Chemical Optimization for a New Brackish Groundwater Reverse Osmosis Treatment Plant

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Drinking water plants in Florida that use brackish groundwater and surface waters rely on reverse osmosis for treating these water sources. The typical process train utilized in the state uses chemical pretreatment, cartridge filtration, reverse osmosis (RO), degasification, disinfection, and corrosion control (see Figure 1 for an example process flow diagram).

Pretreatment chemicals are used to manage scaling in the RO membranes and reduce the pH to optimize hydrogen sulfide stripping in the degasifiers. Post-treatment chemicals are used to perform primary disinfection and to adjust the pH and alkalinity for corrosion control.

The traditional method of RO pretreatment is to use sulfuric acid and a scale inhibitor to suppress the pH of the feedwater prior to the RO membranes to avoid scaling. The pH remains suppressed entering the degasifier at the optimum pH for hydrogen sulfide stripping. Following recent developments in reverse osmosis elements and manufacturer changes in scale inhibitors, an approach is to adjust pH only prior to degasification, thus maintaining an ambient pH through the RO trains and relying on the scale inhibitor to control scaling in the membranes.

The city of Cape Coral is constructing a new drinking water treatment plant, the North RO Water Treatment Plant (see Figure 2 for current construction status). The utility currently operates one of the oldest reverse osmosis plants in the United States, the Southwest RO Water Treatment Plant.

As part of the design of the new North RO Plant, an evaluation of the pretreatment requirements (and chemical optimization through the entire plant) was completed. The starting point, or basis for this evaluation, was the Southwest RO Plant.

The current practice of this plant is to reduce the pH to 6.0 with sulfuric acid prior to the RO membranes with addition of scale inhibitor, without further pH adjustment through the process, until sodium hydroxide (or caustic soda) is added for alkalinity and pH adjustment for post-treatment. Alternatives to this method were considered for the North RO Plant and are presented in this paper.

Water Chemistry

Pretreatment of RO membranes is focused on chemical and physical methods to
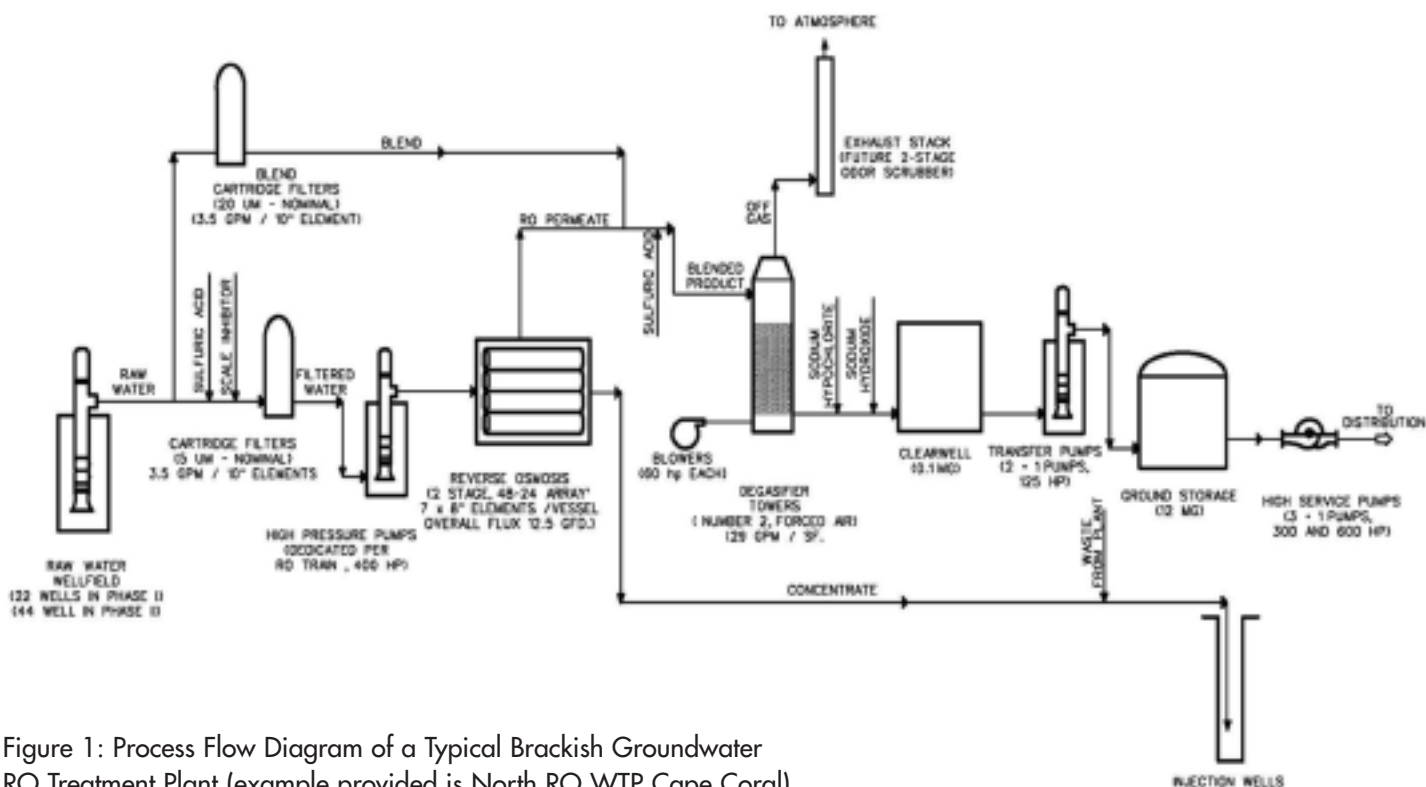
Lead author Karla Kinser, P.E., is the national membrane technology practice leader for MWH Americas, a construction, engineering, construction, and management services firm. She is located in the company's Denver office. Shawn Kopko is the water production superintendent for the city of Cape Coral. Andy Fenske is a chief operator with the city. Gert-Jan Schers is a principal engineer in the Cape Coral office of MWH Americas.

Figure 1: Process Flow Diagram of a Typical Brackish Groundwater RO Treatment Plant (example provided is North RO WTP Cape Coral)

reduce scaling and fouling from scalants and particulates, respectively. Physical methods for particulate removal will not be discussed in this article, but they consist of 5 micron (nominal) cartridge filtration.

Scaling of the RO membrane is caused by the concentration of sparingly soluble salts on the feed side of the membrane, as water passes through to the permeate side and concentrates the salts into the concentrate. Sparingly soluble salts will exceed their solubility products (Ksp) and precipitate in the concentrate onto the membrane surface; therefore, it is the accumulation of the salts in the concentrate that is of concern for the system.

At 80 percent water recovery, the concentration of salts in the concentrate is approximately five times the feed concentration. The main constituents of concern are calcium carbonate, sulfate salts such as barium sulfate, calcium fluoride, and silica.

Table 1 presents a summary of water quality parameters in the raw water in 2008 and projected to the year 2033, which is the design life of the plant. The aquifer is expected to become more saline over this 25-year period, based on groundwater modeling of the aquifer.

Calcium Carbonate Scaling Control

For concentrate total dissolved solids (TDS) concentrations of 10,000 mg/L or less, the concentrate Langelier Saturation Index (LSI) is used to control calcium carbonate scaling. For concentrate TDS concentrations above 10,000 mg/L, the Stiff Davis Index (SDI) is used.

The concentrate TDS level for the North Plant will be just below 10,000 mg/L for the first 10 years, and as the raw water salinity increases, the concentrate TDS will increase to above 10,000; however, since the concentrate TDS concentration is very close to 10,000 throughout the life of the plant, RO manufacturers recommend using the LSI to control scaling.

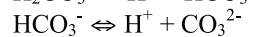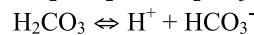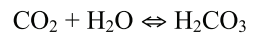The calcium carbonate scaling potential can best be illustrated by looking at the dissociation of carbon dioxide in water. The reactions of carbon dioxide dissolving in water and the subsequent dissociation reactions to the bicarbonate ion (HCO_3^-) and then to the carbonate ion (CO_3^{2-}) were described by Snoeyink and Jenkins (1981) as seen in Formula 1 below.

For instance, at a pH of 6.4 the concentration of H_2CO_3 (equal to CO_2) and HCO_3^- are the same, but at a pH of 5.5 only 10 percent of the carbonate exists in the HCO_3^- form (see Figure 3 for carbonate dissociation).

There are two methods to control calcium carbonate scaling: acid addition prior to the membranes and the use of a scale inhibitor. By definition, the RO manufacturers require the concentrate LSI to be less than zero if scale inhibitor is not used, so that calcium carbonate does not precipitate. For example, Hydranautics RO system design guidelines recommend a concentrate LSI of < -0.2. Adding scale inhibitor allows the con-

Formula 1

$$CO_2 + H_2O \Leftrightarrow H_2CO_3$$
$$H_2CO_3 \Leftrightarrow H^+ + HCO_3^- \qquad pK_1 = 6.4 \ (25°C)$$
$$HCO_3^- \Leftrightarrow H^+ + CO_3^{2-} \qquad pK_2 = 10.3 \ (25°C)$$
$$LSI = pH - pH_s$$

pHs is equilibrium pH value in the raw water (7.7 at 25°C)
(Water is scaling when LSI>0 and aggressive when LSI<0)

Parameter	Units	2008 (sampled)	2033 (projected)
Barium	mg/L	0.030	0.059
Fluoride	mg/L	1.40	1.89
Sodium	mg/L	343	730
Total Dissolved Solids	mg/L	1544	2398
Chloride	mg/L	589	1200
Sulfate	mg/L	269	346
pH		7.70	7.81
Bicarbonate Alkalinity	mg/L as CaCO$_3$	141	178
Calcium	mg/L	90	140
Hydrogen Sulfide	mg/L	3.0	3.0
Magnesium	mg/L	85.6	108.7
Potassium	mg/L	19.0	29.6
Silica	mg/L	14.6	23.0
Strontium	mg/L	17.4	33.7

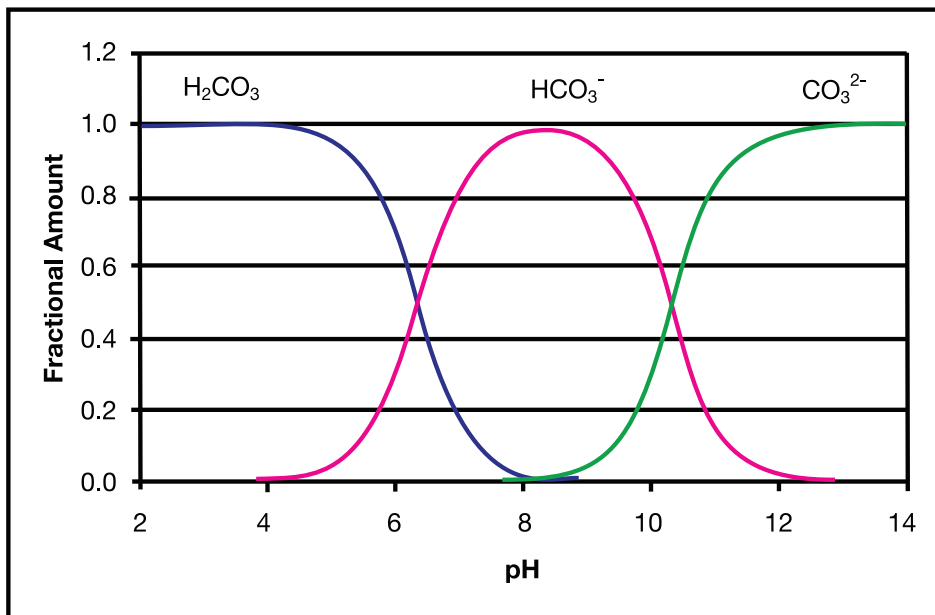Table 1: Typical Raw Water Quality for North RO Water Treatment Plant Design Life

Figure 3: Carbonate Dissociation in Water as a Function of pH (MWH, 2005)

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centrate LSI to be higher than zero (see discussion in following paragraphs).

Adding acid to the RO feed water will lower the concentrate LSI, since the alkalinity in the form of bicarbonate shifts to carbon dioxide, which will not combine with calcium and scale the membrane. Dissolved gases such as carbon dioxide pass through RO membranes and this potential alkalinity is retained in the permeate.

Table 2 illustrates the range of the concentrate LSI calculated by three RO manufacturers using their proprietary RO design software. Each manufacturer calculates the LSI slightly differently, which reflects differences in the performance of their membranes. The table illustrates that to obtain a concentrate LSI of approximately zero without scale inhibitor addition, an influent pH of less than 6.0 must be achieved. A pH of 6.0 requires a 98 percent strength sulfuric acid dose of approximately 90-120 mg/L over the 25-year life of the plant.

According to the RO manufacturers and scale inhibitor manufacturers, however, with the addition of an approved scale inhibitor, the concentrate may have an LSI up to approximately 3 without increasing the scaling potential of the membranes. Each scale inhibitor manufacturer uses proprietary scaling projection programs to determine the highest range of concentrate LSI at which they can guarantee no calcium carbonate scaling. For example, American Water Chemicals proprietary software determined that their product A102 has a concentrate limit LSI of 2.6, and their software modeling results recommend acid addition after the year 2018. The recommendations of the scale inhibitor manufacturers are discussed more in the next section.

Protection of the membranes and allowing for higher recoveries dictates adopting a concentrate LSI standard lower than the scale inhibitor manufacturers. MWH recommends adopting a concentrate LSI of no higher than 1.8, which would require the addition of some acid to the pretreatment stream to obtain a pH of 7.0 or lower in combination with scale inhibitor.

Sparingly Soluble Salt Control

Scaling of the membranes by the other sparingly soluble salts in the concentrate is also a concern. When compounds such as barium sulfate exceed their solubility products, scaling may occur on the membrane.

RO concentrate quality was projected for the years 2008 and 2033 using proprietary RO manufacturer software and by the scale inhibitor manufacturers using their proprietary software. This quality was projected for 80 percent water recovery with no acid addition. The concentrate levels are compared against the solubility product (Ksp) and a maximum concentration that is allowed by the RO manufacturers, with and without scale inhibitor addition. Using scale inhibitor keeps the salts from precipitating.

Three leading scale inhibitor manufacturers were consulted to provide recommendations of their maximum Ksp factors and determine the recommendations based on the present and projected raw water qualities. The results of the analyses indicate that 2-5 mg/L of scale inhibitor is typically required over the life of the plant.

Table 3 summarizes the results of the manufacturers' analyses. All the manufacturers recommended operating the RO membranes without acid addition until after the year 2018.

Hydrogen Sulfide Fouling Control

Another constituent of concern for Cape Coral is the hydrogen sulfide (H_2S) in the feed water and the potential for fouling. H_2S is common in Florida aquifers and is present in the Lower Hawthorn Aquifer. Utilities that utilize this aquifer must provide for hydrogen sulfide removal through treatment plant processes.

The chemistry of hydrogen sulfide can be illustrated in two ways, first as the dissociation of the sulfide gas to form the bisulfide ion (HS^-) and then the sulfide ion (S^{2-}). The

Pretreatment pH	Sulfuric Acid 98% Required (mg/L)		Concentrate LSI	
	Year 2008	Year 2033	Year 2008	Year 2033
6.0	93	116	0.0-0.15	0.3-0.5
6.5	52	64	0.75-1.0	1.0-1.25
7.0	19	24	1.4-1.6	1.75-1.95
7.7 (Ambient)	0	0	2.1-2.4	2.4-2.8

Table 2: Projected Concentrate LSI at Various Pretreatment pH Conditions

| Scale inhibitor Manufacturer	Product Recommendation	Dose Range (mg/L)
American Water Chemical (AWC)	A102	3-5
King Lee	Pretreat 100	3-4
Avista	Vitec	2-4

Table 3: Recommended Doses of Scale inhibitor

Figure 4: Sulfide Dissociation in Water as a Function of pH (Rees, et al, 2002)

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equations from Snoeyink and Jenkins (1981) are as follows:

$$H_2S \Leftrightarrow H+ + HS^- \qquad pK_1 = 7.0 \ (25\ °C)$$
$$HS^- \Leftrightarrow H+ + S^{2-} \qquad pK_2 = 12.0 \ (25\ °C)$$
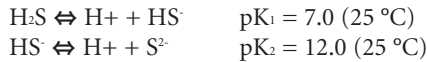
As the pH of the solution decreases below 7, more of the sulfide is present as H_2S gas. Assuming an ambient pH of 7.7, less than 30 percent of the sulfide is present as H_2S gas. The remaining fraction is present as dissolved bisulfide. At pH of 6.0, 90 percent of the ion is present as H_2S. According to the chemistry, at a pH of 5.1, nearly 100 percent of the ion is present as H_2S gas (see also Figure 4 for the sulfide dissociation).

Reverse osmosis membranes allow the passage and equilibration of gases through the surface. There is no rejection of gases such as H_2S or CO_2; therefore, at lower pH, more of the gas is present in the feed and passes through the membrane to the permeate side.

At the higher pH where ions are present in the dissolved form, these dissolved ions such as HS^- are rejected by the membrane. The rejection is dependent on the membrane.

Koch Membrane Systems engineers report that typically 95 percent of the ion is rejected similar to HCO_3^-; therefore, the sulfide will be more predominant in either the concentrate streams or the permeate streams, depending on the pH through the membranes. For instance, at a high pH, more dissolved sulfide will be rejected and be present in the concentrate.
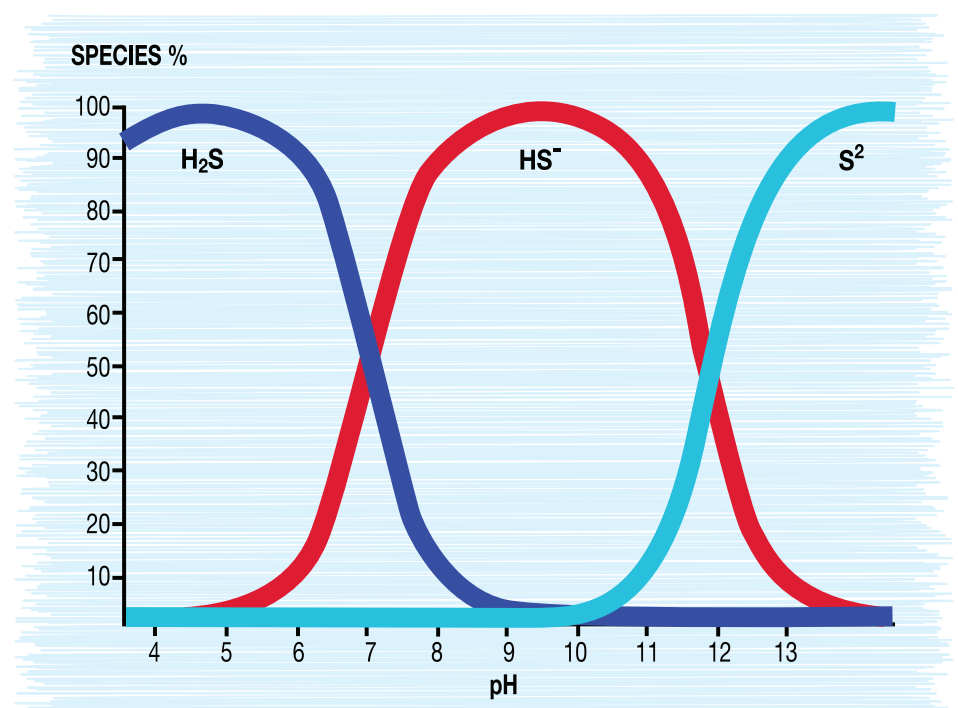
Fouling from H_2S on the RO membrane will occur primarily by elemental sulfur, metal sulfides, or oxides of sulfur. These reportedly are difficult to remove from the membranes. The chemical reaction for formation of elemental sulfur is from the American Water Works Association (1990):

$$2H_2S + O_2 = 2H_2O + 2S_{(s)}$$
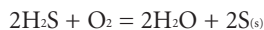
Good design and operation practice for RO membrane plants is to minimize, or avoid, introducing air (oxygen) into the system to prevent the oxidation of H_2S or oxidation of other potential foulants, such as ferrous compounds.

Impacts of Pretreatment Chemicals on the Downstream Treatment Processes

While the previous section describes the proposed pretreatment operations of the plant to control all forms of scaling and fouling on RO membranes, impacts to processes downstream of the RO membranes must be considered prior to analyzing the overall optimization of pretreatment operations.

Pretreatment recommendations on pH directly affect the removal of H_2S, a nuisance gas which must be removed prior to distribution by a degasifying process that uses aeration (stripping) to remove the H_2S from the blended product. The blended product is defined as the RO permeate plus the portion of the raw water which was bypassed around the RO and then mixed together.

Bypassing is used commonly in Florida RO plants for approximately 10 to 25 percent of the feed water. This blend has a better "stability" for corrosiveness than RO permeate on its own, and since the RO permeate is less than 100 mg/L TDS and the secondary standard is 500 mg/L, some of the feed can be blended with the RO permeate while meeting all treated water standards.

The removal of H_2S gas should be as complete as possible to maintain low odor and chlorine consumption in the treated product. Thus, the pH of blended permeate should be adjusted to 5.8 to ensure that the largest fraction (greater than 90 percent) of sulfide is present in gas form for nearly complete removal through the degasifier.

In addition, a portion of the carbonate (lower than 70 percent) is present in gas form (see Figures 3 and 4), which will also be stripped in the degasifiers. Lowering the pH below 5.8 would provide only marginal additional sulfide removal, while the additional (potential) alkalinity removal could be substantial, requiring reminerilization downstream.

Now examining the North RO Water Treatment Plant with the RO feed water pH summarized in Table 1, ranging from 6.0 to 7.7 (ambient) results in the pH of the blended product ranging from 5.7 to 7.1. Additional pH adjustment may be needed to adjust the blended product to a pH of 5.8 prior to degassing, depending on the pretreatment target pH. That is, entering the RO at an ambient pH of 7.7 results in a blended water pH (bypass plus RO permeate) of 7.1, requiring pH adjustment down to 5.8 just prior to the degasifier (acid feeds).

A substantial portion of alkalinity is first removed over the RO system and the remaining alkalinity is converted to CO_2 and second stripped in the degasifiers. This requires adjustments to condition the finished water prior to distribution. Post-treatment stabilization is commonly accomplished using lime or a combination of CO_2 and caustic soda.

The difference of alkalinity and its components, namely CO_2 versus the soluble species, is different, depending on when the pH is adjusted during the process. The effects are demonstrated in optimizing the overall treatment process in the following sections.

Full Plant Chemistry Analysis & Operating Cost Results

Based on the previous analyses, the North Plant can be operated safely at any pretreatment pH as long as the recommended scale inhibitor dose is applied. The informa-

quality changes from chemical addition.
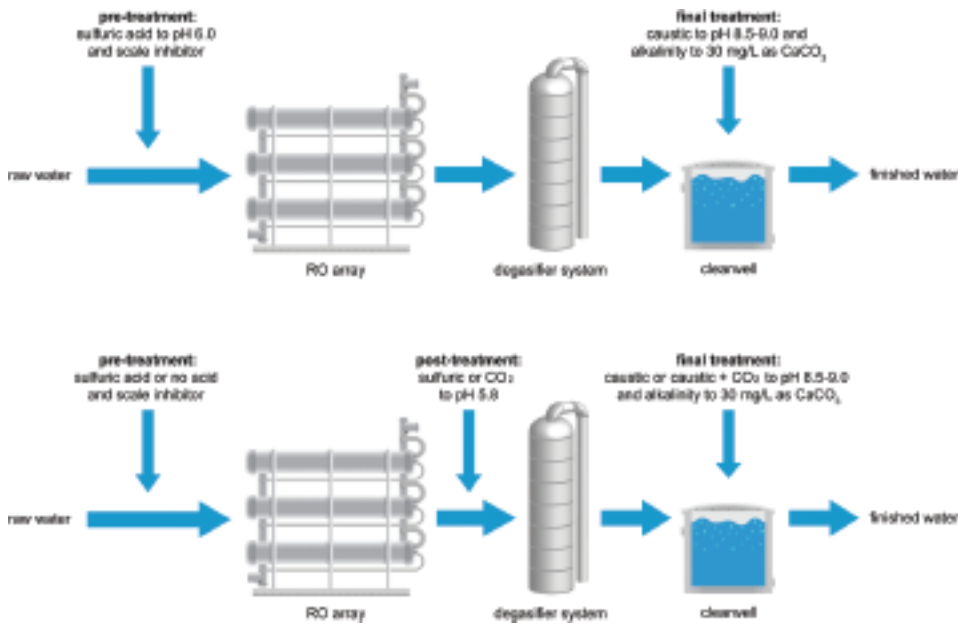
2. MWH Blending Model: A spreadsheet-based model for predicting the resulting alkalinity, pH and carbon dioxide of blending two waters, in any percent mix.

3. MWH CO_2 Stripping Model: A spreadsheet-based model for predicting the pH and carbon dioxide after passage through a stripping tower.

4. RO Manufacturer Proprietary Models: An input-based model to determine the RO permeate quality. The RO feed pH was entered into this model.

Detailed analysis using these models under the conditions for 2008 and 2033 for the chemical addition points, illustrated in Figure 5, resulted in the water quality and chemical requirements summarized in Tables 4 and 5. Table 4 illustrates using only sulfuric acid through the plant, and then stabilizing with caustic and CO_2. Table 5 illustrates using sulfuric at the RO feed, but then CO_2 prior to degasification, then caustic and CO_2 for stabilization.

The operating costs were calculated using the following assumptions for all these analyses:

◆ Sulfuric Acid, 98 percent, $57.50/liquid ton (source: City of Cape Coral)

◆ Carbon Dioxide, 100 percent gaseous, $0.07/lb (source: Fort Myers Water Treatment Plant)

◆ Caustic Soda, 50 percent, $446.58/liquid

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tion obtained by analyzing the pretreatment requirements of the RO membranes determined that a pH of ambient or 7.0 would be optimum for this plant, but the pretreatment pH also has an effect on the final pH, the final alkalinity, and the quantities of chemicals required. This section of the analysis presents the full plant chemistry and the operating costs of each scenario.

A cost analysis was conducted on several chemical addition alternatives common to the Southwest Florida area and the chemicals required. Figure 5 illustrates the options considered for chemical addition and stabilization of the North Plant feed and finished water.

The water quality was determined for the following locations: blended water after RO, after pre-degasifier acid addition, after degasifier, after final chemical addition for stabilization. Several models were used to calculate these water qualities, effects of air stripping, and chemical requirements at each point in the process. Here are the models and the use of each:

1. RTW Version 4.0: The Rothberg, Tamburini, and Windsor Model is a spreadsheet based tool available through the American Water Works Association to predict the chemical dosages required for a given water quality or resulting water

RO Feed Water			Blended Water		Post-Treatment (pH ¡Â 58)			Degasifier Discharge		Finished Water					Chemical Costs				
H₂SO₄ (mg/L)	WQ pH	Alk	WQ pH	Alk	H₂SO₄ (mg/L)	WQ pH	Alk	WQ pH	Alk	NaOH (mg/L)	NaOCl (mg/L)	CO₂ (mg/L)	WQ pH	Alk	H₂SO₄ $ / yr	CO₂ $ / yr	NaOH $ / yr	NaOCl $ / yr	Total $ / yr
Year 2008																			
93	6.0	50	5.8	25	0	5.8	25	6.7	25	13	4	4.3	9.0	42	105,000	11,000	216,000	77,000	**409,000**
19	7.0	123	6.4	28	16	5.8	12	6.8	12	23	4	19	9.0	41	39,000	48,000	374,000	77,000	**538,000**
0	7.7	142	7.1	29	20	5.8	8	6.8	8	24	4	21	9.0	38	21,000	54,000	390,000	77,000	**542,000**
Year 2033																			
116	6.0	64	5.8	32	0	5.8	32	6.8	32	10	4	0	8.9	45	262,000	0	329,000	154,000	**745,000**
24	7.0	156	6.4	37	21	5.8	16	6.8	16	14	4	8.3	9.0	33	98,000	42,000	446,000	154,000	**740,000**
0	7.7	179	7.1	39	27	5.8	12	6.8	12	18	4	13.6	9.0	34	57,000	69,000	572,000	154,000	**852,000**

Table 4 (Above) Sulfuric Acid Addition; Table 5 (Below) CO₂ Addition

Pre-Treatment (RO Feed Water)			Blended Water		Post-Treatment			Degasifier Discharge		Finished Water					Chemical Costs				
H₂SO₄ (mg/L)	WQ pH	Alk	WQ pH	Alk	CO₂ (mg/L)	WQ pH	Alk	WQ pH	Alk	NaOH (mg/L)	NaOCl (mg/L)	CO₂ (mg/L)	WQ pH	Alk	H₂SO₄ $ / yr	CO₂ $ / yr	NaOH $ / yr	NaOCl $ / yr	Total $ / yr
Year 2008																			
93	6.0	50	5.8	25	0	5.8	25	6.7	25	13	4	4.3	9.0	42	105,000	11,000	216,000	77,000	**409,000**
19	7.0	123	6.4	28	55	5.8	28	6.8	28	9	4	0	9.0	40	22,000	140,000	150,000	77,000	**389,000**
0	7.7	142	7.1	29	69	5.8	29	6.8	29	11	4	0	9.0	43	0	176,000	179,000	77,000	**432,000**
Year 2033																			
116	6.0	64	5.8	32	0	5.77	32	6.8	32	10	4	0	8.9	45	262,000	0	329,000	154,000	**745,000**
24	7.0	156	6.4	37	70	5.8	37	6.8	37	11	4	0	8.8	50	54,000	357,000	348,000	154,000	**913,000**
0	7.7	179	7.1	39	89	5.8	39	6.8	39	11	4	0	8.8	53	0	454,000	361,000	154,000	**969,000**

RO Plant	Influent TDS (mg/L)	Influent pH	Blended Water Alkalinity (mg/L as $CaCO_3$)	Degasifier Influent pH	Finished Water Method of Stabilization	Finished Water Alkalinity (mg/L as $CaCO_3$)
Cape Coral SW	2000	6.0	26-30 [1]	6.1–6.2	Caustic soda to pH 9.0	28-32
North Lee County	2200	6.0	27-30 [1]	6.1–6.2	Caustic soda to pH 9.0	28-32
North Collier County	3300	5.9	90 [2]	6.2	Caustic soda to pH of 8.5	45-55
Bonita Springs Utility	3200	6.8	N/A	6.1	No caustic dose applied [3]	75-80
Fort Myers	2500	7.4	30 [1]	5.9 (sulfuric addition)	Caustic soda to pH 10.5 and CO_2 to reduce pH to 8.5	28-30
Island Water	2500	7.6	30	No degas [4]	Caustic to pH of 7.7	28

[1] The alkalinity in the blend originates from the RO bypass (generally between 10 and 15%).
[1] The alkalinity in the blend originates from NF permeate, which is blended with RO permeate. The blend ratio is preferably 1:2 (RO:NF).
[1] RO permeate is blended with finished water from the lime softening plant in the clearwell. The blend ratio is preferably 2:1 (RO:Lime). The blend is dosed with 4.5ppm sodium hypochlorite.
[1] IWA doses chlorine gas to transform sulfide into sulfate.

ton (source: City of Cape Coral)
- Blend percentage – 15.6 percent
- Carbon dioxide stripping in degasifier = 100 percent
- All mixing and chemical reactions to completion
- Finished water pH = 8.5 to 9.0
- Finished water alkalinity > 30 mg/L as $CaCO_3$

The two tables illustrate the following major points:

1. Reduction of pre-treatment pH to 6.0 requires no additional acid adjustment prior to the degasifiers. Higher pH values do require additional acid, whether it is sulfuric or CO_2.
2. Using a higher pretreatment pH leads to more alkalinity removal by RO because less CO_2 passes through the membrane and requires some remineralization with carbon dioxide and caustic soda downstream of the degasifiers to stabilize the water.
3. Using a higher pretreatment pH results in overall lower sulfuric acid doses, which results in lower sulfuric acid consumption per year; however, as Point 2 states, more carbon dioxide and caustic soda are required downstream and the costs of these chemicals are greater than sulfuric acid, resulting in an overall higher plant operating cost.
4. Using CO_2 in place of sulfuric acid prior to the degasifiers may be considered initially but becomes less attractive with increasing raw water salinity, again due to the cost differential of the two chemicals (Table 4 versus Table 5).

Area References

Site visits to RO plants at the city of Fort Myers, North Lee County, Collier County, Bonita Springs, and Sanibel Island indicated that the pretreatment strategies employed vary widely. The following describes the area experience and water quality of these plants (see Table 6).

Three of the six utilities correct the RO feed water pH to around 6.0. Two of the six utilities apply only a limited sulfuric acid dose and operate the RO under close to ambient pH conditions. One utility operates the RO at a midway pH value.

Also, two utilities blend with finished water from other treatment plants to stabilize the RO permeate (North Collier County and Bonita Springs), while the other three utilities apply a RO bypass to stabilize the RO permeate and to reduce the operating cost.

Summary & Recommendations

The pH and alkalinity profiles through the plant were examined using water chemistry and unit process analyses. The study found that acid consumption could be reduced but required a simultaneous increase in chemicals for post-treatment.

The results reported are highly dependent on the alkalinity of the source water and the finished water alkalinity goals. For example, when the RO permeate can be blended with another stable finished water (from a lime softening plant), costly stabilization through the addition of carbon dioxide and caustic soda is not required and operation of the RO membranes under more ambient pH conditions may be cost advantageous. Also, if the feed water quality has much higher alkalinity values, the removal through the plant may not be as great as at the levels found in the Lower Hawthorne Aquifer, and may result in a different operating cost model (Christopher, et al, 2001).

The cost savings for a new 12-mgd plant at low feed water alkalinity could be as high as $150,000 per year in operating costs for the optimum solution. Overall, it was recommended that the existing pH pretreatment regime at the Southwest RO Water Treatment Plant should be utilized for the new North Plant.

Sulfuric acid addition for post-treatment was added to the plant to provide flexibility and to ensure full hydrogen sulfide removal. For long-term flexibility, other proven, alternate scale inhibitors should be considered. This evaluation may be revised when prices of the suggested chemicals change significantly.

The significance of this evaluation to the industry indicates that a utility considering shifting to use only scale inhibitors because of safety concerns over sulfuric acid may realize some cost increases in post-treatment chemical consumption. In general this model should be applied, since looking only at the RO process will not provide a full picture of the water treatment plant.

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