Long-term performance decline in a brackish water reverse osmosis desalination plant. Predictive model for the water permeability coefficient

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Abstract

Transport models in reverse osmosis (RO) desalination have been extensively studied taking into account various factors such as temperature, fouling, etc. However, there are not many models that describe the behavior of a desalination plant over long time periods. These models depend on operating time and empirical parameters to estimate the flux or the average water permeability coefficient (A) decline. The proposed model separates the decline of A in two stages, the first stage refers to a more pronounced decline due to initial compaction and irreversible fouling and the second stage describes a more stable period with less slope. The model is based on the superposition of two exponential functions, which depends on operating time, empirical parameters and fouling potential of the feedwater (k_{fp}). Ten years operating data of a brackish water reverse osmosis (BWRO) desalination plant were used. The obtained results with the proposed model showed a slightly better fit than previous models, but giving meaning to two different behaviors separated in two stages.

Keywords: fouling potential, long term, reverse osmosis, water permeability, operating data.

1. Introduction

The reverse osmosis (RO) is currently the undisputed leading desalination method [1-3]. Despite the progress made in this field [4, 5], there is a continuing need for improvement and expansion. The understanding of the mechanisms at play in RO membrane separation is crucial to improve and raise this technology. Transport models are the tools used to understand membrane transport.

Different models have been proposed to give explanation to solvent and solute transport through dense or "nonporous" membranes [6]. Perhaps the more popular are the Spiegler-Kedem [7] and solution-diffusion [8] models, Eqs. (1) and (2) respectively.

$$J_{\rm w} = \frac{1}{R_{\rm m}\mu} (\Delta p - \sigma \Delta \pi) \tag{1}$$

$$J_{\rm w} = A(\Delta p - \Delta \pi) \tag{2}$$

Where J_w is the water flow (solvent), R_m is the membrane resistance, μ is the viscosity, Δp is the trans-membrane pressure, σ is the reflection coefficient (σ <1 indicates a semi-permeable solute, while σ =1 indicates an impermeable solute, complete rejection [9]), $\Delta \pi$ is the osmotic pressure difference across the membrane, and A is the membrane water permeability coefficient. The value of $R_{\rm m}$ and A are characteristic of the membrane in both cases and they are key in optimal design and operation of RO processes. The above mentioned coefficients depend (among other things) on the operating conditions and fouling potential of feedwater $(k_{\rm fp})[10]$ becoming a permeate flow decline in time. A few coefficients have been added to A in order to fit the model to real behavior of desalination plants, TCF (temperature correction factor), FF (fouling factor), etc. Maybe the most relevant coefficient due to operational, environmental and economic implications is the FF.

Schippers et al. [11] studied the permeate flow (J_w) decline (based on Modified Fouling Index (MFI)) because of fouling and two terms were added to R_m

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(Eq. (1)) coefficient, the first related to concentration polarization and the second one to a resistance of the cake. Although the fouling phenomena, as well as the development of new fouling-resistant membranes, has been extensively studied [10, 12-30], only a few authors [31-33] have proposed equations to estimate the decline of J_w in time due to variation of A in long-term. These correlations are applicable for the respective membrane type and for specific operating conditions. To obtain a model rigorous enough, it would be necessary to have long-term operating data in a wide range of operating conditions and different types of membrane. The mentioned model would depend not only on time, but on the characteristics of the membrane and the $k_{\rm fp}$.

A previously proposed model to predict the decline of J_w , due to membrane compaction (short-time periods), was used by M. Wilf et al. [31] to estimate the $J_{\rm w}$ decline in long-term. Three years of experimental data from different sea water reverse osmosis (SWRO) desalination plants were used to identify the parameter of the model. They calculated the parameter for permeate flow decrements of 25 and 20%. R.A. Mohamed et al. [34] used four years' data from a SWRO desalination plant with the TFC 2822 Fluid systemTM, initial feed pressure of 6.70 MPa and the flux recovery about 26-33%. Abbas et al. [32] proposed a model to determine the variation of the normalized average water permeability coefficient $A_n = A/A_0$, where A_0 is the initial average water permeability coefficient. It was an exponential equation depending on three parameters and time, the utilized membrane was the BW30-400 FilmtecTM. Five years of operating data were used for the parameter identification. The feedwater temperature was between 28 and 30 °C, the concentration in a range of 2,540 to 2,870 mg/L, and the feed pressure was around 1,200 kPa. Zhu et al. [33] also proposed a model to predict the coefficient A, it was an exponential equation, but in this case a hollow fiber membrane was utilized (DupontTMB-10) during one year of operating This correlation is not based on experiments time. but on model-based simulation: variable feed pressure (6.28-7.09 MPa), constant feedwater concentration and temperature (35,000 mg/L and 27 °C respectively). Belkacem et al. [35] used the Zhu model in terms of membrane resistance increase. The membrane used was the BW30LE-440 FilmtecTMin a two stage desalination plant with re-circulation during one year of operation. This model was not taken into account in this work since a proper fit was not achieved.

mainly due to membrane compaction, irreversible fouling (strongly adherent films) and $k_{\rm fp}$. The stage II is related to a gradual decrease mostly due to irreversible fouling, and frequency and efficiency of the chemical cleaning (CC). The operating conditions also play an important role in the decline of A, and therefore on model parameters. The model describes the mentioned stages by the superposition of two exponential functions. The work's aim was to identify the parameters of the different models to be compared. The experimental data were used in order to obtain three equations for each model. One related to maximum values of the normalized water permeability coefficient (A_n) (Post chemical cleaning (Post-CC)), average and minimum values (Pre chemical cleaning (Pre-CC)). This allowed to obtain equations to estimate a range of values for the coefficient A_n in time. The data of about 3,300 operating days of a brackish water reverse osmosis (BWRO) desalination plant were used to evaluate the different mod-2. Material and methods 2.1. Plant description

Two stages were differentiated in the decline of A.

Stage I shows a more pronounced decline than stage II,

The plant is located northwest of island of Gran Canaria (Canary Islands, Spain). This BWRO desalination plant and its operating data have been described in previous works [36, 37]. The feedwater is taken from a groundwater well located 1 km from the cost and 52 m hight. The well has a depth of 40 m and the feedwater flow was 25 m³/h. The plant (Fig. 1) had cartridge filters of 5 µm in the pretreatment stage and the antiscalant products used were Vitec 3000 (AVISTA® Technologies) and Osmotech 1141 (BKG Water Solutions, currently Kurita Water Industries Ltd), being 6 mg/L the dose in both cases. The RO system was equipped with 5 pressure vessels. The arrangement was 3+2 and the number of elements by pressure vessels was 6 (30 total RO elements). The membrane element used was the BW30-400 FilmtecTM, whose characteristics are available in the literature [38]. The production capacity was about 15 m³/h. This BWRO desalination plant was built for agricultural irrigation and has no post-treatment.

2.2. Operating data

This BWRO has been operating with the same membranes since June 2004. The data were collected monthly and the chemical analyses every two months. The feedwater source has a great impact on the RO

els.



Fig. 1. Desalination plant flow diagram

membrane fouling. High-quality source water, such as well water with silt density index (*S DI*) less than 3 (in this case 2.2-2.7), has a lower chance of fouling a RO membrane than lower quality source water, such as surface water (*S DI* of 5). Table 1 indicates the feedwater inorganic composition ranges. Fig. 2 shows the fluctuation of the feedwater conductivity no exceeding 13,120 μ S/cm. These fluctuations are due to many factors such as rain, temperature, etc. The drastic decrease in the day 274 was due to the plant's inactivity for two days (problems with the well pump) and the feed water conductivity decreased as the well water ceased to be agitated. The raw water temperature has been quite constant over the ten year period ~ 22°C.

Table 1Feed water inorganic composition

Ion	Concentration range (mg/L)
Ca ²⁺	68.14-336.47
Mg ² +	79.40-467.43
Na ⁺	635.90-2,319.92
K+	17.99-79.37
HCO ₃	505.25-1,041.61
$SO_4^=$	254.11-1,177.82
NO ₃	30.38-423.46
Cl ⁻	1,017.35-3,344.94
SiO ₂	27.50-46.00
TDS	3,144.70-7,790.76





Fig. 3 shows that the feed pressure was around 1,372.93 kPa at the beginning and increased up to approximately 2,353.60 kPa due to performance decay, specially fouling along these years. The feed pressure decreased in relation to feedwater conductivity and of course chemical cleaning (CC). Ideally, a cleaning is scheduled when the performance changes by 10% and should be completed by the time the performance has changed by 15%. Waiting too long to clean can result in irreversible fouling and/or scaling of the membrane. Membranes with good pretreatment can expect to be cleaned about 4 times per year depending on the quality of the feedwater. In this case, the chemical cleanings

were not always carried out when it should due to the owner's financial situation. As shown in Fig. 4 the permeate flow was oscillating around 15 m³/h due to feedwater conductivity variations and the efficiency of CC. Fig. 5 shows the permeate conductivity which has been in a range between 100 and 300 μ S/cm. The inorganic composition of the permeate water is shown in Table 2.



Fig. 4. Permeate flow

Fig. 6 shows the evolution of the A_n and when the CC were carried out, indicating increases in the mentioned coefficient (up to 35 % in the first 1500 days due to CC performance and not exceeding 20 % the following days). Chemical analyses were necessary to calculate the molal concentration, osmotic pressures and the coefficient A. As the chemical analyses and conductivities were measured every two months and monthly respectively, the molal concentration was in-



Fig. 5. Permeate water conductivity

 Table 2

 Permeate water inorganic composition

Ion	Concentration range (mg/L)
Ca ²⁺	0.40-2.40
Mg ² +	0.12-2.68
Na ⁺	19.08-60.00
K ⁺	0.78-3.52
HCO ₃	6.71-18.31
$SO_4^=$	0.48-8.17
NO ₃	3.72-35.39
Cl⁻	16.67-84.04
SiO ₂	0.15-0.42
TDS	48.11-214.88

terpolated for each month to have a value of coefficient *A* for each month. Eq. (3) shows how the coefficient *A* was calculated. The $k_{\rm fp}$ (Eq. (5) was calculated taking into account the data before the first CC since after this, the value of $k_{\rm fp}$ is "altered". The $k_{\rm fp}$ was 5.08 × 10⁹ Pa s/m².

$$A = \frac{J_{\rm w}}{(\Delta p - \Delta \pi)} \tag{3}$$

and

$$(\Delta p - \Delta \pi) = \left(p_{\rm f} - \frac{\Delta p_{\rm fb}}{2} - p_{\rm p} - \pi_{\rm fb} + \pi_{\rm p}\right) \qquad (4)$$

where $p_{\rm f}$, $\Delta p_{\rm fb}$, $\Delta \pi$, $p_{\rm p}$, $\pi_{\rm fb}$, $\pi_{\rm p}$ are feed pressure, average feed-brine pressure drop, average feed-brine osmotic pressure, permeate pressure and permeate osmotic pressure respectively. The $\pi_{\rm fb}$ and $\pi_{\rm p}$ were calculated using the ASTM standard D 4516 - 00.

$$k_{\rm fp} = \frac{\frac{1}{A_0} - \frac{1}{A(t)}}{\int_0^t v(t) \, dt}$$
(5)

where A_0 is the initial average water permeability coefficient and v(t) is the average permeate velocity. Thus the denominator is the cumulative volume in the time interval.



Fig. 6. Normalized average water permeability coefficient

3. Models

3.1. Existing models

Eqs. (6) and (7) were proposed by M. Wilf et al. [31] and Abbas et al. [32] respectively. Both models aim to describe the permeate flow decline in time or the variation of the coefficient A_n due to compaction, fouling etc. Fig. 7 shows the trend of each equation indicating a more pronounced decrease in the first 100 days for the Wilf et al. equations, but Abbas et al. equation shows a more gradual decrease.

$$A_{\rm n} = t^m \tag{6}$$

$$A_{\rm n} = 0.68 e^{\left(\frac{79}{t+201.1}\right)} \tag{7}$$

where *m* is a parameter with values between -0.035 and -0.041 [31] related to permeate flow decline of 20 and 25% respectively, *t* is the operating time in days.

3.2. Proposed model

To explain the philosophy of the proposed model, the Fig. 8 [32], which describes the decline of A_n of a different facility, was used. The model is based on the superposition of two exponential functions (Eq.(8)). The



Fig. 7. Normalized average water permeability coefficient decline according to literature correlations [31, 32] (Eqs. (6) y (7)) for a period of 500 days.

first is three parameters dependent (δ_1 , τ_1 and $k_{\rm fp}$), and it is related to the behavior in the stage I (Fig. 8), while the second is two parameters dependent (δ_2 , τ_2 and $k_{\rm fp}$) and it is more related to the stage II (Fig. 8). The first function gets closer to zero as the stage I ends. The δ are related with the weight of each exponential, the lower is δ_1 and the higher is δ_2 the higher is A_n when the desalination plant is stabilized. The τ concern to the decline in each stage (i.e. how fast is the irreversible effects (mainly fouling) affecting performance), the larger the value, the more constant is the function. Generally, the higher $k_{\rm fp}$ results en a faster decline of A_n in the stage I and II.

$$A_{\rm n} = \delta_1 \cdot \mathrm{e}^{-\frac{\tau}{\tau_1} \cdot k_{\rm fp}} + \delta_2 \cdot \mathrm{e}^{-\frac{\tau}{\tau_2} \cdot k_{\rm fp}} \tag{8}$$

3.3. Parameter identification

The parameters' identification of the mentioned models was calculated using the Nelder-Mead Simplex Method [39] implemented in MATLAB[®] software. The created function had as input the parameters of the models, $m, \alpha, \beta, \gamma, \delta_1, \tau_1, \delta_2$ and τ_2 (Eqs.(9), (10) and (8)).

$$A_{\rm n} = t^m \tag{9}$$

$$A_{\rm n} = \alpha \cdot e^{\left(\frac{\beta}{l+\gamma}\right)} \tag{10}$$

Data collected past the first 20 hours were considered as initial conditions, so the coefficient A_0 was calculated at that time and the different models were forced to pass through this point. Since the pre-CC and post-CC points are few in the operating time, an interpolation between those points was carried out to calculate the



Fig. 8. Schematic presentation of the two stages in A_n decline. (I) initial more pronounced drop due to compaction and irreversible fouling;(II) gradual decline mainly caused by irreversible fouling.

coefficient A_n in both cases. In these cases the error was measured considering the interpolation.

The index chosen in this paper is the standard deviation defined as:

$$\sigma_{\rm d} = \sqrt{\frac{\sum_{i=1}^{N} (y_{i,\rm exp} - y_{i,\rm calc})^2}{N-1}}$$
(11)

where $y_{i,exp}$ are the experimental data, $y_{i,calc}$ are the calculated data and N is the number of samples.

4. Results and discussion

Table 3 shows the values of the calculated parameters for each model. The standard deviation was quite close to zero in the three cases. Figs. 9, 10 and 11 show the experimental data of the coefficient A_n and the three curves obtained for each model.

Wilf et al. model was not found to be an acceptable estimator in the first 300 operating days of this BWRO desalination plant (Fig. 9,) and it was not possible to force this model through the initial point, so errors measured for this model cannot be compared directly with those of the other two models. The three functions estimated values of A_n below the experimental data but after that time, they fitted quite well. The corresponding curve for average values of A_n is closer to the post-CC curve than pre-CC due to compensation of the pronounced error in the first year. It should be taken



Fig. 9. Normalized average water permeability coefficient using Wilf et al. model [31].



Fig. 10. Normalized average water permeability coefficient using Abbas et al. model [32].



Fig. 11. Normalized average water permeability coefficient using the proposed Eq.

 Table 3 Calculated parameters for each model

	Wilf et al.	Abbas et al.				Proposed model		
Data	m	α	β	γ	δ_1	$ au_1$	δ_2	$ au_2$
Post-CC	-0.077	0.491	239.248	335.737	0.427	1.407×10^{12}	0.574	1.955×10^{14}
Average	-0.091	0.437	211.118	255.049	0.429	8.418×10^{11}	0.571	6.380×10^{13}
Pre-CC	-0.115	0.3907	134.594	142.391	0.534	9.359×10^{11}	0.468	8.539×10^{13}

Table 4 Standard deviation

Model	Post-CC	Average	Pre-CC
Wilf et al.	0.0068	0.0061	0.0050
Abbas et al.	0.0023	0.0046	0.0030
Proposed model.	0.0018	0.0044	0.0029

into account that it is one parameter dependent model, so it is not very flexible.

Fig. 10 shows the three fitted curves of Abbas et al. model. This model had a better fitting than Wilf et al. [31] as it is a three parameter dependent model. Therefore, Abbas et al. model showed to be closer to experimental data. Spite of using the same membrane, the values obtained for the parameters by Abbas et al. [32](Eq. (7)) are far from the values obtained in this work (Eq. (3)). The obtained equations had a more pronounced decrease in the first 1000 days of operation than Abbas et al., mainly due to $k_{\rm fp}$, frequency of CC, etc. The feedwater of this BWRO desalination plant had higher TDS than Abbas et al [32], where inorganic composition or SDI, or some indication on the fouling potential of feedwater was not contemplated, so further comparison was not carried out. The model depends on the time of operation and three parameters not taking into account the $k_{\rm fp}$ as i,t also happens with the Wilf et al. model [31].

The proposed model showed a good fit to the experimental data, being slightly better than the model proposed by Abbas et al. In the Fig. 12, the behavior of the exponential function of the proposed model is shown. Between the operating days 700 and 1,200, the first exponential operation tends to zero in all three cases, which is logical, since the value of τ_1 (8.418 × 10¹¹ for the average case) is low when comparing with τ_2 (6.380 × 10¹³ for the average case). From that moment, the decline of A_n is estimated by the second exponential. With the Abbas et al. and proposed models satisfactory estimates were obtained.



Fig. 12. Comparison of the proposed model in the three scenarios (post-CC, average and pre-CC), where Exp-I and Exp-II are the exponencial function in the stages I and II respectively

5. Conclusions

During ten years, data has been collected from a BWRO desalination plant; including antiscalant usage, CC products and frequency. After the second operating year, the coefficient A_n was fluctuating between 0.4 and 0.6. The evolution of A_n allowed to evaluate the predicting capacity of the above mentioned models. The parameters identification was carried out under three considerations (Post-CC, Average and Pre-CC) to obtain a range of values in which it would be appropriate to carry out a CC and an estimation of its effectiveness. A comparison between the existing models and the proposed model was done. Wilf et al. model showed a "poor" flexibility due to its dependence on only one parameter. Abbas et al. model showed to be more accurate with the disadvantage of being a purely empirical model. The parameters obtained by Abbas et al. and those calculated in this work differed markedly spite using the same membrane element. This is mainly due, amongst other things, to the fouling potential difference between both feedwaters. The proposed model proved to be slightly more accurate than Abbas et al. model in fitting terms. The main difference is that the Abbas et al. model is purely empirical, whereas the proposed model takes into account two different and usual behaviors in the decline of A_n in reverse osmosis systems considering $k_{\rm fp}$, which is an intrinsic property of feedwater. The model depends on operating time, $k_{\rm fp}$ and four parameters related to A_n in terms of operating conditions. This allows to have a predictive model in long operating time closer to the phenomenology concerning the decline of A_n , as opposed to existing models. One of the main objectives of this type of model is to carry out assessments on the evolution of the specific energy consumption, CC efficiency, and operating costs in long term.

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