

Desalination and Water Treatment

www.deswater.com

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51 (2013) 1116–1123 January



Integrated Membrane System (IMS) to treat brackish water with high salinity: a new treatment concept

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Received 15 March 2012; Accepted 15 June 2012

ABSTRACT

The drinking water treatment plant of the Consorci d'Aigües de Tarragona (CAT) is located in the town of L'Ampolla (Spain) and supplies drinking water in the province of Tarragona (345.000 m³/d). The raw water intake is located at the low river basin of Ebro river, which has sporadically high values in salinity. In this sense, a semi-industrial plant has been developed and operated by the CAT and Abengoa Water in order to obtain data for the development of a large-scale plant with a capacity of 250,000 m³/d. The main objective of these tests are maximised in the salt rejection and the global water recovery as well as minimised in the costs for the industrial plant. The idea was to develop and test different configurations of integrated membrane systems. Therefore, ultrafiltration and reverse osmosis were initially installed and tested. Nowadays, microfiltration and nanofiltration has been installed and tested with the existing systems. This paper presents the main characteristics of the pilot plant, results and conclusions obtained during the last operation stage up to the date.

Keywords: Brackish water; Drinking water; Integrated membrane system (IMS); Hollow fibre membrane; Microfiltration; Ultrafiltration; Reverse osmosis; Nanofiltration; Multi cartridge filter; Membrane filtration integrity

1. Introduction

The Consorci d'Aigües de Tarragona (CAT) is operating a drinking water treatment plant (DWTP) in the town of L'Ampolla, in the province of Tarragona (Spain). This plant supplies drinking water around 65 towns (near 1,000,000 people in summer) and 33 industries, with a pipe network of 392 km. In Fig. 1, it can be observed that the geographical location of the plant has a drinking water production capacity of $4 \text{ m}^3/\text{s}$ (345,600 m³/d).

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The raw water comes from two irrigation channels in the Ebro River (right and left side) before flowing into the Mediterranean Sea. This kind of water is continuously having problems of salinity [1–3]. The main source is geological, increased by anthropogenic activity, especially by the intensive agriculture in the area around the middle basin of the river [4]. Therefore, this fact produces anomalous values in order to fulfil the European Directive of Drinking Water [5] and its transposition to Spanish Legislation [6]. The episodes of high salinity are mainly due to the high concentration of chloride, sulphate, etc. reaching values of

Presented at the International Conference on Desalination for the Environment, Clean Water and Energy, European Desalination Society, 23–26 April 2012, Barcelona, Spain



Fig. 1. Geographical location of l'Ampolla DWTP: 40°48′04.54′′N; 0°40′20.07′′E.

 $270 \text{ mg Cl}^-/\text{L}$ and $370 \text{ mg SO}_4^{2-}/\text{L}$ which are higher than the established parametric value of 250 mg/L. These episodes usually last between three and six months per year. The Regional Health Authority has recently recommended the urgent solution of salty problems in order to supply drinking water with less salinity.

It is necessary to highlight that L'Ampolla DWTP mainly consists of: bar screening, straining, coagulation, flocculation-lamella settler, gravity sand filter, granular activated filter and a final disinfection with chlorine. These processes do not remove salts and for that reason, it has recently started the ELSA pilot plant project to study the best treatments and operation conditions to achieve its goal.

In this sense, the CAT will develop a new project in the DWTP with a new partial brackish treatment to remove salts (especially monovalent ions) from drinking water with a capacity of $233,280 \text{ m}^3/\text{h}$ (first phase $155,520 \text{ m}^3/\text{d}$). This water will be mixed with untreated drinking water in order to reach the parametric values specified by the legislation as previously stated.

This kind of solution has been previously implemented for similar DWTPs in Spain. At the moment, 4 plants are operating:

- El Atabal DWTP (Malaga): since 2004 with RO (two steps, recovery of 85% and a production capacity of $165,000 \text{ m}^3/\text{d}$)
- Abrera DWTP (Barcelona): since 2009 with EDR (recovery of 85–90% and a production capacity of 200,000 $m^3/d)$

- Sant Joan Despi DWTP (Barcelona): since 2010 with UF-RO (three steps, recovery of 85–90% and a production capacity of 198,720 m³/d)
- Almoguera DWTP (Madrid): since 2010 with UF-RO (two steps, recovery of 85% and a capacity of 172,800 m³/d)

In order to decrease the salt content in the drinking water in the L'Ampolla DWTP, the CAT and Abengoa Water designed, developed and operated the ELSA pilot plant to check the kind of systems applied for this case. Thus, the most appropriate operation conditions and design criteria for the industrial plant will be determined.

The ELSA pilot plant has been developed in a scale of 1:1,000 having a treatment capacity of $12 \text{ m}^3/\text{h}$ to treat salty drinking water from the DWTP. It has a filteration of salts and recovery higher than 90% in both cases as it will be shown later. The current paper presents the last obtained results up to date by comparing microfiltration (MF) vs. ultrafiltration (UF) and reverse osmosis (RO) vs. nanofiltration (NF).

2. Materials and methods

2.1. General overview of ELSA pilot plant

The feedwater to be treated in the pilot plant is the water (drinking water) let-out by the DWTP. The pump takes this water $(12 \text{ m}^3/\text{h})$ from the water tank of the DWTP till a storage tank installed at first of pilot plant. Here, the pH is corrected with CO₂ (approximately 5 mg/L) up to 7.

From this point, the water is pumped and treated by means of a MF or UF system (pressurised; out-in; dead-end); this step is named as MFM or UFM. In this paper the results will be focused on MFM (MF).

After that, water flows directly to a new storage tank where is taken up by a high pressure pump (1 mg/L of sodium bisulphite (37%) and 2 mg/L of antiscalant) to be desalted in the RO racks (brackish water). Here, the adopted configuration has been divided into three stages with 5, 2 and 1 pressure vessels, respectively. Between the second and third stage, a booster pump has been installed because, depending on each case, it might be necessary to increase the pressure by a few bars. The product water goes to a product water tank. In particular, this step is named OIM. In any case, both MFM/UFM and OIM form the part named M as it can be observed in the Fig. 2.

The brine of the previous RO (OIM) passes to a lamella settler equipped with flocculation to be treated in order to remove precipitates of salts because the brine is highly salty as well as non-balanced. After that, pH is adjusted to 7 by means of CO_2 (20 mg/L). Next, a new UF system is installed to remove small precipitated particles that can escape from the lamella settler; this step is named UFS and is similar to the previous UF (UFM). In this case, the flow and flux are around 2 m³/h and 40 lmh, respectively. The purpose is to avoid some particles that reach a new RO stage (OIS) where previously sodium bisulphite (1 mg/L (37%)) and antiscalant (7 mg/L) are dosed. In order to optimise the system, tests using NF membranes instead of RO membranes were carried out (NFS). The

new permeate is obtained to be mixed with the previous one from OIM. These steps, UFS and OIS/NFS forms the part named S.

All pumps are equipped with variable frequency drives. The UF system can be cleaned by backwashes with water and air free of oil in order to remove the materials on the UF membrane surfaces. On the other hand, a chemically nhanced backwash (CEB) can be applied using chemical reagents. The backwash waters and CEBs are treated by means of a new lamella settler equipped with flocculation in order to be recovered and treated again at the first pilot plant, reaching values around 99.99% (that means around $1 \text{ m}^3/\text{h}$). Finally, the pilot plant has an auxiliary clean-in-place (CIP) and permeated water displacement to be applied if it is necessary. The overall water recovery was 92.5%. Salt rejections were 93.9 % (M), 97.8% (OIS) and 94.2% (NFS). All brines and permeates are balanced as follows:

- OIM1: 60%; OIM2: 50% and OIM3: 40%.
- OIS or NFS: 50%.

The main characteristics and specifications of ELSA pilot plant are shown in the Table 1.

The pilot plant has installed a PLC and SCADA to control the process. Forthis purpose, the pilot plant is monitored online by means of instruments installed: pressure transmitters, differential pressure transmitters, flow metres, pH metres, temperature probes, conductivity metres (EC20°C), turbidity metres, OxRed potential transmitters, etc. Apart from online mea-



Fig. 2. Diagram of the ELSA pilot plant.

T T : L	Flam and a	Madal	Othora	
Unit	Elements	Model	Others	
M part				
Disc filter	1		PE 200 μm	
UF UFM (1+1)	3+3 (8´´)	DOW SFP-2860	PVDF 0.03 μm	
MF MFM (1+1)	16 + 16	Micronet porous fibres PVF/1800	PVDF 0.4 μm	
RO OIM 1	5×6 (4′′)	DOW LE-4040	Low energy brackish water element	
RO OIM 2	2×6 (4′′)	DOW LE-4040		
RO OIM 3	1×6 (4′′)	DOW XLE-4040	Ultra low energy brackish water element	
S part				
Flocculation	$1 \times 2 m^3$		2 h contact time	
Lamella settler	$1 \times 11 \text{ m}^2/\text{m}^3$	Tubodek	POLYPROPILENE	
Disc filter	1		PE 200 μm	
UF UFS (1+1)	1+1 (8´´)	DOW SFP-2860	PVDF 0.03 μm	
RO OIS ^a	1×6 (4'')	DOW XLE-4040 ^a	Ultra low energy brackish water element	
NF NFS	1×6 (4'')	DOW NF90-4040	NF element	
BW&CEB water treats	ment			
Coagulant			10 mg/L 40% FeCl3	
Coagulation	1×501		2 min contact time	
Floculant			1 mg/L PolyDADMAC	
Flocculation	$1 \times 1 m^3$		1 h contact time	
Lamella settler	$0.5\times 11m^2/m^3$	Tubodek	POLYPROPILENE	

 Table 1

 Main characteristics and specifications for ELSA pilot plant

^aIn the first part of the operation, DOW SW30HR LE-4040 was used.

sures, several offline measures are carried out such as: temperature, pH, conductivity, turbidity and chlorine.

2.2. Analytical methods

The general physical, chemical and microbiological parametres are the following to be monitored: pH, turbidity, conductivity (EC20°C), total dissolved solids (TDS), N⁺, K⁺, Ca²⁺, Mg²⁺, Sr²⁺, Ba²⁺, Fe^{2+/3+}, Mn²⁺, CO₃²⁻, HCO₃⁻, Cl⁻; SO₄²⁻, NO³⁻, SiO₂, heavy metals, total organic carbon (TOC), heterotrophic plate count (HPC) at 20°C, total coliforms, *Escherichia Coli*, faecal *Streptococcus* and *Clostridium perfringens*. All of them are based on AENOR water quality [7] and international standards [8].

Silt density index (SDI) at 15 min (SDI₁₅) follows the ASTM method D4189-95 [8]. Particle counting and size distributions are based on Standard Methods 2560 [9]. The characterisations of materials on the discs of the SDI were realised by Genesys Membrane Products, S.L.

Prior to carry out any test, in order to estimate the behaviour of the system, several specific softwares have been used: ROSA by DOW, AVISTA Advisor 3 by AVISTA, Master 3VC by GENESYS and RO 12.5 by NALCO. These softwares were used previously to estimate the behaviour of the processes. Finally, ionic equilibrium was studied to detect the scaling activity for the following compounds: $CaCO_3$, $CaSO_4$, $BaSO_4$, $SrSO_4$, CaF_2 , Fe, Al, Mn, SiO_2 and $Ca_3(PO4)_2$. In the raw water, (Table 2) there is no presence of Fe, Al and Mn (<DL) and the concentration of P in the raw water of the river is <0.05 mg P/L.

3. Results

This work presents the results of the pilot tests carried out during 50 weeks, between 2010 and 2012 and in particular, the last results up to the date. The startup of the pilot was at the beginning of 2010; during this phase, the main parametres of the system were set-up for each case: chemical doses, backwashings, CEBs, CIPs, etc.

One of the main challenges during the operation has been the use of brackish water membranes instead of seawater membranes for the OIS stage. Initially, the last process resulted in bad performance. The rejection of salts was excellent at the beginning, but quickly decreased and there was a necessityto increase the acid CIPs applied to maintain the percentage of salt rejection constant. This fact determined the use of brackish water membranes which improved the stability of this part of the process.

Table 2	
Pilot plant feedwat	ter quality

Parametres	Unit	Average	Maximum	Minimum
Turbidity	NTU	0.20	0.84	0.10
Temperature	°C	18.69	27.40	10.90
pH	Uds. pH	7.70	7.85	7.31
EC 20°C	μS/cm	986.28	1.376.00	512.00
TDS	Mg TDS/L	733.78	1.031.00	365.00
Sodium	mg Na ⁺ /L	95.19	155.50	32.30
Potassium	mg K ⁺ /L	2.71	4.40	1.00
Calcium	mg Ca ²⁺ /L	102.21	129.30	63.50
Magnesium	mg Mg ²⁺ /L	23.53	31.80	11.90
Strontium	mg Sr ²⁺ /L	1.93	3.37	0.84
Barium	μg Ba ²⁺ /L	29.64	42.22	18.02
Ammonium	mg NH ⁴⁺ /L	< 0.05	< 0.05	< 0.05
Aluminium	μg Al/L	<15	<15	<15
Iron	μg Fe/L	<15	<15	<15
Manganese	µg Mn∕L	<5	<5	<5
Carbonates	mg CO_2^{3-}/L	<6	<6	<6
Bicarbonates	mg HCO ³⁻ /L	180.53	204.10	147.00
Chlorides	mg Cl ⁻ /L	144.06	228.40	49.90
Sulphates	mg SO4 ^{2–} /L	204.75	308.60	86.20
Nitrates	mg NO ^{3–} /L	7.83	10.50	5.20
Nitrites	mg NO ^{2–} /L	< 0.01	< 0.01	< 0.01
Fluorides	mg F^-/L	0.17	0.19	0.15
Silica	mg SiO ₂ /L	3.75	5.40	1.20
Boron	mg B/L	< 0.1	<0.1	< 0.1
TOC	mg TOC/L	1.59	1.70	1.40
UVI 254	Abs	2.16	2.50	1.40
UVI 210	Abs	114.63	147.30	72.40
HPC at 20°C	CFU/ml	67	200	5
Total coliforms	CFU/100 ml	14	118	<1
E. Coli	CFU/100 ml	1	12	<1
Faecal streptococcus	CFU/100 ml	<1	<1	<1
Clostridium perfringens	CFU/20 ml	<1	2	<1

3.1. Pilot plant feed water

The feed water quality for pilot plant can be observed in the Table 2. It corresponds to the product water of DWTP. The ClO₂ is used in the DWTP to avoid the presence of Trihalomethane's. It only uses Cl₂ in the distribution network. In general, this water has a COD of 2 mgO₂/L, TOC < 3 mg C/L and the turbidity <1 NTU. The Electrical Conductivity 20°C and TDS range from 600 to 1,600 μ S/cm and from 400 to 1,200 mg/L, respectively.

3.2. MFM results

As explained previously, this paper will focus on MF tests, i.e. MFM stage. In the following paragraphs, the main results for the MFM are presented. It is neces-



Fig. 3. SDI for UFM and MFM.

sary to highlight that the system has been treating $12 \text{ m}^3/\text{h}$ at 85 lmh (with UF was implemented, the flux was 80 lmh) of water. Fig. 3 shows a group of SDI values (inlet vs. outlet) for MFM. In order to compare with UFM, data collected for UFM are presented. SDI₁₅ values are found to be better using UF (2.41) than MF (3.22). However, following the recommendations given by the manufacturers of RO membranes (the upper limit is 5), the values obtained with both systems are adequate and the treated water can be supplied to RO stage (OIS).

On the other hand, the organic matter rejection was unimportant and the particles $(1-100 \,\mu\text{m})$ were removed nearly 99%. The final results of the microbiological parametres were <1 CFU/100 ml. This fact indicates the low biological activity presented to feed the RO stage (OIM) and therefore, the low probability that biofouling appears on the RO membrane surface. This result is in accordance with the kind of water that is fed to the pilot plant which is the product water of DWTP.

Regarding the transmembrane pressure, the best values were found for MF (0.50 bar) compared to those found for UF (1.23). The chemical consumption (CEB and CIP) was similar for both cases

3.3. OIM results

This part of the process is the same for every test. Only at the beginning, some tests with seawater RO membranes were carried out. After that, they were replaced by brackish water RO membranes; this decision was based on the worse results obtained as explained previously. In consequence, from this point,

Table 3

Salt rejection percentage in th	he O	IM
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every test was carried out using the brackish water RO membranes.

The OIM is fed by the product water from the MFM with a constant flow of $11 \text{ m}^3/\text{h}$. By adjusting the valves and the rest of devices, the first stage (OIM1) worked at 60% of recovery, corresponding with a concentration factor (CF) of 2.5. The projections made with specific software at pH 7 did not show any scaling risk of CaSO₄, SrSO₄ and SiO₂ except for CaCO₃, BaSO₄ and CaF₂.

In the second stage (OIM2) there was a 50% of recovery with a CF of 5; this value did not show any scaling risk of $SrSO_4$ and SiO_2 .

Finally, the third and last sage (OIM3) resulted in a 40% of recovery with a CF of 6.7 which showed a high scaling risk. In this sense, it is necessary to highlight that selected antiscalant worked very well and therefore, an acid CIP should be applied every month, and as preventive measure, an EDTA CIP regularly.

Table 3 shows the percentage of salt rejection for divalent ions such as SO_4^{2-} (>99%) and Ca^{2+} and Mg^{2+} (\approx 98%) as well as monovalent ions with values around 90%. In terms of TDS, the rejection was around 93.9%. The global recovery for this part (MFM + OIM) was around 85%.

3.4. UFS results

For this part of the process, UF membranes were used every time and no changes were applied. For that reason, the results almost did not vary along the time, i.e. they remained constant.

Parametres	Unit	OIM feed	OIM perm.	% Rejection
Temperature	°C	16.5	18.5	
pH	Uds. pH	7.02	6.60	
EC 20°C	μS/cm	986	73	92.5
TDS	Mg TDS/L	734	45	93.9
Sodium	mg Na ⁺ /L	95	12.7	86.6
Potassium	mg K ⁺ /L	2.7	0.4	85.2
Calcium	$mg Ca^{2+}/L$	102	2.3	97.7
Magnesium	$mg Mg^{2+}/L$	23.5	0.5	97.9
Bicarbonates	mg HCO ³⁻ /L	180.5	20.8	88.5
Chlorides	mg Cl ⁻ /L	144	12.5	91.3
Sulphates	$mg SO4^{2-}/L$	205	1.5	99.3
Nitrates	$mg NO^{3-}/L$	7.8	3	87.2
Silica	mg SiO ₂ /L	4.0	0.8	80.0
UVI 254	abs	3.6	0.2	94.4
UVI 210	abs	122	56	54.1



Fig. 4. SDI for UFS.

Thus, the main results for the UFS are presented in the following paragraphs (Fig. 4). It is necessary to highlight that this system has been treating $2 \text{ m}^3/\text{h}$ at 40 lmh. In this case, the feedwater is the brine from OIM stage after being treated in a lamella settler with flocculation.

Thus, the suspended solids are totally different from solids found in MFM or UFM feed. The nature of these solids is mainly crystalline. This type of materials explain the hydraulic behaviour of the UFS. The TMP for UFS is less than for UFM due to the nature of the solids to be retained. Therefore, UFS presents permeability values around 2001mh/bar. In this case, a citric acid CIP was only applied. As a preventive measure, displacements were applied to remove salty water from the system to prevent scaling on the UFS membrane surface. 3.5. NFS results

In this case, during the previous weeks of operation, RO was tested. After that, in order to optimise the global process further, a NF has been tested. Therefore, as explained previously, this paper will be focused on NF tests, i.e. NFS stage. In the following paragraphs, the main results for the NFS are presented.

The NFS is the last treatment and is fed by the product water that comes from UFS (around $2 \text{ m}^3/\text{h}$) working at 50% of recovery. In Table 4, the percentage of ions rejected compared to the previous OIS tested was observed. In the case of OIS, divalents present higher values of rejection (>99%) than monovalents (>90%), except for the NO³⁻. For NFS, the global rejection is >90%. For monovalents, it was <90 and <45% for NO³⁻.

Table 4 shows the main results obtained for NFS in salt rejection percentages. At the same time, to compare with UFS, the results obtained previously are also presented.

To visualise the data collected in Table 4, Fig. 5 shows the different ion selectivity between NFS and OIS. As it can be observed, rejection for some divalent ions such as calcium, magnesium, sulphates, etc. is similar in NFS and OIS. However, rejection for some monovalent ions such a as sodium, potassium, bicarbonates, chloride, etc. is less in the NFS than OIS. As well, the nitrate salt rejection is very low being its final concentration <50 mg/L (parametric value of the European Water Directive).

Table 4 Salt rejection percentage in the OIS and NFS

Parametres	Unit	OIS			NFS		
		Inlet	Outlet	% Rejection	Inlet	Outlet	% Rejection
Inlet	Outlet	% Rejection	Inlet	Outlet	% Rejection		
EC 20°C	μS/cm	8,839.9	299.6	96.6	7,236.53	686.21	90.5
TDS	Mg TDS/L	8,659.4	189.3	97.8	7,137.21	414.47	94.2
Sodium	mg Na ⁺ /L	1,119.3	57.2	94.9	884.13	136.93	84.5
Potassium	mg K ⁺ /L	41.5	1.6	96.1	26.62	4.41	83.4
Calcium	$mg Ca^{2+}/L$	1,131.9	4	99.6	1,035.74	13.57	98.7
Magnesium	mg Mg ²⁺ /L	264	0.5	99.8	237.66	2.15	99.1
Bicarbonates	mg HCO ^{3–} /L	1,942.4	29.5	98.5	1,682.32	80.61	95.2
Chlorides	mg Cl ⁻ /L	1,809.3	76	95.8	1,414.67	183.36	87.0
Sulphates	$mg SO_4^{2-}/L$	2,607.3	3.1	99.9	2,314.35	4.61	99.8
Nitrates	mg NO ^{3–} /L	58.2	15.1	74.1	38.79	21.44	44.7
Silica	mg SO ₂ /L	32.7	0.7	97.9	30.62	2.72	91.1



Fig. 5. Salt rejection for NFS and OIS.

3.6. Overall results

The Integrated Membrane System (IMS) system tested consists of MF/UF-RO/NF divided in two parts: M and S in order to avoid spending product water from the DWTP. In general, it can be inferred that the performance is very good with a global salt rejection higher than 94% and being exactly 93.9% and 94.2–97.8% for M and S (NF-RO), respectively.

Therefore, the estimated cost is $0.0097 \notin /m^3$ ($\approx 0.01 \notin /m^3$) and $0.0362 \notin /m^3$ ($\approx 0.04 \notin /m^3$) for the part M and S, respectively, giving an overall cost of $0.0137 \notin /m^3$. The overall recovery is around 92.5%.

L'Ampolla DWTP has an average cost (reagents and energy) of $0.0173 \in /m^3$ ($\approx 0.02 \in /m^3$). According to the information obtained during these pilot tests, the average cost in the step M with MF is $0.0448 \in /m^3$, i.e. a final cost of $0.0621 \in /m^3$ (an increase of 259%) if all produced water is treated in the integrated membrane plant. On the other hand, it could be concluded that the decrease of salinity processes take six months and the rest of the year, the integrated membrane plant will not be working. Therefore, the awaited cost will be around $0.0397 \in /m^3$ (an increase of 129%). It can be highlighted that the final average cost will be increased up to $0.04 \in /m^3$ (nowadays $\approx 0.02 \in /m^3$). These costs mean an important increase in the final water cost.

The step S will increase the average cost around $0.0207 \in /m^3$ with RO and $0.0155 \in /m^3$ with NF. However, this stage will be reserved in other time to obtain an extra flow rate of desalted water.

Therefore, the final decision about what kind of technology will be finally implemented on industrial scale will depend on the equilibrium between the investment and the variable costs. For this case of study, the MF reduces the production cost around 5% and therefore, it could be considered as solution if the investment costs are adequate.

Regarding NF, it reduces the cost around 25%, but in general, the investment cost seems to be higher than in the case of RO.

In this case, one of the main goals for the near future will be to make consumers aware of the new water price.

4. Conclusions

- (1) The process proposed can obtain an overall recovery of 92.5% (high recovery) and an overall rejection of salts higher than 94%.
- (2) The UF system improves the performance of the RO, removing bacterial, flocculent materials, precipitates, etc. till reaching SDI values around 2.
- (3) The MF works like UF with less energy and therefore, it could be considered.
- (4) The NF can be an alternative to RO depending on the investment cost.
- (5) The energy cost in the part S is three times higher than part M due to the chemical cost are four times higher for the first one.
- (6) The industrial plant can be developed and constructed by phases to minimise the impact in the final cost of the product water. First of all, the part M will be installed to reduce salts with a recovery around 85%. Immediately, the part S will also be installed to reach the experimental recovery of 92.5%, supplying 150 L/s of additional product water.

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