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Reverse osmosis process chemistry relevant to the Gulf

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Abstract

The cost of seawater desalination by reverse osmosis technology (RO) continues to drop with time. This portends accelerating adoption of RO around the world in the 21st century. Since chemicals are used in the operation of RO systems as coagulants, antifoulants and cleaners, continuing and increasing the adoption of the RO process in the Gulf necessitates conservation and efficient utilization of these chemicals. This is possible through understanding and optimization of the RO process chemistry. In this paper we survey the chemical factors that affect the operational characteristics of RO systems. The factors include the chemistry of the feedwater, membrane fouling mechanisms, composition of foulants found in membrane elements from many plants around the world and examples of antifoulants and their application. Current approaches towards the development of antifoulants to prevent fouling and membrane cleaners for plant maintenance are discussed in this context. The chemistry presented is part of a conceptual framework in which process optimization and chemical usage conservation can progress well into the next century.

Keywords: Reverse osmosis; Desalination; Chemistry; Process; Membrane foulants; Antifoulants; Cleaners; Conceptual framework; Optimization

1. Introduction

In the Middle East, particularly around the Gulf, over 50% of the world's desalination plants are found. A large segment of the desalination installations use reverse osmosis (RO) membrane technology. Recent bid prices of seawater desalinated with RO were offered for sale for as low as US\$0.60/M³ in Florida (USA), which amounts to less than half of any reported costs

for a plant currently in operation [1]. This portends accelerating the adoption of RO around the world in the 21st century. Since chemicals are used in the operation of RO systems as coagulants, antifoulants and cleaners, continuing and increasing the adoption of the RO process in the Gulf necessitates conservation and efficient utilization of these chemicals. This is possible through understanding and optimization of the RO process chemistry.

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2. Feedwater chemistry

The oceans are the most abundant sources of water, amounting to 99.37% of water on earth [2]. The Gulf water can be differentiated from typical seawater due to its relatively low volume, high evaporation rate, restricted circulation and mixing with fresh seawater and high utilization rate. These factors are known to result in significantly higher salinity (TDS 55,775 mg/l compared to 35,159 mg/l of standard seawater) [3]. Sand (50 microns–2 mm), silt (5–10 microns), clay (<5 microns) and submicron colloidal particles are introduced by natural and industrial drainage and erosion. Just as active is the process of growth, excretion, death and decay of animals and microorganisms. The biotic life in the Gulf waters can be expected to be different from other localized areas of the seas and should be studied and considered in the control of RO processes. The amount of different components in seawater is not proportional to the quantity of the components that enter the sea, but is inversely proportional to the facility with which the components in the seawater are made insoluble by chemical actions in the sea [4]. When river water meets the highly saline seawater, much of the suspended colloidal matter coagulates and is

deposited as organic-matter-rich silt in the delta regions. This is the same mechanism as the deposition of coagulated colloidal matter in the RO system with the increase in salinity of the brine. Complexation interactions and precipitation of particles are active natural processes in seawater. Despite the high salinity, biotic life abounds. Some of the types of colloids that exist in the natural waters, especially in the sea, are listed in Table 1. Polyphenolic complexes such as humic acid, lignin and tannin are decay products of woody tissues of plants that are noticeable due to their tendency to foul RO membranes by themselves or coagulate with other foulants [5]. Polysaccharides that constitute cell walls of microorganisms and plants and excreted variously as gums, slimes and biofilms are prominent players in membrane fouling. In the ocean, they are added to polysaccharides from sea organisms, especially the plankton [6], which upon aging become kerogen. It is believed that a white material called “marine snow” that has been observed to fall in deep oceans are these aged and ubiquitous polysaccharides [7].

Table 1
Colloidal matter in natural waters

Microorganisms
Biological debris (plant and animal)
Polysaccharides (gums, slime, plankton, fibrils)
Lipoproteins (secretions)
Clay (hydrated aluminum and iron silicates)
Silt
Oils
Kerogen (aged polysaccharides, marine snow)
Humic Acid/Lignins/Tannins
Iron and manganese oxides
Calcium carbonate
Sulfur and sulfides

3. Membrane fouling mechanisms

Feedwaters to RO systems typically are concentrated by a factor of 2–4 (50–75% recovery) during desalination. For simplicity in classification, three classes of fouling [8–14] can be observed.

First, the solubility limits of various dissolved salts in the brine stream may be exceeded, leading to deposition and growth of crystals in the flow channels and membrane surface of the RO elements. This type of fouling is referred to as “scaling”. Most common examples of scales are calcium carbonate, sulfates of calcium, strontium and barium, and calcium fluoride. In the microscopic examination of foulants [5], crystals appear to be well defined. Inhibitors injected into feedwaters to suppress crystallization are called antiscalants.

Secondly, particulate matter pre-existing in feedwater can agglomerate and adhere to the membrane and brine flow channels due to increased concentration, salinity, compaction, flocculation, surface interactions and other physical and chemical factors. The particles may be large enough to be removed with 1 to 5 micron RO prefilters such as silt, carbon fines, coagula and microbial clusters. There may also be colloidal particles that escape filtration even with 0.45 micron filters used in Silt Density Index measurements. The colloids can be organic, inorganic or composites. Ferric, aluminum and manganese hydroxides and silica grow through increasing particle sizes by polymerization and cross-linking with organic and inorganic polymers to become gels and amorphous foulants commonly seen on membranes [5,15]. Biotic debris such as polysaccharides and dead cellular matter contribute greatly to this type of foulants. We have found that anticoagulants and anti-deposition agents show promise in inhibiting this fouling process. “Antifoulants” then can be a term applied specifically in this particulate fouling context. In a broader sense, the term

antifoulant is used in the field to include antiscalants as well [9].

Thirdly, a prominent source of fouling, as is true of all water treatment or distribution systems, is the growth and anchoring of microorganisms. Moderate temperatures and minimal nutrient levels in RO waters can at times support explosive growths of microorganisms. Bacteria with cell division times of 20 min can grow from a normal count per unit volume of water to a too numerous count in the period of an 8-h shift. Due to the tendency of bacteria to secrete polymers that anchor themselves to surfaces to facilitate growth as the biofilm, this fouling mechanism is unique and poses a serious threat to the operation of RO systems [16]. This threat is compounded by the great difficulty of completely removing biofilm from membrane surfaces.

4. Composition of foulants

Through analysis of the composition of foulants on seawater and brackish water RO membrane elements sent from plants across the US and some overseas locations, we have seen examples of simple scales and more frequently the more difficult to clean composites of foulants and biomass [5]. Inorganic scales are relatively easy to identify by qualitative tests and quantitation by ion emission spectroscopy using inductively coupled plasma (ICP) or by scanning electron microscopy with energy diffusive X-ray analysis (SEM–EDX) methods. Our experience confirms the limited set of calcium carbonate, sulfate of calcium, strontium and barium, and calcium fluoride as the common scales found in RO systems. Known responses to specific cleaning formulations also allow us to confirm the identity of the scale.

Elemental gravimetric analyses of carbon, hydrogen, and nitrogen along with quantitation of residual ash after combustion allowed us to examine the organic components of foulants (in

the limited sense of coagulated particulate matter) and biomass. After correlating the results of these analytical data on foulants with feedwater chemistry, plant design and operations data, we have made the following observations [5]. These observations have been helpful in guiding process improvements to minimize fouling and improve cleaning strategies.

1. Organic matter contributes more than 10% of the mass of foulants in all recent samples, with the majority of the samples containing 50–100% of organic matter.

2. Biomass constituted the major part of the organic contents of the foulants.

3. Synthetic polymers used in water pretreatment can often be differentiated from biomass in the foulants.

4. Anionic polymers used as antiscalants and dispersants can be deposited on membranes and act to concentrate high valence metal ions (Fe, Al, Ca) on the membrane surface.

5. Colloidal silica and clays have complex and variable contents of iron, aluminum and calcium.

6. Elemental phosphorous is associated with biomass, phosphate and phosphonates.

These six aspects of fouling can each occur alone or frequently or with two or more aspects simultaneously. Discernment and differentiation of the contributing fouling mechanisms are a developing science, which is vital to the development of antifouling chemicals and effective cleaning methods.

5. Antifoulant design and application

The term “antifoulant” used here is in its broadest meaning covering scaling, particulate fouling and microbial fouling — the three classes of fouling mechanisms discussed above. We will briefly discuss strategies aimed at each of these three areas.

For scale control, the development and application of antiscalants is well known and reviewed in the field of boiling water and cooling water chemistry [17–19] and applied to boilers, evaporators, cooling towers and cooling systems. Anionic polymers, polyphosphates and organophosphorous compounds, sometimes referred to as threshold inhibitors and dispersants, are used in substoichiometric amounts, usually in the range of 1–5 mg/l concentrations. By binding to surfaces of growing crystal nuclei, the rates of crystallization from supersaturated solutions are retarded, and crystal packing orders are modified. By this mechanism, crystallization rates are so retarded that although supersaturation of solutes in the water will eventually equilibrate through crystallization, within the residence time of the water in the system, there is little or no scale formation. The uniqueness of RO among water conditioning systems is that the residence time is very short (a few seconds), concentration of seed crystals is low, and temperature is constant. For this reason, higher levels of supersaturation without crystallization are possible. On the other hand, the limits of saturation and rates of scaling are hard to model, measure and predict. Interference comes from other solutes in the water, organic or inorganic [19]. Assumptions of RO fouling limits vary considerably among practitioners.

For controlling fouling by preexisting particulate matter, the task is much more challenging due to the variety of types of potential foulants and the complexity of their interactions [4,5,11,13,15], with each other in the same water and with the membrane. Stability and agglomeration of colloidal particles are subjects of major importance in natural waters [4] as well as in the treatment of process waters [2]. Drawing on the basic science of colloids and testing of model foulants suggested by our RO foulant analysis data [5], progress is steadily being made with the development of anticoagulants and

anti-deposition agents. Case studies from various plants will be presented in the near future.

Concerning prevention and management of biofouling in water treatment systems, the literature is extensive. Much of the art and science found generally useful is applicable to RO systems as well. Several factors peculiar to the RO system can be mentioned. Chemicals used to sanitize and clean the system have to be chemically compatible with the thin, salt-rejecting, barrier membrane. Of prime concern is that accumulation and exponential growth of the microorganisms should not be allowed to occur within the system. Pretreatment of feedwater, adequate maintenance of upstream unit operations, continuous flow of water through the RO unit, good monitoring and sanitization program and use of preservatives during downtime [20] are important to this end. Normalized flow and differential pressure in the system are sensitive indicators of biofouling. To prevent irreversible fouling, trend charting and readiness to perform adequate cleaning are important aspects of the system operation. Immediate assistance should be sought where cleaning has been inadequate. Powerful new cleaners are now available providing alternatives to membrane replacement in the worst cases of biofouling.

6. Practical implications for Gulf RO systems

Given the above observations and in our opinion, an enlarged conceptual framework from what is commonly considered, we can look at some practical implications for RO systems in the Gulf region. Since scaling is not generally a problem for seawater and very high TDS well waters, we would like to address here, firstly, antifoulants for controlling the coagulation of colloidal and polymeric organic matter. Petroleum sources, active biotic life in warm waters, low exchange rate in the Gulf with fresh

seawater through tides, and high industrial utilization present unique conditions of high levels of organic matter prone to coagulation in RO systems, bringing down with them as foulants, inorganic colloidal particles. Secondly, as we have found in our extensive analysis of composition of foulants and correlations with responses to cleaning, the presence of biomass and natural and synthetic organic polymers on membranes make cleanings challenging. We will briefly discuss improved cleaning with cleaning enhancers.

Exciting developments have occurred in the expansion of a line of antifoulants [21] that not only prevent coagulation in RO systems, but also act as dispersants and antideposition agents for partially coagulated matter. Small RO systems typically used in making bottled drinking water from poor quality surface waters with biomass and particulate contents are helped by injection of antifoulants without other pretreatment. For larger RO systems where pretreatment can be designed and controlled, the economy and convenience of treating colloidal particulate matter with antifoulants alone can be an attractive alternative to clarification and Silt Density Index reduction with coagulant addition and demanding filtration schemes. Case studies will be published elsewhere.

Difficult cleaning problems typically call for resolution by expert companies. Through thorough review of the water and pretreatment chemistry, analysis of the foulant composition and source, and selection of proprietary antifoulants and high performance cleaners, both fouling avoidance and reliable maintenance cleaning can be attained. Most RO systems are cleaned with generic chemicals recommended by membrane manufacturers such as shown in Table 2. While these generic chemicals often produce satisfactory results, many systems require better cleaners to achieve full recoveries from fouling. Some key ingredients from

Table 2

BetterClean™-boosted cleaning solutions formulated with generic chemicals in concentrations recommended by major membrane manufacturers

BetterClean™-	A	B	C	D	S
TYPES OF FOULANTS-	Acid Soluble Foulants	Biofilm, Bacterial Biological Matter	Oils and Organic Matter	Organic and Inorganic Colloids	Silica and Silicates
Fluid Systems	1% citric Acid	/ <----- 1% STPP (TFC only) -----> /			
			1% EDTA.Na ⁺ 1% TSP 1% Borax (TFC only) 0.1% Triton X-100 (CA only)		
Hydranautics	2% Citric Acid 0.1% TritonX-100 or Tergitol 8 etc	2% STPP 0.26% SDDBS pH 7.5	0.5% Na Perborate 0.1% TritonX-100 or Tergitol 8 etc.	2% STPP 0.8% EDTA.Na ⁺ 0.1% TritonX-100 or Tergitol 8 etc	-
Filmtec	0.2% HCl or 0.5% H ₃ PO ₄ or 2.0% Citric Acid or 0.2% Sulfamic Acid	/ <----- 1% EDTA.Na -----> / 0.1% NaOH to pH=12 or following pH=12 solutions: 0.05% Na-DDS or 0.1% STPP + 1% EDTA.Na, or 0.1% TSP + 1% EDTA.Na		0.1% NaOH 0.05% Na-DDS pH=12	-
Desal	HCl pH = 3 or Citric Acid pH = 2	/ <----- 1% TSP -----> / 1% STPP 0.1% Na-DDS 1% EDTA.Na pH = 8 - 11.5			
Permasep	0.5% HCl (pH 2.3 min) or 0.5% H ₃ PO ₄ (pH 2.3 min) or 0.2% Sulfamic Acid or 2% Citric Acid, pH=4 or 2% Citric Acid 2% Na ₂ EDTA NH ₄ OH to pH = 4 or 1% Na ₂ S ₂ O ₄ or 1% NaHMP	1% Na ₂ EDTA 0.1% NaOH pH = 11 max	0.3% NaPerborate 0.25% Na-DDBS pH = 10 or 1% Na ₂ EDTA 1% STPP 1% TSP, pH=11 or 2% STPP 0.25% Na-DDBS pH = 10 or 1% NaHMP	0.3% NaPerborate 0.25% Na-DDBS pH = 10 or 2% STPP 0.25% Na-DDBS pH = 10 or 1% NaHMP	0.5% NaOH, pH=11 or 1% Na ₂ EDTA 0.1% NaOH, pH=11 or 0.3% NaPerborate 0.25% Na-DDBS pH = 10 or 2% STPP 0.25% Na-DDBS pH = 10
KEY:	STPP = Sodium Tripolyphosphate EDTA.Na = Sodium Salt of Ethylenediaminetetraacetic Acid TSP = Trisodium Phosphate Triton X-100 and Tergitol 8 = Non-ionic Surfactants Na-DDS = Sodium Dodecylsulfate Na-DDBS = Sodium Dodecylbenzenesulfonic Acid Na ₂ EDTA = Disodium Salt of Ethylenediaminetetraacetic Acid NaHMP = Sodium Hexametaphosphate				

proprietary products that are missing in generic cleaner formulations (Table 2) are made into concentrated cleaning enhancers [21] to be used in v/v dilution ratios of up to 1:250 in conjunction with the generic formulations (see Table 2). A case study on the use of the cleaner enhancers (BetterClean™-A and D) in an off-line pilot cleaning of single elements from an industrial waste water reclamation RO is given in Appendix 1.

7. Conclusions

This paper provides the conceptual framework in which contributions to the RO process chemistry are being made. The review of the chemistry of seawater and the natural processes that lead to deposition of solids is instructive for the control of the same or similar processes within the RO system. Known membrane fouling mechanisms are reviewed. Analysis of the composition of foulants sampled from membranes from RO plants provided insight on the specific contributors to fouling. Such knowledge provides the basis for designing antifouling strategies and testing methodology for developing antifoulants. Reference is given to new antifoulants developed to control coagulation of organic foulants in the RO system and to new cleaner enhancers to improve the cleaning effectiveness of generic cleaning chemicals. We look forward to the expansion of this conceptual framework and reporting on advances towards the day when RO plants can be uniformly sturdy in operation and predictable in performance. Optimal utilization of RO chemicals should be a continuing target into the next century, particularly important to the conservation of Gulf water quality and the economical production of water.

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Appendix 1

Case study on cleaning enhancement of reverse osmosis membranes with BetterClean™

To improve the effectiveness of routine maintenance cleaning of a reverse osmosis system used to recover water from an industrial waste water stream, the use of BetterClean products are investigated. A batch of thin-film composite membrane elements (TriSep ACMII 8.3"×40") were test-cleaned off site. The routine cleaning conditions using phosphoric acid at pH 3 followed by sodium hydroxide at pH 11.0–11.5 were found helpful in the plant, but can be improved upon. This study resulted in a recommendation to use BetterClean A and BetterClean D in conjunction with the low pH and high pH generic cleaners, respectively, to attain two- to five-fold improvement in permeate flow.

Elements were cleaned individually by recirculation of 20 gal of cleaning solution at low pressure at 30–40 gal/min (GPM) cross flowrate

and no permeation for 30 min. The temperature throughout was approximately 77°F (25°C). Tests were performed at 112.5 psi (half of 225 psi manufacturer specification condition). Permeate flow rates were recorded as GPM before each cleaning, and after 10 or more minutes of rinsing with San Diego city water, which is filtered through an activated carbon bed to remove residual chlorine. To normalize to specification conditions of 225 psi, each increase of 0.10 GPM in the observed permeate flow is equivalent to approximately 288 GPD under specification conditions.

The increase in GPM of permeate flow after each cleaning step is recorded and tabulated below (Table A1). The magnitudes of the increase with and without BetterClean A and D are compared.

The use of BetterClean A with phosphoric acid and BetterClean D with sodium hydroxide at pH 13 would increase the overall effectiveness of the two step cleaning by three- to five-fold.

Table A1

Increase in permeate flow (GPM) after each cleaning step

Element no./ cleaners	H ₃ PO ₄ , pH 3	H ₃ PO ₄ , pH 3 + BC-A	NaOH, pH 11–11.5	NaOH, pH 13 + BC-D	Total improvement in permeate flow ^a (in GPM)
1. 80127	0.015	—	0.105	—	0.120
2. 80077	0.027	—	0.094 ^b	—	0.121
3. 80198	0.039	—	0.070	—	0.109
4. 80152	—	0.036	—	0.317	0.353
5. 80178	—	0.023	—	0.334	0.357
6. 80175	—	0.090	—	0.418	0.508
7. 79298	—	0.025	—	0.549	0.574

^aSalt rejections have improved or are unaffected.

^bCleaning the element with pH 13 NaOH after this step only increased flow by 0.080 GPM.

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