Potential of Scale Formation during the Desalination of Power Plant Wastewater in a Bench Scale Evaporator

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Abstract—Concerned with its impact on the environment, Eskom has implemented policies that work in the interest of preserving water. The 'zero liquid effluent policy' highlights the importance of water recycle and reuse within the electricity generation sector. Eskom utilizes reverse osmosis (RO) to treat power plant wastewater, but this process yields a retentate with high concentrations of scale forming compounds. Multi-effect distillation can serve as a practicable means to treat RO reject water. The scale potential on the heat exchanger surface within an MED system is high, compelling the pre-treatment of the reject solution. In this study, RO reject water collected from a power plant was analyzed to determine the physico-chemical characteristics, including the anion and cation content. The French Creek and the PHREEQC models were then used to predict the formation of scale, as well as the proposal of suitable antiscalants to delay the scale formation. Gypsum and barite saturation levels were high in the feed brine. PMA and HEDP antiscalants were proposed for the RO reject sample. The saturation indices of scale forming agents were seen to increase for an increase in temperature from 18.5°C to 75°C, as well as during the evaporation of the sample by 34.5%. According to the prediction, HEDP and PMA would be able to control scale formation for pH variations between 7 and 9, but will not able to control barite scale for any pH value. Additional treatment is then required to remove barite scale that would form in the MED system.

Keywords—Antiscalant, MED (Multi-effect distillation), RO reject, Scale.

I. INTRODUCTION

Water scarcity has been the topic of discussion in many fields for decades. South Africa is reported to be one of the driest countries in the world, experiencing almost half of the global average of 860 millimeters of rain per year (amounting to about 500 millimeters per year) [1-29]. Large scale usage of groundwater resources is highly restricted (by almost 80%) due to the country's geology [30]. Roughly 8% of available water in the country is used by the power generation sector (and other large industries). This percentage may seem low, but these industries are situated in water management areas (WMAs) that are strictly water constrained [31]. Knowledge of the energy-water nexus and the problem of water scarcity has led to the analysis of water usage trends within most industries, including the electricity generation sector [32]. In the same breath, Eskom's Acting General Manager made it clear that strategies to reduce the company's water footprint will be implemented, which include the diversification of Eskom's water and energy mix [33]. Over the past decade, waste water recovery and recycle have become very popular to minimize water availability issues. The reclamation of waste water contributes to achieve both volume minimization of liquid waste, and reducing the withdrawal of water (which severely affects ecosystems). By achieving these goals, it is possible for waste water to be viewed as a resource to achieve water sustainability in the near future. A water management strategy that aims to reuse and recycle waste water to minimize the need for disposal comes in the form of the zero liquid discharge (ZLD) strategy. The risks that the disposal of liquid waste poses to the environment are emphasized by this strategy [34]. The costs associated with waste water disposal due to regulations seem to outweigh the costs incurred by implementing ZLD [35]. ZLD processes that are thermal-based are energy-intensive, thus reverse osmosis (RO) has been introduced as a precursor to lower this energy requirement [34]. To prevent rupture, RO membranes are only operated at pressures up to between 55 and 80 bars. To ensure that the osmotic pressure is below this limit, the salinity of the retentate should not exceed 70 000 ppm [36] – [37]. It then follows that there are two main challenges that Eskom needs to tackle. Firstly, Eskom needs to comply with ZLD stipulations stated in their policies, which stipulate that

salts be disposed of onsite and that the disposal of liquid waste into the environment is forbidden. Secondly, a drop in the target for raw water consumption from 1.39 to 1.34 liters per kilowatt hour, has elevated the significance of waste water reclamation [38]. The main factors that contribute to the dangers of waste water for the environment include the salinity level, temperature and residual additives. The residual additives that are found in brines usually come in the form of antiscalants, which have a low degradability, hence making them dangerous to the surrounding environment. RO reject that is discarded can cause variations in pH of water resources, the spread of heavy metals and even eutrophication [39]-[40]. Evaporative thermal desalination processes have been successful in treating RO concentrate, and hybrid systems combining membrane and thermal processes have achieved water recoveries of up to 99% - given that the feed brines were pretreated to reduce scaling. Multi-effect distillation (MED) systems, specifically, are thermal desalination technologies, based on fluid mechanics and the mechanisms of heat transfer. Multiple cells make up a typical MED system, wherein the pressure, as well as temperature, decreases from effect – to – effect. The heat source for the next step is the water vapor that rose from the previous step [41]. Horizontal tube falling film MED systems often experience scaling problems, with scale being deposited on the outside of the heat exchanger tubes. As a result of scale formation, the thermal resistance of the tubes is increased, decreasing the performance of the system. Oversizing the heat transfer surface and adding chemical additives are some methods used in industry to minimize scaling, which unfortunately result in high capital and operating costs [42]. The factors to pay attention to that affect scaling in MED systems consist of the composition of the feed fluid, the thermal field, the flow field and the surface properties of the heat exchange surface. Investigating the limitations of the optimum process conditions, as well as adding chemical additives constitute an effective strategy in combating scaling. Furthermore, the pretreatment method is affected by the characteristics of the RO feed brine. The compounds of interest, in the brine characterization, are those that are most likely to form scale on the heat exchanger surface. These compounds mainly include calcite (all three polymorphs), anhydrite, gypsum and silica [43]. Any variations in the brine characteristics can result in changes in the composition and concentration level of resulting brine. Scale is typically crystalline in nature, exhibiting a surface charge that can be either directly or indirectly dependent on pH [44]. The pH has such a large effect on the likelihood of scaling, that a mere change in pH from 7(neutral) to 10 (basic) can lead to the premature precipitation of scale forming compounds. Temperature is another important factor when it comes to scale formation, with scale formation said to begin around 30-35°C and goes on to increase as temperature is increased. The abovementioned stresses the importance of the pretreatment method being able to handle potential changes in brine characteristics, without compromising the quality of product water [45]. The deciding factor between utilizing a membrane or thermal desalination lies in the total dissolved solids (TDS) content level. At high levels of TDS, about 40 000 ppm, in the RO reject, it has been found that thermal desalination technologies are crucial to minimize brine volume [41]. The saturation limits of each salt is to be kept below the upper limit to inhibit scaling in thermal desalination systems. The easiest way to achieve this is to introduce an acid or an antiscalant into the system. Antiscalants are usually made up of either phosphonate, acrylic acid or a blend of the two – in industry [38]. Supersaturation is the driving force for salt precipitation at the heat-and-mass transfer surface of an MED system. This does not mean that all solutions in the supersaturated state will experience precipitation, however. Metastability describes the state of a supersaturated solution that remains supersaturated for an extended period of time or even indefinitely, because nucleation does not occur in this state. The period of time during which the solution is metastable is called the induction period. The degree of supersaturation of the precipitating salt plays a role in how long the induction period of a solution is going to be. Instability of the solution begins at the onset of nucleation [46]. It then follows that, for thermal desalination processes, as the solution becomes more concentrated, the main limiting factor for precipitation is the degree of supersaturation that the solution is able to handle before becoming unstable. The phenomenon of scaling involves crystallization and transport mechanisms, and instead of eliminating the presence of scale forming compounds altogether, rather, antiscalants delay crystallization and retard crystal growth [47]. The main mechanisms of scale inhibition exhibited by antiscalants are known to be threshold action, chelation, dispersion and crystal distortion [44]. Weakening the adherence of scale to the flow surface and inhibiting further crystal growth is the best way to control scaling, which is also known as the threshold inhibition of scale [48]. The knowledge of how scale forms can assist in determining how antiscalants can function to prevent scale formation. The successful use of chemical inhibitors in industry can be attributed to their ease of handling, low dosages and low costs [49]. The optimum dosage of antiscalants is important in that if the dosing rate is too low, the onset of scale formation will occur, while a dosing rate that is too high results in sludge formation [50].

Thus, successful desalination involves the determination of the best type of inhibitor for the feed brine, as well as the optimum dosing rate. The geochemical simulator, PHREEQC, is employed to obtain the metal speciation of RO brine before and after thermal treatment, whereas the French Creek software proposes a suitable anti-scalant for the solution to achieve successful scale control in the MED system.

II. EXPERIMENTAL PROCEDURE

A. Water Sampling

Samples of RO water reject were collected from the Grootvlei Power Station, located in Mpumalanga province. The water samples were collected in 25 L clean, plastic water containers directly from the reverse osmosis brine sample

points. The sample points are situated roughly one meter above ground level with valves that can be opened or closed. Each water container was labelled after being filled. Samples were taken from both Train 1 and Train 2 of the RO system. The RO brine samples were covered in black plastic bags to prevent the penetration of external light, and refrigerated to prevent pre-mature precipitation [51-55]. The samples were only kept and used for a maximum of 4 weeks, and were discarded of thereafter.

B. Laboratory Analysis

The characterization of the samples involved measuring the conductivity, pH and ORP with the use of the Lovibond SensoDirect 150 multiparameter water quality meter. The Multiparameter Bench Photometer HI 83099 (Hanna Instruments Inc., USA) was used to determine the sulfate concentration in the samples, while the 2100Q portable turbidimeter was used to measure the turbidity level. Furthermore, the chloride content and total alkalinity were measured by the chloride portable photometer and the handheld colorimeter alkalinity for fresh water (Hanna), respectively. In order to determine which metals were present, as well as their relevant concentrations in the samples, four samples were externally measured using inductively coupled plasma optical emission spectrometry (ICP-OES) (Agilent Technologies, USA) at the Water Lab (Pty) Ltd. This company has received SANAS (South African National Accreditation System) accreditation. Two samples from each train (raw and diluted) were analyzed. Every instrument was calibrated utilizing buffer and standard solutions before being used.

A control water sample was made by dissolving salts to achieve concentrations of 0.005 mol/dm^3 CaCl$_2$.2H$_2$O and 0.005 mol/dm^3 CaCO$_3$ in distilled water [56].

C. Metal Speciation using PHREEQC Modelling

The speciation of metals in the RO reject samples was predicted using the geochemical modelling software PHREEQC. PHREEQC is open – source modelling package developed by the US Geological Survey. The software requires a chemical database and an input file (that can be modified by the user) to generate chemical equations [57,58]. This modelling software is based on an ion association aqueous model and has advanced capabilities with regards to speciation and saturation calculations. Given this, PHREEQC determines and reports the speciation of metals in a sample in terms of their inorganic complexes and free hydrated ions in order to identify the scaling potential of aqueous solutions [59]. The MINTEQ database, one amongst many offered by PHREEQC, was used in this study. A PHREEQC analysis was conducted on both the raw sample and the concentrated brine from evaporation to predict the effect of evaporation on the scaling potential of the RO brine.

D. Determination of Antiscalants using French Creek Software

The French Creek Software was used to predict the scaling potential, based on the solubility of scale forming species, as well as predict the optimum antiscaling agent for the water sample. The software makes use of multiple saturation and stability indices to accomplish this. The Langelier saturation and Ryzner stability indices are amongst the many that the French Creek software utilizes to give accurate results [57]. The optimum dosage of the proposed antiscalant was also obtained from the software simulation.

III. RESULTS AND DISCUSSION

A. Sample Characterization

The concentrations of the cations of interest in the feed RO brine, being calcium, barium and silicon were determined as 777.491 ppm, 0.76 ppm and 152.91 ppm, respectively. Whereas, the alkalinity as CaCO$_3$ was measured as 132 ppm, while the sulfate content was determined to be 3363 ppm. The pH of the feed brine was 8.18.

B. PHREEQC Modelling

Modelling in PHREEQC showed that the calcium ion is likely to form calcium sulfate and calcium carbonate at the feed conditions of 18.5°C and a pH of 8.18. More calcium sulfate is likely to form compared to calcium carbonate. Results also show that barium will not form complexes with free sulfate ions, and amorphous silica (SiO$_2$) will not form, at these conditions. Simulating evaporation via PHREEQC yielded an increase in the saturation indices of the common scale forming compounds, depicted in Table I. An evaporation of 34.5% was simulated (which is the evaporation achieved in our Bench Scale MED [56-57]). This makes it evident that the concentration of scale forming compounds drives the solution closer to supersaturation, and thus precipitation, increasing the potential of scale formation on the surface of heat exchanger. The pH after evaporation was measured as 9.39.

TABLE I: SATURATION INDICES OF COMMON SCALE FORMING AGENTS

Compound	Before evaporation	After evaporation
Anhydrite	0.12	0.33
Aragonite	0.68	0.86
Barite	2.21	2.41
Calcite	0.82	1.00
Gypsum	0.16	0.36
Amorphous Silica	0.3	0.48

TABLE II: SATURATION INDICES OF COMMON SCALE FORMING AGENTS

Compound	18.5°C	40°C	75°C
Anhydrite	0.12	0.26	0.44
Aragonite	0.68	0.7	0.77
Barite	2.21	1.85	1.33
Calcite	0.82	0.85	0.99
Gypsum	0.16	0.09	-0.01
Amorphous Silica	0.3	0.07	-0.25

The behavior of metal species in the sample was also simulated at 75°C. Table II depicts the increase in saturation indices for an increase in temperature. The saturation indices of calcite and anhydrite showed a direct relationship with temperature, increasing when the temperature of the sample was increased. However, the potential for barite scale to form was decreased when the temperature was increased. This behavior is also seen for gypsum and amorphous silica. This indicates that the combined effect of temperature increase and evaporation that the brine will experience in an MED system makes it highly likely for the formation of scale, if no preventative measures are put in place.

C. French Creek Software Analysis

The concentrations of metals obtained from the ICP-OES analysis were used as input for the French Creek software analysis of the feed RO brine. Table III depicts the saturation limits of the common scale forming compounds. Gypsum and barite are above their upper limits of 4.00 and 80.00, respectively. This means that these compounds are highly likely to precipitate out of solution. Additional treatment will be required to remove the barite, as it is highly insoluble and resistant to chemical treatment [60].

TABLE III: LIMITS OF COMMON SCALE FORMING COMPOUNDS

Compound	Saturation limit	Lower limit	Upper limit	Status
Calcite	65.41	N/A	150.00	OK
Gypsum	5.53	N/A	4.00	ABOVE LIMIT
Celestite	5.09	N/A	12.00	OK
Barite	396.72	N/A	80.00	ABOVE LIMIT
Amorphous Silica	0.00	N/A	1.20	OK
Fluorite	5.67	N/A	120	OK
Anhydrite	3.39	N/A	4.00	OK
Aragonite	60.50	N/A	150.00	OK
Langelier Index	2.28	N/A	2.50	BORDERLINE
Ryznar Index	3.44	N/A	2.80	ABOVE LIMIT

The proposed antiscalants were PMA (polyamaleic acid) and HEDP (1-Hydroxy Ethylidene-1,1-Diphosphonic Acid), which belong to the carboxylic and phosphonate groups, respectively. The recommended dosages were 0.062 and 0.428 ppm for HEDP and PMA, respectively. Phosphonates act in the nucleation and crystallization stages of crystal growth of many sparingly soluble inorganic salts, efficiently hindering these processes. They are also highly stable at elevated temperatures [61]. Additionally, PMA inhibitors do not experience loss of activity due to hydrolysis, and are more hydrolytically stable at high levels of pH. Unfortunately, these polymers have low calcium tolerance and can react to form calcium-polymer salts, which are not soluble [62]. Fig. 1 to Fig. 3 illustrates this, as PMA is able to control calcite, anhydrite and gypsum scale at pH values from 7 to 9. Whereas HEDP shows a similar trend for calcite, but is able to control anhydrite and gypsum scale at any pH level (see Fig. 4 to Fig. 5). Calcite is the most thermodynamically stable of the three polymorphs of calcium

carbonate, while vaterite is the least stable [63]. Both antiscalants were unable to prevent barite scale formation at any pH. The saturation level of barite did not change over a range of pH values from 7 to 13, regardless of whether antiscalants were added or not.
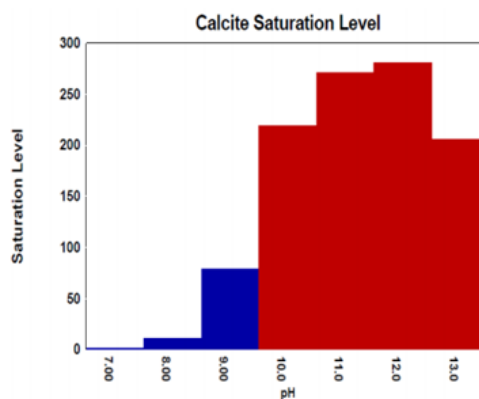

Fig. 1: Saturation level of Calcite vs pH for PMA (blue – safe range; red – scaling cannot be controlled, remedial action required)
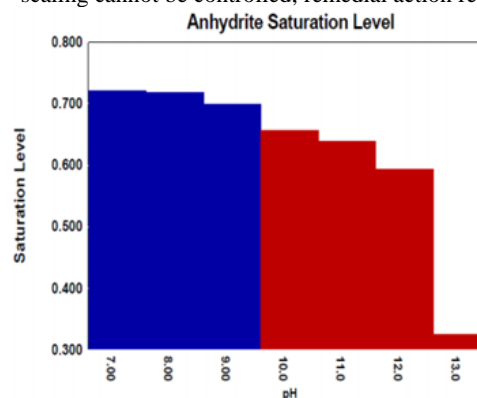

Fig. 2: Saturation level of Anhydrite vs pH for PMA (blue – safe range; red – scaling cannot be controlled, remedial action required)
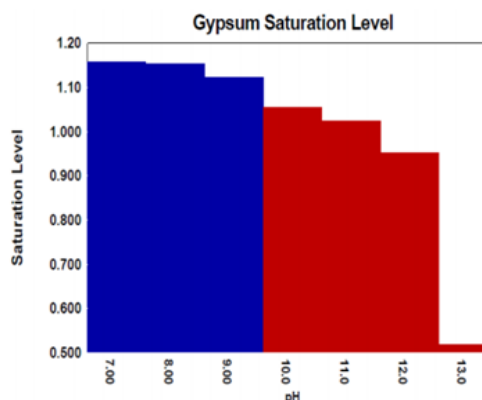

Fig. 3: Saturation level of Gypsum vs pH for PMA (blue – safe range; red – scaling cannot be controlled, remedial action required)
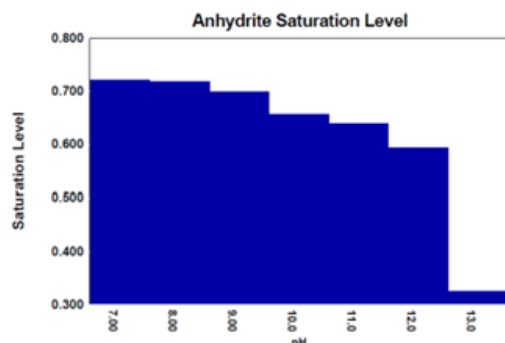
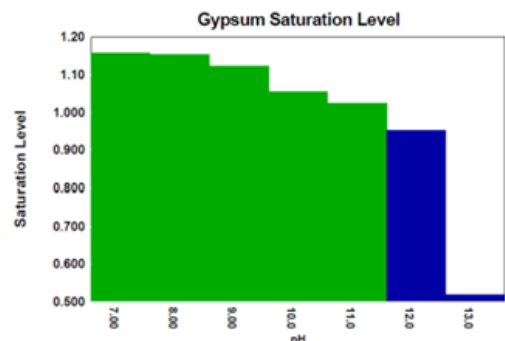Fig. 4: Saturation level of anhydrite vs pH for HEDP

Fig. 5: Saturation level of gypsum vs pH for HEDP (green – mild problem potential if conditions change slightly)

This gives an indication that the HEDP antiscalant is more flexible than the PMA antiscalant, for the RO brine sample.

IV. CONCLUSION

RO systems have limitations that necessitate the need for downstream desalination processes that will reduce the large volumes of concentrate produced. Thermal desalination has been successful in doing this, making use of the excess heat from low pressure spent steam in power plants. Scale formation poses a problem in thermal desalination systems, with the common scale forming agents being calcite, gypsum and anhydrite. The gypsum and barite saturation levels in the feed RO brine were above their limits, at 5.53 and 396.72, respectively. Evaporation of the RO brine illustrated an increase in the saturation indices of the common scale forming compounds. The same pattern is seen (for most of the scale forming compounds) for an increase in temperature to 75°C, which is the usual temperature for the first effect of an MED system. The proposed antiscalants, HEDP and PMA were both able to control scale formation for pH variations between 7 and 9, but were not able to control barite scale for any pH value. Additional treatment is required to remove the barite scale. HEDP performed better based on French Creek modelling, as it was able to fully inhibit both anhydrite and gypsum, unlike PMA. The pretreatment of RO brine is necessary because of the high propensity of the feed brine to form scale at conditions that are found in an MED system. To substantiate the viability of the use of antiscalants, the induction tests can be carried out to prove that the antiscalant extends the induction period of the brine, hopefully enough to improve the performance of MED systems.

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