INFLUENCE OF NANOFILTRATION PRETREATMENT ON SCALE DEPOSITION IN MULTI-STAGE FLASH THERMAL DESALINATION PLANTS

by

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Scale formation represents a major operational problem encountered in thermal desalination plants. In current installed plants, and to allow for a reasonable safety margin, sulfate scale deposition limits the top brine temperature in multi-stage flash distillers up to 110-112 °C. This has significant effect on the unit capital, operational, and water production cost. In this work, the influence of nanofiltration pretreatment on the scale deposition potential and increasing top brine temperature in multi-stage flash thermal desalination plants is modeled on the basis of mass transfer with chemical reaction of solutes in the brine. Full and partial nanofiltration pretreatment of the feed water were investigated. The top brine temperature can be increased in multi-stage flash by increasing the percentage of nanofiltration treated feed. Full nanofiltration pretreatment of the make-up allows top brine temperature in the multi-stage flash plant to be raised up to 175 °C in the case of di-hybrid nanofiltration/multi-stage flash and up to 165 °C in the case of tri-hybrid nanofiltration/reverse osmosis/multi-stage flash. The significant scale reduction is associated with increasing flashing range, unit recovery, unit performance, and will lead to reduction in heat transfer surface area, pumping power and therefore, water production cost.

Key words: *nanofiltration, pretreatment, scale deposition, sulfate removal, desalination*

Introduction

Scale formation represents a major operational problem encountered in thermal desalination plants. In today's operating plants, and to allow for a reasonable safety margin, sulfate scale deposition limits the top brine temperature (TBT) in multi-stage flash (MSF) distillers up to 110-112 °C. Limited TBT and flashing range have significant effect on unit capital, operational, and water production cost. In addition, scale deposits have a direct influence on the thermal units performance and water cost. This appears in the effect of scale on fouling factor, overall heat transfer coefficient, specific heat transfer area and, therefore, the specific capital cost (CAPEX). On the other hand, the scale influences: (1) surface friction losses, pressure drop, and

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pumping power, and (2) the frequency of ball cleaning and the chemicals dosing rate and, therefore, the specific operation cost (OPEX). The nanofiltration (NF) is commonly considered as an intermediate membrane filtration process between ultrafiltration (UF) and reverse osmosis (RO). It offers, due to the loose structure of its membrane porosity, a higher fluxes compared to RO process. Compared to UF, NF membranes have a higher retention capability of organic matters, synthetic dyes, antibiotics, and all viruses. With molecular weight cut-offs (MWCO) from 200 to 2000 Da, NF is capable also of divalent ions rejection depending on their molecular weight. NF membranes can be classified into tight structure and loose structure membranes. Tight structure NF membranes are characterized by small pore size and hence high rejection and low permeate flow, while loose structure NF membrane has larger pores and hence low rejection and high flux. Although the use of NF process is relatively recent, it has used in a wide range of applications such as food, pharmaceutical, water desalination, and wastewater treatment industries [1-4].

In seawater desalination, NF has been proposed for divalent ions removal from seawater feed entering the thermal processes [5-7]. The Saline Water Desalination Research Institute (SWDRI) in Saudi Arabia has been actively involved in the development of NF pretreatment technology application for both seawater reverse osmosis (SWRO) and multistage MSF desalination processes. For MSF process, NF has the advantageous over the conventional up-to-date antiscalant dosing method in that operating with NF feed pretreatment can increase the TBT above the present operational limits of 110-112 °C. This is because: firstly, the efficiency of antiscalants decreased at high temperatures due to the thermal degradation and secondly, due to the formation of calcium sulfate salts at high temperature which can not be avoided by available antiscalants [8].

In case of using NF, sulfate ions are almost completely removed from seawater, and TBT can easily be increased above the present operational limits. NF pretreatment has a significant capability to lower the concentration of hard scale elements in seawater especially Ca^{2+}, Mg^{2+}, SO_4^{2-}, and HCO_3^-. Hassan [9] has reported the following NF salts rejection values: Ca^{2+} 80.7%, Mg^{2+} 87.7%, total hardness 86.5%, SO_4^{2-} 93.3%, and HCO_3^- as $CaCO_3$ 63.3%. In the same paper, the authors reported more than 26% rejection to monovalent ions such as Cl^-, Na^+, and K^+ by the NF membrane. The total seawater salinity was also decreased from 44,046 to 27,619 ppm after the NF treatment which is equivalent to 37% reduction in seawater total dissolved solids (TDS). The feed pH was also reduced from 8.2 to 7.85 after the NF pretreatment. As a result, it was able to increase the TBT of MSF to >130 °C. The recovery rate of MSF operating on NF permeate was found to reach 80% and 70% at TBT 130 °C and 120 °C, respectively. Three different types of NF membrane were tested for salt rejection and permeate flux. For seawater softening, a special NF membrane was used which was characterized by an average rejection rate and permeate flow [9]. In a similarly related work, Awerbuch [10] presented the benefit of using NF membranes in the removal of scale elements from seawater. Awerbuch suggested using NF permeate-seawater mixture feed (partial feed pretreatment) to thermal process to reduce the cost of NF pretreatment. The study showed the feasibility to increase TBT in MSF up to 125 °C with only 25% NF permeate mixture with seawater.

In this work, the influence of NF pretreatment on sulfate scale potential and the consequent TBT increase range were studied. A theoretical simulation program developed by Hydranautics Nitto Denko Company (IMS software), was used for calculating NF rejection rate and membrane permeate flow. The NF permeate was used as a feed and its characteristics were used as input to calculate scale formation potential in BR-MSF evaporators. A model based on mass transfer with chemical reaction of solutes in the brine was used.

Table 1. Characteristics of NF (ESNA1-LF2) membrane

Parameters	Values
Membrane type	ESNA1-LF2
Membrane polymer	Composite polyamide
Nominal membrane area [m^2]	37.2
Maximum applied pressure [bar]	41.6
Maximum chlorine concentration [ppm]	< 0.1
Maximum operating temperature [°C]	45
Feed water pH range	3-10
Maximum feed water turbidity	1 NTU
Maximum feed water SDI (15 minutes)	4
Maximum feed flow (m^3/h)	17
Membrane length (mm)	1016
Membrane diameter (mm)	201.2

Feed seawater NF pretreatment

The integrated membrane solutions (IMS) program, developed by Hydranautics NITTO DENKO Company, was used for simulating NF pretreatment to seawater. The specifications of the spiral wound NF ESNA1-LF2 membrane used for seawater pretreatment are shown in tab. 1. ESNA1-LF2 NF membrane is classified as a softening element for seawater treatment. It has a high rejection rate to scale elements combined with moderate permeate flow rate. The compositions of feed water considered in this study were similar to that used by Awerbuch [10] for typical Arabian Gulf seawater.

Table 2 shows the resultant composition of feed water before and after the NF pretreatment. The feed temperature, applied pressure and recovery rate were 30 °C, 32.8 bar, and 60%, respectively. According to IMS, the rejection rate values are: Ca^{2+} 70.5%, Mg^{2+} 82.4%, Na^+ 29%, SO_4^2 87.69%, HCO_3 62.54%, and Cl^- 33.62%. These results are in agreement with the experimental data obtained from the pilot plant in Umm Lujj, Saudi Arabia [9]. Umm Lujj

Table 2. Feed water compositions before and after NF pretreatment

Ions	NF feed [ppm]	NF permeate [ppm]	Rejection rate [%]		
			This work	Hassan [9]	Awerbuch [10]
Ca^{2+}	600	176.7	70.5	81	23
Mg^{2+}	1550	272.4	82.4	88	76
Na^+	14840	10523.0	29.0	26	10
K^+	500	483.0	3.3		14
Ba^{2+}	0.07	0.017	75.7		
Sr^{2+}	18	4.3	76.0		
SO_4^2	3440	423.2	87.7	93	98
HCO_3	128	48.0	62.5	63	86
CO_3^2	38	3.2	91.7		
Cl^-	26253	17425.5	33.6	26	4
TDS	47367	29360.0	38.0		
pH	8.2	7.8			

pilot test was operating on 64% recovery rate and feed temperature 33 °C [11]. The applied feed pressure was between 18-25 bar. This signified the proper selection of NF membrane in this study which yielded acceptable results compared to the data from the Umm Lujj typical experimental work. There are slight differences in the operating parameters between the theoretical and experimental data in which the latter used slightly higher recovery rate and feed temperature. However, both membranes showed a high rejection rate to Ca^{2+}, Mg^{2+}, and SO_4^{2} which is normally expected in NF process. The reduction in feed TDS was 38% and 37% for theoretical and experimental data, respectively. According to IMS design program the NF module ESNA1-LF2 is able to reduce the concentration of scale elements in feed solution to the same level achieved in Umm Lujj pilot plant.

Scale potential in MSF distillers

MSF configuration

In the MSF process, two different configurations can be differentiated (fig. 1, from Al-Shayji [12]). In once-through distillers, the concentrated brine from the last stage is discharged to the sea. In recycle distillers, a portion of the concentrated brine from the last stage is mixed with the feed water. When the brine enters the first flash chamber in MSF distillers, due to the sudden reduction of the CO_2 partial pressure, CO_2 is released into the vapour space. Consequently, scales ($CaCO_3$, $Mg(OH)_2$, and $CaSO_4$) precipitate in the bottom of the flash chamber

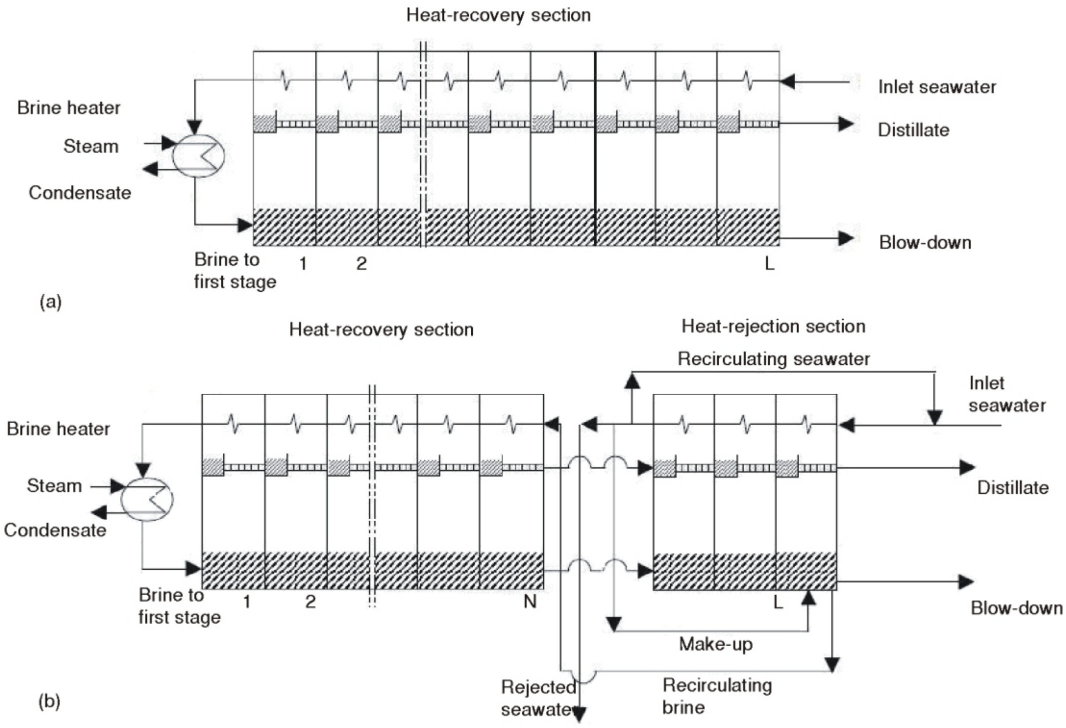

Figure 1. MSF process: (a) once-through distiller and (b) recycle distillers [12]

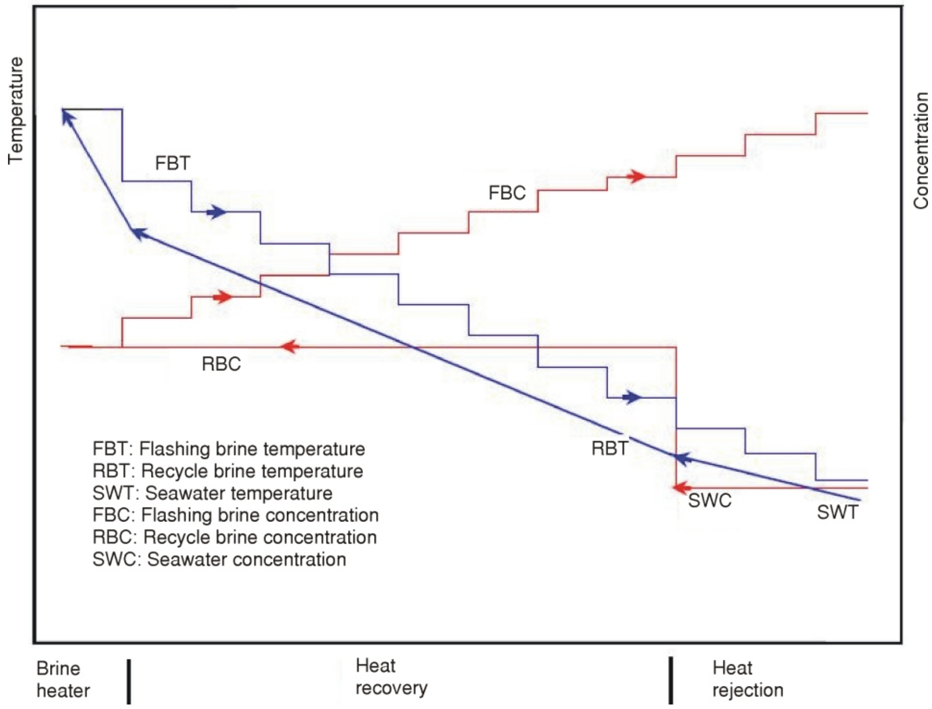

Figure 2. Temperature and concentration gradients in MSF distillers

and inside the tubes. Figure 2 [13] shows the trend of temperature and concentration gradients in various sections of a typical MSF distiller as the one shown in fig. 1(b).

$CaSO_4$ scale potential

Scaling in MSF evaporator tubes is a complex process depending on a variety of factors such as seawater composition, tube flow velocity, heat flux, and surface and bulk temperatures. The composition is thought to have a significant effect on scaling. Sulfate scales result from the direct crystallization of anhydrite ($CaSO_4$), hemi-hydrate ($CaSO_4 \cdot ½H_2O$) or gypsum ($CaSO_4 \cdot 2H_2O$) from seawater, once their solubility limits are exceeded. Most of the deposited calcium sulfate found in seawater desalination plants, is in the form of hemi-hydrate. The sulfate minerals are insoluble in common chemicals and their development inside a distiller should be avoided by all means. This is achieved by operating the plant at temperatures and/or brine concentrations not allowing saturation and precipitation conditions [14, 15]. Calcium sulfate is two orders of magnitude more soluble than calcium carbonate. This means that the sulfate is much less likely to drop out of solution when both are present. At higher temperatures, calcium sulfate becomes more influencing, because calcium carbonate scale, if not inhibited, begins precipitate at lower temperature and can be easily cleaned, chemically by acids or mechanically, by sponge ball cleaning.The solubility of calcium sulfate is strongly affected by the presence and concentration of other ions in the system.

Skillman developed a simple sulfate solubility index for estimating the likelihood of calcium sulfate scaling. Skillman index [16] is a ratio between the actual concentration, $[i]_{\text{actual}}$, of either calcium or sulfate and its theoretical or equilibrium concentration whichever is the limiting species:

$$\text{Skillman index} \quad \frac{[i]_{\text{actual}}}{(\sqrt{x^2 \quad 4K_{\text{sp}}} \quad x)\ 10^3} \tag{1}$$

where x is the absolute value of the excess common-ion concentration of calcium and sulfate ions:

$$x \quad \left|2.5[\text{C}^{2} \ ] \quad 1.04[\text{SO}_4^{2} \ ]\right| \ 10^{\ 5} \tag{2}$$

The solubility product constant (K_{sp}) can be determined from the equation of solubility (s), in g/L, established by Linke *et al.* [17] as:

$$s = 2.091 + 0.003173T - 8.193 \cdot 10^{-5}T^2 \tag{3}$$

The NF-pretreated seawater was used as a feed for MSF evaporators. The scale potential was calculated using a C++ code. Details on calculation procedures can be found in Al-Rawajfeh [18,19].

Figure 3 shows the $CaCO_3$ deposition (gram per ton distillate) in the individual stages flash chambers of typical MSF once-through and recycle distillers [18]. $CaCO_3$ deposition rates notably decreases from the first to the last stage and this can be attributed to the following reasons: (1) because of $CaCO_3$ precipitation in a certain stages, TA of solution entering the next stages becomes lower, and (2) CO_2 release rate decreases, because the difference between the concentration of CO_2 in the bulk and at the phase interface, desorption driving force, decreases. This can be attributed to the increase in salinity with evaporation which causes the solubility of CO_2 to drop.

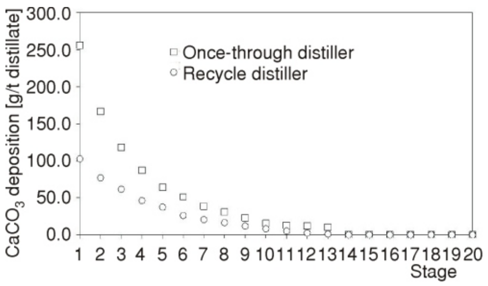

Figure 3. The $CaCO_3$ deposition rates in the individual stages flash chambers of the MSF once-through and recycle distillers

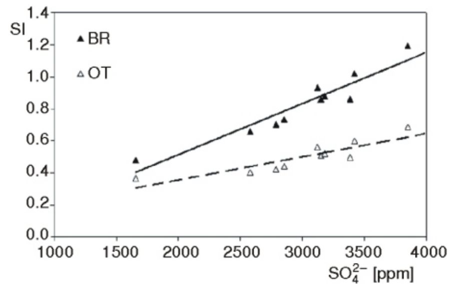

Figure 4. The effect of sulfate ion concentration in the feed for different feed water composition in different intakes from the Arabian Gulf

Figure 4 shows the effect of sulfate ion concentration in the feed for different feed water composition in different intakes from the Arabian Gulf for once-through (OT) and brine recycle (BR) MSF distillers. The $CaSO_4$ scale potential increases with increasing the sulfate ion content in the feed water. The typical heat transfer resistances in MSF are as follows: 34-39%, 34-36%, 20-25%, and 7-8% for fouling factor (FF), internal film coefficient (R_i), outer film coefficient (R_o), and tube wall (R_w), respectively. Fouling factor is a controlling parameter in the overall heat transfer coefficient.

According to Al-Sofi [13], certain abnormalities in sludge and scale presence in various parts of MSF distillers were reported in the past. Heavy or uneven depositions were reported to occur inside heat exchanger tubes from lower end to mid-section up to high temperature parts in heat recovery stages. There were also cases when inlet sides of brine heater tubes were fouled to a higher degree than outlet tube ends. Moreover, there are references to alkaline scale inverse temperature dependence solubility. It is worth stressing that the so-called inverse behavior resulting from the rate of generation of anions rather than the temperature dependence of alkaline scale solubility. In addition to the above, uneven sludge depositions were reported especially in water boxes and on the face of tube sheets. Such uneven presence were either restricted to certain areas of the water box and the tube sheet or very heavy in some specific stages along the flow path of recirculating brine from cold to hot end of the recovery section or across the brine heater. Uneven depositions of somewhat similar pattern were also reported to take place inside water boxes and on inlet tube sheets and those of heat recovery or rejection section inlets in particular. There were also cases when heavier depositions reported in lower temperature flash chambers and more specifically carry over of scale into their demister pads.

Results and discussions

Di-hybrid NF-MSF

The general idea to control all types of scale deposits is to maintain the concentrations of the scale forming ions below the solubility product of the precipitating salt. This may be achieved by pretreatment of the feed water. Recent studies [5-11] showed that NF is a promising approach for pretreatment of seawater offering a viable alternative to avoid the limitation in top brine temperature. Two different approach have been tested; di-hybrid NF-MSF and tri-hybrid NF-RO-MSF. A schematic flow diagram of di-hybrid NF-MSF desalination system is shown in fig. 5 [20]. The seawater intake is first pretreated by passing it through a dual media filter followed by a fine sand filter. The pretreated seawater is then sent to the NF membrane. The NF product is used as a make-up to the MSF plant. It is assumed that the process is achieved without acid treatment which is normally used to prevent scaling. Sulfate concentrations may be elevated by the addition of H_2SO_4.

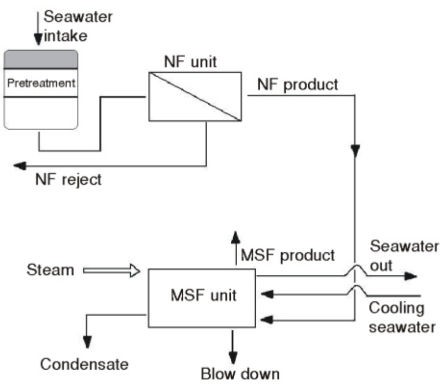

Figure 5. Schematic flow diagram of di-hybrid NF-MSF desalination system [20]

As a rule of thumb, it was suggested [21] that $CaSO_4$ scale would not form unless the product $(Ca^{2+})(SO_4^{2-})$ exceeds 50,000 ppm. Alternatively, to avoid sulfate scale formation, water hardness should be maintained below 900 ppm (as $CaCO_3$) or the pH should be approximately 6.50. Removing the scale forming ions from the raw seawater by NF opens the possibility to safely increase TBT above the current MSF operating temperature, which will materialize in significant reduction in unit water production cost. Increasing TBT has, on one hand, some advantages such as using less specific heat transfer surface area, leading to better design of

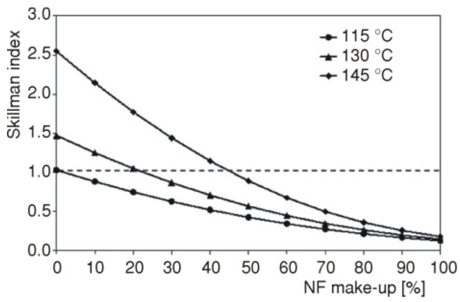

Figure 6. Influence of NF on sulfate scale potential in BR-MSF plant

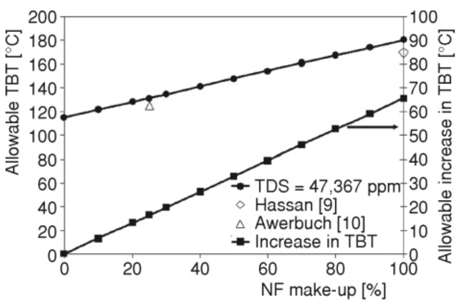

Figure 7. Shifting the maximum TBT by NF

dual-purpose power-desalination plants, lowering brine to distillate and cooling water to distillate ratios, consequently, lowering the pumping energy, and applying less vacuum duty especially for the high temperature MSF stages. On the other hand, high TBT plants need higher quality steam, higher pressure design for the evaporators and pumps, and higher quality maerials to face the problems associated with corrosion and thermal expansion [22].

Figure 6 shows the influence of NF on the sulfate scale potential, expressed by Skillman index, for seawater at 115, 130, and 145 °C in BR-MSF reference plant. The scale potential increases with increasing temperature and decrease with increasing the percentage of NF-treated feed. For seawater with no feed pretreatment (0% NF), the scale can start deposit at 115 °C. However, the maximum TBT, at which sulfate scale begins to precipitate, is shifted to higher temperature with increasing the NF-treated portion. The temperature is shifted to 120, 135, and 145 °C when the NF-treated portion increased from 10, 25, and 50%, respectively, as shown in fig. 7. For 100% NF feed pretreatment, TBT can reach as much as 175 °C.

The results presented here were calculated without considering the influence of antiscalant. It should be noticed that with 100% NF no need to use antiscalants while with 30% NF pretreatment, a TBT of 135 °C can be reached without antiscalants. The results obtained from fig. 2, are in a good agreement with the results of Awerbuch's [10], in which he showed the feasibility to increase TBT in MSF up to 125 °C by retreating only 25% of the seawater feed [9]. Full NF pretreatment of the feed reduce sulfate scale potential by ~95% (fig. 8) and allows TBT to rise up to 175 °C which is closed to the results reported by Hassan [9].

It should be noted that the scale formation is very much influenced by the brine temperature within the fluid boundary layer near the wall. The inside wall temperature and the boundary layer temperature is itself depends on: (1) the heating steam terminal temperatures 2-3 °C in the evaporator and 5-10 °C in brine heater, and (2) the tube wall material and thickness and the outside condensation coefficient of heat transfer (which again depends on the condensation film thickness and the presence of non condensable gases. For this reason, a safety margin should be taken to avoid scale deposits on the walls. *i. e.*, the real temperature to be avoided should be TBT + ΔT (boundary layer safety). Typical 8-10 °C is taken in operating plants so that a scale limit of 120 °C will only allow TBT to be 110-112 °C.

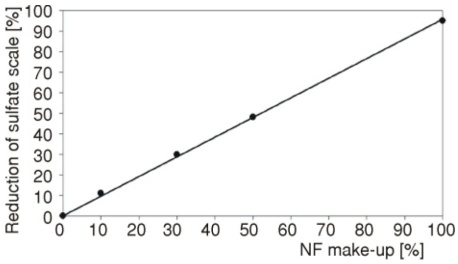

Figure 8. Influence of NF on sulfate scale in MSF-BR plant

Tri-hybrid NF-RO-MSF

Evaluation tests which were carried out by SWDRI [20] using the NF process for feed seawater pretreatment to reverse osmosis units, revealed that the RO rejects consist of a relatively low concentration of the scale forming components. Hence, it is worthwhile to investigate the possibility to send the RO reject to a BR-MSF plant and to check the maximum TBT at these conditions. A schematic flow diagram of tri-hybrid NF-RO-MSF desalination system is shown in fig. 9. The seawater intake is first pretreated by passing it through a dual media filter followed by a fine sand filter. The pretreated seawater is then sent to the NF membrane. The NF product is sent to the RO unit and its BR is used as a make-up to the MSF plant.

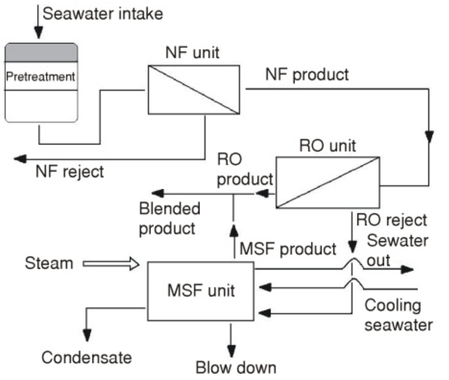

Figure 9. Schematic flow diagram of tri-hybrid NF-RO-MSF desalination system [20]

Figure 10 shows the sulfate scale potential for seawater with 0, 20, 40, 50, 60, 80, 100% NF-treated make-up at 60% RO recovery in a NF-RO-MSF hybrid plant. The scale potential increases with increasing temperature and decrease with increasing the percentage of NF-treated feed. Referring to fig. 11, the TBT is shifted from 115 °C to 123, 132, 138, 142, 152, and 165 °C when the NF-treated portion increased from 0 to 20, 40, 50, 60, 80, and 100%, respectively.

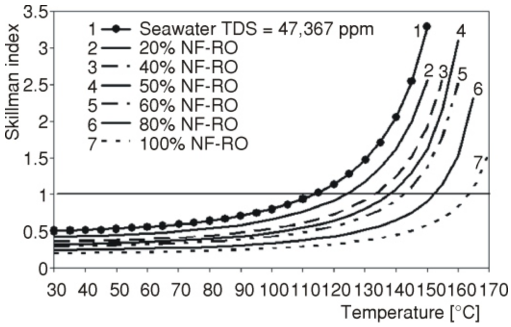

Figure 10. Scale potential in tri-hybrid NF-RO-MSF plant at RO recovery of 60%

The removal of scale forming components from seawater by NF increases the reliability of RO units [23]. According to Hamed *et. al*. [11], full integration of NF, RO, and MSF processes would result in enhancing reliability, flexibility, plant productivity and ultimately reduce water production cost. The very low concentration of the sulfate and calcium ions in the brine recycle which were below the saturation limits enabled to operate the MSF unit safely up to a top brine temperature of 130 °C and water recovery ratio of about 69%. The NF-RO-MSF hybridization: (1) reduces scaling potential throughout the entire processes, (2) provids the opportunity to blend the high purity distillate of the MSF plant with the RO permeates, (3), the flexibility inherited in the hybrid RO/MSF configuration provides the prospect for the RO unit to accommodate the power load variation without the need of an auxiliary boiler, when the power demand in a conventional dual purpose MSF/power plant is changing, it is essential to provide an auxiliary boiler to provide supplementary fuel for energy to keep water production constant, (4) increasing the TBT of the MSF distiller increases the distiller's flash range and subsequently will enhance the water production and recovery ratio, and (5) also feeding the RO process with a NF product resulted in the increase of the permeate recovery ratio. Blending the products of the MSF and RO process allows operating the RO unit with relatively high recovery ratio.

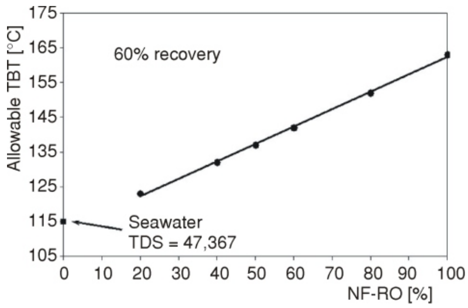

Figure 11. Influence of NF on the allowable TBT in tri-hybrid NF-RO-MSF plant

This hybridization would lead to cost reduction in at least four different components: (1) reducing equipment such as MSF brine recycle pump which could become redundant when RO reject is used to replace typically circulated brine stream with the MSF process, (2) improving recoveries would reduce input power for pumping special by the input to RO high pressure pump(s), (3) membrane feed heating especially in winter season and the operation of permeation processes (NF and RO) at narrower temperature ranges thus higher recovery and prolonged membrane life, and (4) markable reduction in treatment chemicals.

Conclusions

The sulfate scale potentials is analyzed and modeled in hybrid membrane-MSF evaporators. The results show that sulfate scale potential decreases with increasing the percentage of NF pretreatment. Consequently, the TBT is shifted to higher temperature with increasing the NF-treated portion. In NF-MSF hybrid plant case, full NF pretreatment of the feed reduce sulfate scale potential by ~95% and allows TBT to rise up to 175 °C. In NF-RO-MSF hybrid plant case, full NF pretreatment of the feed increase the TBT up to 165 °C. The results of the study may be help to: (1) determine the theoretical maximum TBT which can be reached utilizing NF pretreatment and the possibility to apply such a process to the existing plants, and (2) to carry out a detailed techno-economic study for the development and construction of MSF plants operating at higher TBT.

Nomenclature

[*i*] – ion concentration, [molkg^{-1}solution or meqL^{-1}]
K_{sp} – solubility product constant, [on the basis of $\text{mol}^2\text{kg}^{-2}$]
R – resistance to heat transfer
s – solubility, [gL^{-1}]
T – temperature, [K]
x – the absolute value of the excess concentration, [mgL^{-1}]

Supscript

i – internal film coefficient of heat transfer
o – outer film coefficient of heat transfer
w – tube wall thermal resistance

Acronyms

BR – brine-recycling
IMS – integrated membrane solution
MSF – multi-stage flash
NF – nanofiltration
NTU – nephelometric turbidity units
OT – once-through
RO – reverse osmosis
SDI – silt density index
TBT – top brine temperature
TDS – total dissolved solids, [ppm]

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