships of operating pH and membrane flux with trans-membrane pressure and membrane permeability.
- Compare the lime dosage in this research with the lime dosage used in conventional lime softening plant, treating similar water source.
- Evaluate the operating parameters important in designing a full scale plant. These include membrane flux rates, permeability, recovery, backwash intervals, cleaning intervals, and transmembrane pressure.

3.2. Pilot lime softening ultrafiltration unit

The pilot lime softening ultrafiltration system is a full scale commercial operating plant with a lime reactor and one SpiraSep UF membrane immersed in a reactor tank.

The lime reactor is a polyethylene cylindrical conical bottom tank, with maximum capacity of 200 gallons, to allow 30 min retention at the maximum flow of 5.7 gallons/min (gpm). The tank was provided with discharges at three different levels for the different flow rates. The elevation of the lime reactor is adjustable, in order to allow gravity flow into the membrane reaction tank. The lime solution or slurry was fed by a peristaltic metering pump, with a maximum capacity of 2.3 gallons/h (gph). The flow rate of the metering feed pump was controlled by the pH transmitter. The lime slurry or solution was fed to the incoming raw water into the mixing chamber which directed the flow to the bottom of the lime reactor. The mixing chamber was provided with a mixer driven by a 1/20 hp electric motor. The precipitate, or sludge, settles at the bottom of the tank. Sludge was expected to be carried over to the membrane reactor tank. The bottom of the lime reactor was provided with a connection for pumping out the sludge at scheduled interval. The pH sensor was installed at the inlet of the membrane reactor tank.

The SpiraSep UF membrane manufactured by Trisep Corp. is an immersed, negative-pressure ultrafiltration process, which will remove suspended solids, turbidity, viruses, bacteria, and some organic compounds. A typical SpiraSep system consists of an array of spiral wound elements submerged inside a process tank as shown in Fig. 2. The membrane elements are attached to a manifold assembly, consisting of a central permeate header with an array of membrane permeate ports, which connects to the SpiraSep membrane. A vacuum is generated by the suction of a centrifugal pump, creating the necessary net drive pressure to "pull" water through the SpiraSep membrane. Air is bubbled up through each membrane element via bubble diffusers, creating tremendous shear forces on the membrane surface that remove any suspended solids as shown in Fig. 4. A small amount of a coagulant is injected into the process influent. The enhanced coagulation process will help reduce organic fouling and improve TOC and color reduction.

Periodically (on a timed basis), permeate water is reversed through the membrane, or back flushed, to help further remove the accumulated suspended solids as shown in Fig. 3. This process also introduces a small amount of disinfectant to help control the microbial activity on the membrane surface. Concentrate is removed from the process tank, and is typically less than 10% of the influent rate. SpiraSep membranes can also be chemically cleaned through one of two processes: a periodic flux enhancement (PFE) or a flux recovery clean (FRC) procedure.

The pilot plant was manually controlled and operated with several automated features, such as backwashing. The automation is controlled by a process logic controller (PLC). Feed from a pressurized source is delivered to the UF system, and is controlled by a feed control valve. The blower was operated continuously to de-



Fig. 2. SpiraSep immersed UF membrane configuration.

liver pressurized atmospheric air to the membrane element. Membrane backwashing was controlled by a timer, and was performed on a timed basis. Membrane cleaning is operator initiated.

The filtrate pump flow rate was adjusted manually with the permeate control valve. The UF membrane was back flushed at set interval. The water required for the membrane back flush was taken from the UF filtrate tank and pumped to the membranes using a separate backwash pump. The backwash pump reverses the flow of water through the UF membranes. A membrane back flush was performed every 15 min for 30 s and is automatically controlled by the PLC. Once filtrate production started, timers for the back flush frequency and periodic flux enhancement (PFE) are started. The blower remains on running at the manually set value.

When a back flush sequence is started, the automatic feed valve was closed, and the filtrate pump and blower were automatically turned off (concentrate valve remains open). UF filtrate water and chlorine were then backflushed through the membrane for a period of about 30 s. A variable frequency drive (VFD) adjusts the back flush



Fig. 3. SpiraSep UF membrane in backflushing mode.



The spiral design open feed channel provides for:

 Efficient distribution of air scour and

(2) Effective air scour via direct controlled contact with the membrane surface by the air scour

Fig. 4. Spirasep UF membrane air scour.

pump speed, to the manually set value. Output of the metering pump was manually adjusted. Excess water introduced to the tank was removed via a tank overflow and/or concentrate line. Once the back flush sequence was completed, the back flush pump and chlorine metering pump were automatically turned off. The blower was turned on and allowed to operate for 10–15 s before the filtrate pump was restarted and the feed valve opened to allow normal filtrate production.

The UF membrane was continuously aerated to prevent and minimize membrane fouling. A blower takes atmospheric air and bubbles them up through individual membrane module via an aeration disc. The blower was operated using a VFD, and the motor speed is set manually. The operation of the blower was controlled by the PLC. Air was delivered to the UF membrane through a coarse bubble diffuser. The air diffuser was attached to an aeration pipe. The aeration pipe contains a manual flow control valve and air flow indicator to ensure proper air flow.

Various chemicals were dosed for various system operations. Chlorine is normally dosed during each back flush, in addition to PFE and clean-in-place (CIP) processes. During the pilot testing, chlorine solution was not dosed. Sodium hydroxide was injected for PFE and CIP processes. Citric acid was dosed for PFE and CIP processes. The flow rates of the chemical dosing pumps were set manually. Operation of the chemical dosing pumps during backwash, PFE, and CIP was controlled by the PLC.

Operating performance can be optimized through the use of PFE. A chemical solution was backwashed through the membranes in-situ to perform a quick chemical treatment. This process was performed while the membrane tank was filled with process water, requiring approximately 20–30 min. This was done on a daily or every two days. When a PFE process was initiated, the feed valve was closed, and the filtrate pump and blower were turned off. UF filtrate and chemicals were then automatically back flushed through the membranes while they are still immersed in the feed water (i.e. membrane tank is not drained for this process). Excess water introduced to the tank was removed via a tank overflow and/or concentrate line.

During membrane cleaning, a cleaning solution was back flushed through the membranes until the filtrate tank was completely filled. The membrane was statically soaked in the cleaning solution for approximately 4–8 h. A CIP process is typically performed once every 3 months when treating municipal water. Actual CIP frequency is determined through pilot testing and actual plant operation. CIP is a manual operation. In high suspended solids environment like in lime softening CIP every 2–3 weeks is acceptable. The UF system is normally designed to allow the membrane elements cleaned in place in the membrane tank. UF filtrate and cleaning chemicals are back flushed through the membranes until the CIP tank is completely filled. At the end of the chemical soak, the tank is drained and then refilled.

3.2. Chemicals

The chemicals used in the pilot test were:

- Hydrated lime, Ca(OH)₂, 93%, CAS 1305-78-8, technical grade. A 3.2% lime slurry was prepared by adding 32.24 g of hydrated lime (93% Ca(OH)₂) per l of water mixed into the slurry tank. The 3.2% lime slurry has a specific gravity of 1.020 or 2.84 Baume,
- Sodium hypochlorite, NaOCl, 12% chlorine, CAS 7681-52-9. The sodium hypochlorite (12% chlorine) was dosed at 10 mg/L during back flush and 100 mg/L during periodic flux enhancement (PFE). The sodium hypochlorite solution for both the back flush and the PFE back flush were dosed by metering pumps drawing directly from the sodium hypochlorite container
- Sodium hydroxide, NaOH, 45%, CAS 1310-73-2. Caustic soda, 45% solution was dosed at 0.1% or 1,000 ppm using chemical feed pump at a rate of 0.63 l/h drawing directly from the caustic soda container
- Citric acid anhydrous 99.5%, C₆H₈O₂, CAS 77-92-9. The citric acid crystals were dissolved in water at 200 g/L solution. From this stock solution, the citric acid was dosed directly to the PFE back flush line at rate of 2 l/h during CIP. The citric acid was dosed to the CIP line at the rate of 20 l/h.

3.3. Experimental procedures

The pilot unit process flow diagram is shown in Fig. 5. The pilot unit was initially operated for one week without any chemical addition to stabilize the flow and calibrate the instruments. After one week the pilot unit was operated for approximately one month with varying dosage of lime to determine the conditions that can provide the desired water quality. The pilot unit was operated for another month at the selected optimum operating conditions. The lime slurry was dosed by peristaltic pump with maximum capacity of 1.25 gph (4.73 lph). This pump is capable of delivering lime up to 346 mg/L when operating at flux of 15 gfd and 120 mg/L when operating at 45 gfd. Operating flux of 15 gfd was selected to be the starting flux, based on previous pilot testing using other coagulants such ferric chloride, ferric sulfate and alum. Trisep recommend the following sustainable flux rates: For municipal secondary effluent: 15–18 gfd; municipal drinking water: 25 gfd; landfill leachate (with chemical precipitation): 15 gfd. The flux was eventually increased to 30, and 45 gfd. Lime slurry was dosed to achieve pH of 8.3, 9.4, 10.6 and 11.2 at the lime reactor overflow or discharge to the membrane reactor tank.

The flux was set by controlling the flow through the filtrate pump through the adjustment of the filtrate control valve. During the test the trans-membrane pressure



Fig. 5. Process flow diagram of pilot unit.

(TMP) was monitored through a digital pressure indicator connected to a pressure transmitter installed at the manifold between the UF membrane filtrate discharge and the suction of the filtrate pump.

The pilot testing log included the following information: Date and time, actual flow rate reading, total flow (from flow totalizer), pH, temperature, raw water and filtrate turbidity, TMP or UF pump suction line pressure located at the same level as the water in the UF reactor tank. The net flow in each segment of test can be determined and used as basis of calculating the average permeability. The flow was indicated by a flow transmitter with digital flow indicator and totalizer, receiving signal from a flow sensor. The pH is indicated by a pH transmitter with digital pH and temperature indicator, receiving signal from a pH probe. The TMP was measured by local mounted pressure transmitter/ indicator. The turbidity was continuously monitored by a low range process turbidimeter, provided with sample connections to allow turbidity measurement of either the raw water or the filtrate.

The permeability was plotted against elapsed time. The permeability was calculated as flux (in gfd) divided by the transmembrane pressure (psi). The permeability has a unit of gfd/psi. The TMP values were also plotted against time.

Composite samples of feed and filtrate were taken daily and were analyzed for pH, alkalinity, calcium and magnesium hardness, conductivity, turbidity, and total organic carbon (TOC). Sample of the water in the membrane reactor was also taken for suspended solids analysis.

3.4. Analytical procedures

The analysis of the water samples were made following the EPA Methods and Standard Methods for the Examination of Water and Wastewater [13]. The water samples taken during pilot testing were sent to Severn Trent Laboratories, Inc. (STLI) in Tampa for analysis. Chemical analyses were also conducted on site using Hach test kits for verification and calibration of instruments. Water analysis was also conducted in the nearby beverage plant's laboratory, for comparison. The parameters measured and/or analyzed are:

- pH and temperature
- Alkalinity
- Calcium and magnesium hardness
- Turbidity
- Total suspended solids
- Total organic carbon

4. Results and discussion

The pilot testing was divided into three phases. The first phase was to stabilized the flows and calibrate control valves and instruments. The first phase started on October 15, 2005, and was supposed to last one week, however it was extended by one more week, due to mechanical and instrument problems. The second phase was performed at varying flux and pH conditions. It started on October 29, 2005 and lasted four weeks. The objective of the third phase was to simulate the operation in the beverage plant, based on the data obtained from the second phase. The third phase started on December 2, 2005 and ended on January 5, 2006. The source of feed water during the test was city of water supply.

4.1. Initial operating conditions without chemical addition

The purpose of running the pilot unit at different flux levels, without the addition of chemicals, is to determine the flow characteristics of the unit and to calibrate the instruments. Based on the UF membrane area of 178 ft², the filtrate flow rates of 1.9, 3.7 and 5.6 gpm corresponded to flux values of approximately 15, 30 and 45 gfd. The pressure indicator in the suction line of the UF permeate pump provide reading down to increments of 0.1 psig. pH and temperature were also continuously displayed. The pressure measured on the UF membrane filtrate discharge and suction of the UF permeate pump pipework is the trans-membrane pressure. The location of the pressure sensor was in the same level as the water level in the UF reactor tank as recommended by the membrane manufacturer. This eliminated the need for correcting for the difference in hydraulic heads. The vacuum pressure reading can be considered as the trans-membrane pressure. The filtrate flow was controlled by globe valve. After the flow and pressure readings were stabilized, the pilot unit was operated with varying flows of 1.9-5.6 gpm. The back flushing was set every 15 min for duration of 30 s. It was expected that the TMP will increase prior to back flushing. During the initial run at 1.9 gpm, the TMP remained at -0.5 psi, before and after back flushing throughout the 2 days of operation. At the flow of 3.7 gpm, the TMP stayed consistently at -1.1 psi after back flushing, and the pressure before back flushing was -1.5 psi. When operating at 5.6 gpm, the TMP after back flushing was -1.7 psi and -2.0 psi before back flushing. Water samples were taken for analysis. Raw water analysis is shown in Table 2. The average pH of the feed water is 7.3 and the water temperature ranges from 20 to 25°C. Chlorine was not dosed during back flushing and during PFE.

4.2. Operation at varying flux and pH

The second phase of the pilot testing was the addition of lime to achieve operating pH values of 8.3, 9.4, 10.6 and 11.2, at flows of 1.9, 3.7 and 5.6 gpm (or flux of 15, 30 and 45 gfd). The pilot unit was operated continuously for 2 days for each flow condition. The pH was set to the desired operating pH and the chemical feed pump automatically dosed the required lime solution. The average TMP values before and after back flushing are shown in the Table 5. The flux and permeability values at different operating conditions are shown in Table 6. The Permeability Profile at various operating conditions is shown in Fig. 6. The permeability values range from 50% to 85% of the clean water permeability for SpiraSep UF membrane, which is 35 gfd/psi. Fig. 7 shows the TMP profile during the test. It can be observed, TMPs tends to increase with increasing flow (or flux) and operating pH.

Composite raw water and filtered water samples were analyzed for Ca, Mg, Alkalinity, pH, turbidity, and TOC.

Table	e 4	
Raw	water	analysis

	10/15/05	11/12/05	12/10/05
рН	7.31	7.5	7.3
Alkalinity, mg/L CaCO3	76	70	75
TOC, mg/L	3.8	4.0	3.6
Ca, mg/L CaCO ₃	65	60	62
Mg, mg/L CaCO ₃	4.2	4.5	4.2
Turbidity, NTU	0.1	0.1	0.1

120

Table 5 Average vacuum pressures or TMP values before and after UF back flushing at various flux values

Table 6

Flux vs. permeability at various operating pH

pН	Flux values		
	15 gfd	30 gfd	15 gfd
7.3	–0.4/–0.5 psi	–1.1/–1.3 psi	–1.8/–2.0 psi
8.3	–0.6/–0.8 psi	–1.2/–1.5 psi	–1.9/–2.2 psi
9.4	–0.6/–0.8 psi	–1.3/–1.8 psi	–2.2/–2.6 psi
10.6	–0.7/–1.0 psi	–1.6/–2.2 psi	–2.4/–2.8 psi
11.2	-0.6/-1.0 psi	–1.7/–2.3 psi	–2.6/–3.4 psi

рН	Flux valu	Flux values			
	15 gfd	30 gfd	45 gfd		
7.3	29.6	26.29	26.12		
8.3	25.81	25.31	23.95		
9.4	25.14	23.03	20.43		
10.6	23.0	19.32	19.21		
11.2	21.4	19.05	18.17		

Note: After BF/Before BF

Table 7



Fig. 6. Permeability profile at various operating conditions.

Analysis of water samples at various operating conditions

Note: Permeability is gfd/psi



Fig. 7. TMP profile at various operating conditions.

Operating pH	7.3	7.3	8.3	9.4	10.6	11.2
pH of the sample	7.31	7.31	8.06	9.2	10.3	10.8
Type of water	Raw	Filtrate	Filtrate	Filtrate	Filtrate	Filtrate
Alkalinity, mg/L CaCO ₃	76	76	62	30	36	36
TOC, mg/L	3.8	3.8	3.7	3.5	3.3	3.2
Ca, mg/L as CaCO ₃	65	65	57	38	41	56
Mg, mg/L as CaCO ₃	4.6	4.6	4.5	4.3	3.9	2.4
Turbidity, NTU	0.1	0.05	0.05	0.05	0.05	0.05

The results of the water analysis are shown in Table 7. Grab water sample from the membrane reactor was also taken for total suspended solids analysis.

An analysis of water sample was also conducted in the beverage plant laboratory and shown in Table 6. Note that there is difference between the operating pH value and the pH of the filtrate analyzed in the laboratory. The pH of the filtrate was expected to change due to the effect of aeration in the UF tank which tends to strip the CO₂ or add CO₂ from the air. Aeration has stabilizing effect on the filtrate. During the test the amount of lime in each run was not monitored, however every time a batch was prepared, the quantity was recorded.

From the tables above, it can be noted that there is a significant reduction of alkalinity and hardness, whereas at pH 10.6, the alkalinity and hardness increased. At pH 10.6 and above, the increase in alkalinity and calcium was due to the lime addition. The magnesium concentration continues to drop as the pH went up as expected.

The dilute sludge that accumulates at the bottom of

the membrane reactor tank can be manually drained, when the unit stopped. During backwashing, the water in the membrane reactor overflows to lime reactor tank. The concentration of the suspended solids in the membrane reactor is shown in Table 7. It was observed that there was slight change in the sludge concentration when operating pH changed as shown in Table 9. The concentrated sludge that accumulated at the bottom of the lime reactor tank was pumped out using another rotary flexible impeller pump rated at 0.25 gpm. Usually, 1/3 of the sludge in the conical section of the lime reactor tank was drained when the volume of sludge reaches the top of the conical section.

4.3. Operation at pH 9.4-9.8

The next phase of the test was to simulate the operation in an actual beverage plant condition. Operation at pH 9.4-9.8 was chosen because the results in the previous tests satisfied the water quality treatment objective using the same source water as used in this test, although the actual operating pH was slightly higher. The resulting alkalinity level was favorable to their operation. The flux selection of 30 gfd (or flow of 3.7 gpm) was based on the following factors: economics, previous experience for similar application, results from the second phase of the test, and guideline of the membrane manufacturer. The test also predicted the intervals between cleaning and estimated the consumption of lime. Water samples were taken and analyzed. The amount of lime used was also monitored. This test lasted for over 30 days. Fig. 8 shows the permeability profile and Fig. 9 shows the TMP profile throughout the duration of the test period. On the 18th day of test the TMP has almost doubled and the permeability dropped to down to 50% from the first day value. Based on experience, when this condition occurs, it is necessary to chemically clean the UF membrane. The cleaning was made as per the CIP procedure recommended by the membrane manufacturer. After cleaning the TMP and permeability values were restored to the first day values. The operation of the pilot unit was continued for another 10 days after cleaning. The TMP and permeability profile after cleaning is similar to the initial profile. The analysis of the filtrate by STLI and in the beverage plant laboratory are shown in Tables 7 and 8.

The concentration of suspended solids in the membrane reactor tank was maintained at 600–700 mg/L range. Backflushing seemed to maintain constant solids concentration in the membrane reactor. During backflushing, the excess water flowed back to the lime reactor tank, carrying suspended solids, and the backwash water diluted the water in membrane reactor. The sludge from the membrane and lime reactor tanks was manually drained.

Table 8

Analysis of the filtrate in the beverage plant laboratory

Operating pH	7.3	9.45	9.6	9.8
pH (Lab)	7.3	8.49	9.65	9.14
Phenolphthalein alkalinity,	4.2	9.6	23.3	14.8
mg/L CaCO3				
Methyl Orange alkalinity,	88.1	35.8	33.7	26.8
mg/L CaCO3				

Table 9

Average suspended solids concentrations in the membrane reactor

Operating pH	Suspended solids conc., mg/L
7.3	10
8.3	580
9.4	600
10.6	600
11.2	680



Fig. 8. Permeability profile at beverage plant operating conditions.



Fig. 9. TMP profile at beverage operating conditions.

5. Summary and conclusions

5.1. Alkalinity reduction

Alkalinity reduction to less than 50 mg/L or to the preferred level of 20-30 mg/L and maintenance of desired phenolphthalein alkalinity and methyl orange alkalinity ($2^{*}P_{alk} - MO_{alk} = 2-7$) can be achieved continuously in the lime softening UF unit with relatively simpler control, operation and maintenance compared to conventional lime softening process. The lime softening UF unit can be started in a matter of minutes, unlike the conventional lime softening which requires hours or days to build up of the sludge blanket before stable operation is achieved. The lime dosage during the third phase of test (operating pH = 9.8) was 70 mg/L, based on raw water alkalinity concentration of 76 mg/L and pH of 7.3 and the filtrate alkalinity and pH are 26.8 mg/L and 9.18 respectively. The theoretical or calculated dosage using the Rothberg, Tamburini, and Windsor model [14] was 65 mg/L. The lime dosage in the beverage plant was in the range of 120-130 mg/L operating at pH of 9.8-10.2 with ferric chloride addition.

5.2. UF filtrate turbidity

The turbidity of the filtrate was consistently observed to be in the range of 0.04–0.05 NTU throughout the duration of the test. The filtrate turbidity was not affected by the incoming feed water turbidity. When the pilot unit was operated without the lime addition, the feed water and filtrate turbidity were 0.1 NTU and 0.05 NTU, respectively. The suspended solids concentration in the membrane reactor tank throughout the test was in the range of 580–650 mg/L. Table 9 shows the average suspended solids concentration in the membrane reactor.

5.3. Transmembrane pressure (TMP) vs. pH and flux

The increase in flux results to corresponding increase in TMP, however as the operating pH increases, the rate of TMP increases as shown in Figs. 11 and 13.

5.4. Permeability

The operating pH vs. permeability profile shown in Fig.10 indicates, the permeability decreases with increasing operating pH. The TMP vs. flux profile shown in Fig. 12 indicates that permeability decrease with increasing flux. The decline in permeability during the second phase of the test was due to the increase in operating pH. The starting and ending average permeability values were 31.25 gfd/psi and 17.53 gfd/psi. The prolonged operation without CIP had not impacted the permeability, because when the third phase of the test started, the



Fig. 10. Permeability vs. operating pH at various flux rates.



Fig. 11. TMP vs. operating pH at various flux rates.



Fig. 12. Permeability vs. flux at various operating pH.

starting average permeability during the first 2 days of operation was 26.93 gfd/psi, which is comparable to 25.5 gfd/psi when the operation started in second phase of the test at pH 9.4.

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Fig. 13. TMP vs. flux various operating pH.

5.5. Total organic carbon (TOC)

The data in Table 7 indicate that there was no reduction in TOC when the pilot unit was operated without lime addition. With the addition of lime, there was a reduction of TOC. The reduction in TOC ranged from 2.6% to 15.8%, when the pilot unit was operated at various pH values.

5.6. Hardness reduction

Table 7 indicates the reduction in Ca and Mg hardness which was expected as a result of the increase in operating pH.

5.7. Operating flux

The operating flux of 30 gfd was initially selected because most of the ultrafiltration membranes used in treating municipal operate at this flux value, although Trisep recommendation is 25 gfd for treating municipal water supply, when dosing coagulants (such as ferric chloride or sulfate, alum and polyaluminum chloride). It was assumed that lime will behave like the other coagulants although there were concerns of excessive fouling and scaling. The results of this research confirmed that the immersed SpiraSep UF membrane can achieve the treatment objectives when operated at flux of 30 gfd, and fed with lime treated water at pH 9.8, with suspended solids concentration of 600 mg/L. The cleaning of the membrane or CIP was initiated when the TMP value was doubled, which correspond to about 50% of clean membrane permeability. The CIP was conducted after 19 days of operation, noting that the pilot unit has been in operation for over 30 days in the first and second phases before the third phase started. The third phase of the test also confirmed the following: the cleaning procedures and chemicals effectively restored the membrane to its starting TMP and permeability; by extrapolating the permeability and TMP profiles the expected next cleaning will be after 48 days. This corresponds to 30 days cleaning interval.

5.8. Chlorination

During the entire duration of test, chlorine was not added to the back flush water or in the PFE. The residual chlorine in the feed water ranged from 0.2 to 0.7 mg/L. Chlorine was dosed only during CIP and when the unit was stopped longer than 24 h.

5.9. Benefits of the integrated lime softening ultrafiltration (ILSUF) process

The benefits of the integrated lime softening ultrafiltration process, based on the results of this study can be summarized in the following:

- There is considerable economic benefit when the conventional treatment processes comprising of chlorination, lime softening, clarification, and filtration, is replaced with ILSUF comprising of a single equipment with smaller footprint. With less equipment, operation and maintenance will be simpler.
- The LSUF process requires shorter time for start-up, unlike conventional lime softening which requires time to build up sludge, stabilize the flow and attain the desired treated water quality.
- The ILSUF process produces less sludge and dirty backwash water. It can be operated at relatively lower pH and with no addition of ferric chloride which significantly reduced the volume of sludge. The water during backflush operation can be returned back to the system. The water wasted is the water that goes with the waste sludge, which is minimal.
- Continuous chlorination of raw water can be eliminated, reducing the formation of the THMs.
- Process control in ILSUF reduced to adjustment of pH and flows. The process is less sensitive to temperature.
- In ILSUF process, the sludge removal is simplified because there is no sludge blanket to maintain.
- The ultrafiltration process provides physical barrier for microorganism and particles, minimizing the contamination in the down stream processes.
- Existing lime softening plants can be retrofitted and their rated capacity can be increased with just the addition of the UF system processes.

5.10. Other applications of integrated lime softening ultrafiltration process

The possible applications of the ILSUF process in process water and wastewater:

- Any lime softening plants that can either be stand alone plants or pre-treatment for additional processes. As stand alone, it can be municipal softening plant, manufacturing plant or as part of Advanced Wastewater Treatment for secondary sewage effluent.
- Pre-treatment for reverse osmosis to increase its recovery as part of water re-use or recycle processes, brine reject reduction, and zero liquid discharge (ZLD)

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when treating water or wastewater. ZLD becomes attractive when there are limitations in the volume of liquid waste discharge or the absence of receiving water.

- Pre-treatment of feed to thermal evaporation processes will reduce scaling, increase recovery, and improve the overall performance of the plant.
- The use of secondary treated sewage as the only source of feed water for industrial and power plants for the production of high quality or ultra pure water is increasing as result of restriction in the use of potable water for industrial applications.
- Recent study by the author indicated possibility of treating seawater RO feed with lime to precipitate calcium and magnesium salts, as CaCO₃ and Mg(OH)₂ respectively. This process will also elevate the pH > 10.3 operating the seawater RO at higher pH will eliminate the need for second pass RO unit for the removal of boron.

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