Economic evaluation of salt recovery from wastewater via membrane distillation-crystallization

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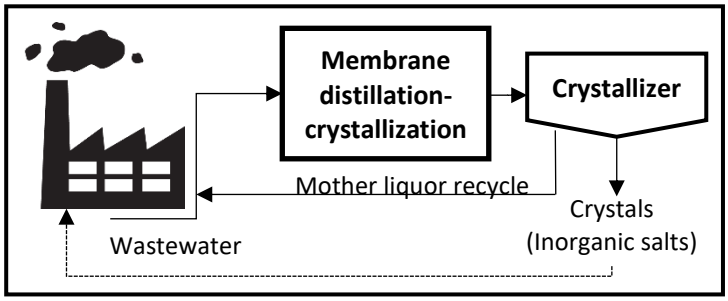
Abstract: Membrane crystallization is a promising method that could reduce the treatment costs of wastewater containing inorganic salts because of its easy functioning, its low energy consumption and the possible revalorization of the salts. However, very few economic evaluations have been conducted in this field of work, which makes the transition from laboratory scale to industrial levels difficult. This work addresses the economic analysis of crystallization of three inorganic salts i.e., sodium sulphate, sodium carbonate and potassium nitrate, using osmotic membrane contactors. Beforehand, the membrane area requirements are analysed because of their significant influence on process viability. Subsequently, the costs, salt sale profits and benefits are evaluated. The results show the membrane area requirements are strongly dependent on the concentration of the osmotic agent used in the contactor. Furthermore, a sensitivity analysis showed that the economic viability of the process is significantly influenced by the market value of the salts to be crystallized, the plant availability, the membrane price and the overall mass transfer coefficient of the membrane, suggesting that recovery of high-value salts or intensified research dedicated to low-cost and high-performance membranes could lead to a process substantially more performant and cost-effective.

Keywords: Membrane crystallization, membrane distillation, salts recovery, economic analysis.



Nomenclature	
a	Amortization factor [year^{-1}]
a_i	Activity of side i (feed/osmotic) [-]
A	Area [m^2]
A_{CC}	Annual capital cost [$ year^{-1}]
A_{CHEM}	Annual cleaning costs [$ year^{-1}]
A_{DISP}	Annual brine disposal cost [$ year^{-1}]

A_{ELEC}	Annual electricity cost [$ year^{-1}]
A_{LABOR}	Annual labor cost [$ year^{-1}]
A_{MAINT}	Annual maintenance cost [$ year^{-1}]
A_{MEM}	Annual membrane cost [$ year^{-1}]
A_{NaCl}	Annual NaCl [$ year^{-1}]
A_{OC}	Annual operating cost [$ year^{-1}]
A_T	Annual cost [$ year^{-1}]
b	Specific cost of brine disposal [$ m^{-3}]
c	Electric cost [$ kWh]
C_c	Crystallizer cost [$]
CE	Chemical engineering cost index [-]
C_{f0}	Initial feed concentration [g L^{-1}]
C_{ff}	Final feed concentration [g L^{-1}]
C_M	Membrane cost [$]
C_{o0}	Initial osmotic concentration [g L^{-1}]
C_{of}	Final osmotic concentration [g L^{-1}]
C_P	Pump cost [$]
D_{CC}	Direct capital cost [$]
f	Plant availability [-]
i	Interest rate [-]
I_{CC}	Indirect capital cost [$]
j	Parameter of interest
J	Transmembrane flux [L m^{-2} h^{-1}]
k	Specific chemical cost [$ m^{-3}]
K_{ov}	Overall mass transfer coefficient [mPa^{-1}s^{-1}]
M_{cf}	Mass of crystals produced [kg h^{-1}]
M_R	Membrane replacement [year^{-1}]
n	Plant life [year]
O	Objective function [$ year^{-1}] or [m^2]
P_f	Pressure at feed inlet [bar]
p^*	Vapor pressure [mmHg]
P_M	Membrane cost per area [$ m^{-2}]
P_{NaCl}	NaCl price [$ ton^{-1}]
P_{salt}	Salt price [$ ton^{-1}]
P_{SS}	Salt sale profit [$ year^{-1}]
Q_{evap}	Plant capacity [L h^{-1}]
Q_{f0}	Initial feed flow rate [L h^{-1}]
Q_{ff}	Final feed flow rate [L h^{-1}]
Q_{o0}	Initial osmotic flow rate [L h^{-1}]
Q_{of}	Final osmotic flow rate [L h^{-1}]
S_j	Sensitivity coefficient of parameter j [-]
sp	Spare parts [$ m^{-3}]
TWF	Total water fraction [%wt.]
UPC	Unit product cost [$ m^{-3}]
w	Electric power consumption [kWh m^{-3}]
γ	Specific cost [$ m^{-3}]
ρ_{NaCl}	NaCl density [kg L^{-1}]

Table 1: Nomenclature

1. Introduction

Industrial wastewater is an abundant source of valuable compounds. Salts such as carbonates, nitrates and sulfates, are typically present in wastewater coming from the textile, petrochemical and metal industries. If recovered with high purity, they can be reused, decreasing the stress generated by the intensive extraction of natural resources. Besides, these salts are naturally present in small quantities in unpolluted water but a high concentration in non-treated wastewater streams can result in environmental issues such as eutrophication [1], deterioration of the soil quality where this wastewater is used as irrigation material and increase of soil salinity or alkalinity [2], [3].

Therefore, efforts need to be put in place to treat wastewater and recover these inorganic substances. Among others, existing methods for removal of inorganic dissolved compounds are ion exchange, nanofiltration, reverse osmosis, electrolysis, chemical precipitation and evaporation [4], [5]. However, most of these treatment processes are complex and expensive and lead to solids in a sludge-form that do not allow easy reutilization [6]. For the purpose of salt recovery, crystallization seems to be ideal because it generates very high-quality products and has a high recovery rate [7]. Several crystallization techniques exist and have been compared in terms of applications, advantages and disadvantages in the context of wastewater treatment [8]. Among them, membrane distillation-crystallization is a promising method because of its numerous advantages: it can process highly concentrated streams, it is easily scaled-up, it can be operated at room temperature, and it permits a well-controlled supersaturation, crystal nucleation and growth [8]–[10]. Membrane distillation-crystallization is performed using membrane contactors, i.e., devices allowing a non-dispersive contact between two phases thanks to a hydrophobic membrane. Water evaporates in the pores of the membrane from the feed side to the permeate side due to a difference in chemical potential caused by activity, pressure and/or temperature gradients [11]. Crystallization occurs when the feed solution reaches supersaturation. The surface of the membrane promotes heterogeneous nucleation and crystals are then constantly driven away by the flow of the solution. They are then allowed to further grow in the crystallizer [12] while the mother liquor is continuously recycled from the crystallizer to the membrane contactor.

Membrane contactors offer a broad range of configurations such as direct contact membrane distillation (two liquid phases maintained at different temperatures), osmotic membrane distillation (DCMD driven by concentration gradient instead of temperature gradient), air gap membrane distillation (air gap at the permeate side), sweeping gas membrane distillation (sweeping gas applied downstream) and vacuum membrane distillation (vacuum at the permeate side) [13]. Direct contact membrane distillation is the most studied configuration in membrane distillation-crystallization. However, this configuration has a low thermal efficiency because of the heat losses and pronounced temperature polarization [14]. The high energy consumption for heating and cooling of the streams in direct contact membrane distillation are the main reasons for limited use in the industry [15]. Other configurations have also been studied for crystallization in terms of modelling, optimization and experimental work. Among others, Anisi *et al.* [16] studied sweeping gas membrane distillation for crystallization of L-ascorbic acid, Quist-Jensen *et al.* [17] studied the crystallization of lithium via vacuum membrane distillation-crystallization and Salmon *et al.* [18] studied osmotic membrane distillation for crystallization of several salts. Osmotic membrane distillation is advantageous compared to the others due to its operational simplicity, the possibility of working at room temperature, and the great availability of osmotic agents [19]. Waste streams such as concentrated brines could be used as osmotic solutions [9], reducing the cost of the process.

Despite the considerable potential of membrane distillation-crystallization and the demonstrated technological feasibility in several applications, very few economic evaluations have been performed in this field [9]. Some works include only a minor part about economics and most of the detailed economic evaluations about membrane distillation systems focus on desalination of seawater or backish water and production of pure water [20]. Kesieme *et al*. [21] compared membrane distillation with other desalination techniques and showed that membrane distillation is competitive with reverse osmosis and multistage flash when the heat source is inexpensive. Nonetheless, only a few authors analysed the economics of membrane distillation-crystallization for crystals recovery. Among them, Drioli *et al*. [22] studied different systems for seawater desalination. They demonstrated that the process including membrane distillation-crystallization considerably reduces the amount of brine produced and that salt sale can cover the desalination costs and even generate benefits. Wang *et al.* [23] studied membrane-assisted crystallization processes and concluded that membrane distillation-crystallization is competitive when the solubility of the solute is high. Quist-Jensen *et al*. [24] studied the economics of membrane distillation-crystallization of seawater as a magnesium source for phosphorous recovery from wastewater and showed that the process is economically feasible. Quist-Jensen *et al.* [17] studied membrane distillation-crystallization for lithium recovery from high-concentrated solutions but did not report their calculations. The lack of economic evaluations makes the transition from laboratory to industrial scale difficult. Therefore, efforts need to be made to study the economic point of view of this technology for various configurations and natures of crystals.

This work focuses on the economic evaluation of salt recovery from wastewater via osmotic membrane distillation-crystallization. The studied salts are sodium sulfate decahydrate ($Na_2SO_4 \cdot 10H_2O$), sodium carbonate decahydrate ($Na_2CO_3 \cdot 10H_2O$) and potassium nitrate (KNO_3). These salts have been chosen because of their common presence in effluents coming from the textile, petrochemical and metal industries [25]–[27]. Indeed, Na_2SO_4 and Na_2CO_3 can be found in wastewater from dyeing industry in concentrations of around 60 to 100 g L^{-1}, depending on the dye and the depth of colour desired [28]–[30]. Their concentration also reaches 60 g L^{-1} in alkaline leaching of uranium [31], and ethylene spent caustic is usually composed of 3 to 7 wt.% Na_2CO_3 [32]. The average concentration of nitrate in raw wastewater coming from 2-ehn production in the petrochemical industry is about 5.2 g L^{-1} [33]. Typical effluent concentration limits are 2000 ppm salinity in the textile industry [25], 250 to 7500 mg L^{-1} of sulfate in the metal industry [34] and 20 mg L^{-1} nitrate, 50 mg L^{-1} sulfate and 2000 mg L^{-1} Total Dissolved Solids in the petrochemical industry [35]. The technical feasibility of crystallization of sulfate decahydrate, sodium carbonate decahydrate and potassium nitrate via osmotic membrane distillation-crystallization has been previously demonstrated in [18] and some data have been extracted therefrom and integrated in the present study. A more detailed description of the experimental setup considered is described in [18]. The osmotic agent considered here is NaCl because of its great water activity difference with the three studied salts (cfr. Appendix A), high solubility and low risk of scaling [36]. First, this work evaluates the membrane area requirements for crystallization and the main influential process parameter. Then, an economic analysis is performed in order to determine the viability of the process with several operating conditions and highlight the main economic parameters affecting the potential benefits. A sensitivity analysis is performed using MATLAB software.

2. Working principle of osmotic membrane distillation

Figure 1 illustrates the working principle of a hollow fiber membrane contactor used for concentration of a feed solution using osmotic membrane distillation. The feed stream is introduced in the lumen side of the membrane whereas the osmotic solution flows in the shell side. A

configuration with the osmotic solution in the lumen side and the feed solution in the shell side is also possible. The non-dispersive contact induced by the membrane allows indirect contact of both streams through the pores. Membrane wetting is not desired, hence, a hydrophobic membrane material is selected. The difference in water activities between the feed and osmotic side is the main driving force that generates evaporation of the solvent (here water) from the feed to the osmotic side. A difference in temperature of both streams could be used as a second driving force in order to enhance the mass transfer but this was not considered in this study because it would require high extra capital and operating costs and the thermal efficiency is known to be low [14].

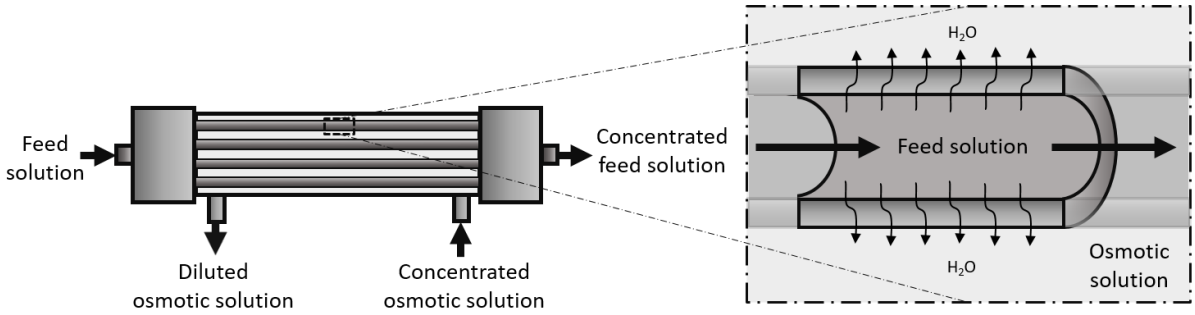


Figure 1: Operating principle of a hollow fiber osmotic membrane contactor. Water evaporates from the feed to the osmotic side through the membrane due to the difference in water activities.

3. Methodology

3.1. Process parameters

The forthcoming economic analysis depends on several process parameters that need to be evaluated beforehand such as the membrane area required to crystallize the salts, the total amount of water to be evaporated, and the mass of crystals obtained at the outlet of the continuous process. These parameters can be determined by studying the system shown in Figure 2:

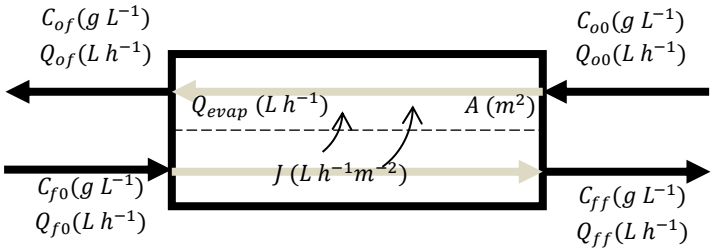


Figure 2: Scheme of the osmotic membrane contactor system for crystallization.

First, a simple mass balance allows determining the total amount of water per hour that has to be evaporated (Q_{evap}) in order to reach a supersaturated solution at the outlet of the feed stream:

$$Q_{evap} = Q_{f0} - Q_{ff}, \tag{1}$$

This quantity will represent the flow rate of water going through the membrane that is required in the treatment plant. The membrane area A can now be expressed as the flow rate of water going through the membrane Q_{evap} divided by the transmembrane flux:

$$A = \frac{Q_{evap}}{J}, \tag{2}$$

The easy linear scale-up of membrane contactors allows estimating the industrial needs from the intrinsic overall mass transfer coefficients K_{ov} obtained in lab-scale devices, as explained in Appendix B. K_{ov} can then be used to compute the fluxes and area requirements at industrial scale with a different driving force. In a previous study [18], Salmón *et al.* studied the influence of concentrations and flow rates on the transmembrane fluxes of a membrane contactor used for crystallization of $Na_2CO_3{\cdot}10H_2O$, $Na_2SO_4{\cdot}10H_2O$ and KNO_3 from synthetic wastewater. Mean flux values corresponding to initial feed and osmotic concentrations (C_{f0} and C_{o0}) of 100 and 300 g L^{-1} and initial feed and osmotic flow rates (Q_{f0} and Q_{o0}) of 18 and 27 L h^{-1} have been extracted from the work aforementioned in order to compute K_{ov}. Also note that, in their work [18], Salmon *et al.* did not report the presence of polarization phenomena, which could have lowered the value of the mass transfer coefficient.

Since no salt is permeating through the membrane, the flow rate at the outlet of the membrane when the final feed concentration C_{ff} is reached (Q_{ff}), can be expressed as:

$$Q_{ff} = \frac{C_{f0}Q_{f0}}{C_{ff}}, \tag{3}$$

The final feed concentration C_{ff} is defined here as the saturation concentration of the salts, i.e. the concentration at which the salts cannot be dissolved anymore in the solvent and hence crystallize (216, 192.3 and 316 g L^{-1} respectively for $Na_2CO_3{\cdot}10H_2O$, $Na_2SO_4{\cdot}10H_2O$ and KNO_3 at room temperature).

By combining equations (1) to (3) and equation (B.1) in the Appendix, the membrane area requirements become:

$$A = \frac{Q_{f0}}{K_{ov}(p_f^* a_f - p_o^* a_o)}\left(1 - \frac{C_{f0}}{C_{ff}}\right), \tag{4}$$

Note that the slight dilution and concentration during the process is not taken into account in the computations. Therefore, the activities are considered to be constant and to be equal to the initial activities. Finally, the mass of crystals produced at the end of the process M_{cf} needs to be determined. In their study, Salmon *et al*. [18] obtained crystals of sodium carbonate decahydrate ($Na_2CO_3{\cdot}10H_2O$), sodium sulfate decahydrate ($Na_2SO_4{\cdot}10H_2O$) and anhydrous potassium nitrate (KNO_3). Therefore, assuming that all the salt crystallizes, the mass of crystals obtained at the end of the process can be expressed as:

$$M_{cf,i} = \frac{C_{f0}Q_{f0}}{1000\ (1 - TWF_i)}, \tag{5}$$

where TWF_i is the total water fraction contained in the crystals of salt i (i.e. 67%, 58% and 6% for $Na_2CO_3{\cdot}10H_2O$, $Na_2SO_4{\cdot}10H_2O$ and KNO_3 respectively according to [18]). All the salt is assumed to crystallize at the outlet of the crystallization process.

3.2. Economic evaluation

In order to assess the viability of the process, the annual costs and salt sale profits need to be evaluated so that the final benefit is estimated. The benefit is defined as the difference between annual cost and salt sale profit. The annual cost A_T is defined as the sum of the annual capital cost A_{CC} (direct and indirect) and annual operating cost A_{OC}. The direct capital cost D_{CC} represents the purchase of equipment (pump, membrane and crystallizer) whereas the indirect capital cost I_{CC} is the contingency cost. The annual operating costs include the electricity necessary for the pumps A_{ELEC}, the cost of the osmotic solution A_{NaCl}, a labor cost for the employees A_{LABOR}, the maintenance cost A_{MAINT}, a

membrane replacement cost A_{MEM}, brine disposal costs A_{DISP} and cleaning costs using chemicals, A_{CHEM}. The relationships necessary to evaluate these costs have been extracted from the literature [37]–[39] and have been modified to suit the case of osmotic membrane crystallization for salt recovery from wastewater. The equations are provided in Table 2. Several assumptions were made for the economic analysis [18], [37]–[44]: (1) plant life is estimated to be 20 years, (2) interest rate is 5%, (3) module and membrane cost per area is 120 \$ m^{-2} (which is the price of the 14x28 Extra-Flow Liqui-Cel module, Membrana GmbH, Germany, including membrane and module [personal communication]), (4) electric cost is 0.09 \$ kWh, (5) plant availability is 0.9, (6) NaCl price is 50 \$ ton^{-1}, (7) specific cost is 0.05 \$ m^{-3}, (8) spare parts are 0.033 \$ m^{-3}, (9) membrane replacement rate is 25 % $year^{-1}$, (10) brine disposal costs via evaporation pounds costs 1.18 \$ m^{-3}, (11) specific chemical cost is 0.1 \$ m^{-3}, (12) $Na_2SO_4{\cdot}10H_2O$, KNO_3 and $Na_2CO_3{\cdot}10H_2O$ prices are 154, 700 and 156 \$ ton^{-1} respectively, and (13) electricity consumption is mainly due to pumping throughout the system (see Appendix C).

Costs	**Equations and assumptions**
Annual cost (A_T; \$ $year^{-1}$)	$A_T = A_{CC} + A_{OC}$
Annual capital cost (A_{CC}; \$ $year^{-1}$)	$A_{CC} = a(D_{CC} + I_{CC})$ $a = \text{amortization factor} = \frac{i \cdot (1+i)^n}{(1+i)^n - 1}$ $n =$ plant life = 20 years [43] $i =$ interest rate = 5% $year^{-1}$ [37]
Direct capital cost (D_{CC}; \$)	$D_{CC} = C_P + C_M + C_C$ $C_P = \text{pumps cost} = 0.0151 \cdot (Q_{f0} + Q_{o0}) \cdot P$ $P = \text{pressure} = 1.5 \text{ bar}$ $C_M = \text{membrane cost} = P_M \cdot A$ $P_M = \text{membrane cost per area} = 120$ \$ m^{-2} $C_C =$ crystallizer cost [39] $= \frac{\text{CE}}{500} \cdot 2.06 \left(2.4422\ 10^{-3} \left(\frac{24\, M_{cf}}{1000}\right)^3 - 5.3146 \left(\frac{24\, M_{cf}}{1000}\right)^2 + 5621.3 \cdot \frac{24\, M_{cf}}{1000} + 237830\right)$ CE = Chemical engineering cost index = 567.5 [44]
Indirect capital cost (I_{CC}; \$)	I_{CC}= 0.1· D_{CC} [37]
Annual operating cost (A_{OC}; \$ $year^{-1}$)	$A_{OC} = A_{ELEC} + A_{NaCl} + A_{LABOR} + A_{MAINT} + A_{MEM} + A_{DISP} + A_{CHEM}$
Annual electricity cost (A_{ELEC}; \$ $year^{-1}$)	$A_{ELEC} = c \cdot w \cdot f \cdot \frac{Q_{evap}}{1000} \cdot 24 \cdot 365$ $c =$ electric cost = 0.09 \$ kWh [37] $w =$ electric power consumption, kWh m^{-3} $f =$ plant availability = 0.9 [37]
Annual NaCl cost (A_{NaCl}; \$ $year^{-1}$)	$A_{NaCl} = \frac{Q_{o0} \cdot \rho_{NaCl}}{1000} \cdot P_{NaCl} \cdot f \cdot 24 \cdot 365$ $P_{NaCl} =$ NaCl price = 50 \$ ton^{-1} [37] $\rho_{NaCl} =$ NaCl density = 1.1085 kg L^{-1}
Annual labor cost (A_{LABOR}; \$ $year^{-1}$)	$A_{LABOR} = \gamma \cdot f \cdot \frac{Q_{evap}}{1000} \cdot 24 \cdot 365$ $\gamma =$ specific cost = 0.05 \$ m^{-3} [37]
Annual maintenance cost (A_{MAINT}; \$ $year^{-1}$)	$A_{MAINT} = sp \cdot f \cdot \frac{Q_{evap}}{1000} \cdot 24 \cdot 365$ $sp =$ spare parts = 0.033 \$ m^{-3} [38]

Annual membrane cost (A_{MEM}; \$ year^{-1})	$A_{MEM} = M_R \cdot C_M$ M_R = membrane replacement = 25% per year
Annual brine disposal cost (A_{DISP}; \$ year^{-1})	$A_{DISP} = b \cdot f \cdot \frac{Q_{of}}{1000} \cdot 24 \cdot 365$ *b* = specific brine disposal cost = 1.18 \$ m^{-3} [41], [42]
Annual cleaning costs	$A_{CHEM} = k \cdot f \cdot \frac{Q_{evap}}{1000} \cdot 24 \cdot 365$ *k* = specific chemical cost = 0.1 \$ m^{-3}
Salt sale profit (P_{ss}; \$ year^{-1})	$P_{ss} = M_{cf} \cdot P_{salt} \cdot f \cdot 24 \cdot 365$ P_{salt}: salt price (\$ ton^{-1}) [40], [45]
Benefit (B, \$ year^{-1})	$B = P_{ss} - A_T$

Table 2: Equations and assumptions for the economic analysis

3.3. Sensitivity analysis

Differential sensitivity analysis can be performed to compare the influence of the different parameters on the system. Two sensitivity analyses are performed in this work: the first one aims at studying the influence of the operating parameters on the membrane area requirements (i.e., initial osmotic concentration and initial feed concentration and flow rate) whereas the second one studies the effect of the economic parameters on the potential benefits *B* (i.e., plant life, interest rate, membrane cost per area, electric cost, electric power consumption, plant availability, NaCl price, specific cost, spare parts, membrane replacement rate and salt price). The effect of the mass transfer coefficient on the benefits is also studied in the second analysis because it is the major parameter characterizing membrane performance. Such information about sensitivity is crucial to determine the parameters with a high leverage on the economic viability that should be targeted first for process improvement in given conditions [46]. The relative sensitivity coefficient S_j of parameter j illustrates the normalized change in objective function *O* (area or benefit in this work) when a parameter changes. It is defined as the ratio of the relative partial derivative of the objective function to the relative partial derivative of the parameter [46], [47]:

$$S_j = \frac{\delta O/O}{\delta j/\mathrm{j}}, \tag{6}$$

This sensitivity analysis method assumes no correlation between the parameters and is only valid for small parameter variations. A positive (respectively negative) relative sensitivity coefficient means that an increase in parameter *j* results in an increase (respectively decrease) in absolute objective function. A coefficient equal to one suggests that the objective function is directly proportional to the parameter of interest. The bigger the absolute sensitivity coefficient, the higher the influence of the parameter on the objective function. Also note that the sensitivity coefficients are dependent on the objective function, as explicitly written in equation (6).

4. Results and discussion

First, a preliminary evaluation of system parameters has been performed to have the material required to complete the economic study. Among these system parameters, membrane area requirements are of uttermost importance because the membrane price will highly influence the process costs [48]. Therefore, special attention is given to membrane area requirements, which will be discussed before the economic evaluation. The forthcoming economic analysis is then performed first in terms of operating parameters and finally in terms of economic parameters. Lastly, a case study

example investigates some plant-related considerations and compares the process with conventional ways of producing the considered salts.

4.1. Membrane area requirements

Equation (4) expresses the membrane area requirements as function of the overall mass transfer coefficient, the feed flow rate, the initial and final feed concentration, the temperature (through the partial pressure) and the concentration of the osmotic solution (through the osmotic activity). A base case (at room temperature (20°C), initial feed concentration and flow rate of 100 g L^{-1} and 500 L h^{-1} respectively and initial osmotic concentration of 350 g L^{-1}) would require 2 068, 3 077 and 2 489 m^2 membrane area to crystallize $Na_2SO_4 \cdot 10H_2O$, KNO_3 and $Na_2CO_3 \cdot 10H_2O$ respectively. At first sight, this area seems quite large, but it is not in terms of occupied space: knowing that the area per volume ratio of membrane contactors typically ranges from 1500 to 3000 m^2 m^{-3} [49], the volume required would be lower than 2.1 m^3. This demonstrates that membrane contactors are genuinely compact compared to conventional equipment, such as mechanically agitated columns that offer 3 to 40 times less area per volume than membranes [50].

The influence of all the parameters involved in equation 4 is studied in this subsection by means of a sensitivity analysis. The sensitivity coefficients for the base case scenario of room temperature, initial feed concentration and flow rate of 100 g L^{-1} and 500 L h^{-1} and initial osmotic concentration of 350 g L^{-1} are summarized in Table 3. Firstly, it can be seen that K_{ov} and Q_{f0} influence the membrane area requirements to the same extent, no matter the salt to be crystallized. The opposite signs indicate that an increase in overall mass transfer coefficient leads to a decrease in area whereas the effect is opposite for an increase in feed flow rate. Secondly, the sensitivity coefficient of temperature, S_T, seems independent of the salt and has a larger influence than K_{ov} and Q_{f0}. However, the parameter that has the highest influence on the membrane area requirements is the initial osmotic concentration. This is because it is strongly influencing the osmotic activity and hence the driving force. The initial feed concentration shows a lower influence on the membrane area requirements, suggesting that it has a lower influence on the feed activity and hence on the flux and the driving force. This can indeed be verified in Figure A.1. This low influence of feed concentration on flux compared to the influence of the osmotic concentration had also been reported experimentally in [18], which confirms the results and the adequacy of the mathematical description.

	$S_{K_{ov}}$	S_T	$S_{Q_{f0}}$	$S_{C_{f0}}$	$S_{C_{ff}}$	$S_{C_{o0}}$
$Na_2SO_4 \cdot 10H_2O$	-1.00	-1.24	1.00	-1.00	1.08	-1.46
KNO_3	-1.00	-1.24	1.00	-0.36	0.46	-1.51
$Na_2CO_3 \cdot 10H_2O$	-1.00	-1.24	1.00	-0.71	0.86	-1.55

Table 3: Sensitivity coefficients for membrane requirements at room temperature with initial feed concentration and flow rate of 100 g L^{-1} and 500 L h^{-1} respectively, and initial osmotic concentration of 350 g L^{-1}.

4.2. Economic evaluation

First, the effect of the operating parameters (C_{f0}, Q_{f0}, C_{o0} and Q_{o0}) on the potential benefits is evaluated: Figure 3 depicts the benefits as function of the initial feed flow rate and feed concentration for the three salts, and Figure 4 illustrates the effect of the initial osmotic concentration on the benefits for the three salts considered. The initial osmotic flow rate is not deeply analyzed because of its marginal influence on the benefits (see Figure 4, annual NaCl cost). Secondly, the influence of the economic parameters is evaluated thanks to a sensitivity analysis summarized in Figure 5.

Figure 3 illustrates the benefits as function of the initial feed flow rate and feed concentration for the three salts with initial osmotic concentration and flow rate of 350 g L^{-1} and 27 L h^{-1} respectively. As a general trend, the benefits increase as the initial feed concentration increases. This is because less water needs to be evaporated if the feed concentration is higher, hence, the expenses decrease. Concerning the feed flow rate, the trend is quite different: depending on the initial concentration, increasing the feed flow rate increases or decreases the benefits. This means that the salt sale profit does not always increase faster than the diverse expenses when the feed flow rate is increased. This suggests that the initial feed concentration is of uttermost importance for the economic viability of the system: there is a minimum initial concentration that must be fed into the system so that benefits can be generated with increasing feed flow rates. Its value depends on the nature of the salt to be crystallized: around 75 g L^{-1} for $Na_2SO_4 \cdot 10H_2O$, 50 g L^{-1} for KNO_3 and 70 g L^{-1} for $Na_2CO_3 \cdot 10H_2O$, for the osmotic conditions mentioned above. In addition, Figure 3 also illustrates the fact that potassium nitrate always shows the highest benefits. This is principally due to its expensive price compared to sodium carbonate and sodium sulfate, which are more than four times less valuable. Indeed, their market values are around 700, 156 and 154 \$ ton^{-1}, respectively [40], [45]. The difference between the benefits generated by the crystallization of sodium sulfate and sodium carbonate is less significant. The salt-dependent parameters that can be responsible for the difference in benefits are the salt price, the total water fraction in the crystals, the saturation concentration and the overall mass transfer coefficient. Sodium carbonate has a higher saturation concentration and led to a lower overall mass transfer coefficient than sodium sulfate which would lead to lower benefits. Therefore, it can be concluded that the higher total water fraction (0.58 versus 0.67 %wt.) and higher salt price (154 versus 156 \$ ton^{-1}) are responsible for the better economic results of sodium carbonate compared to sodium sulfate.

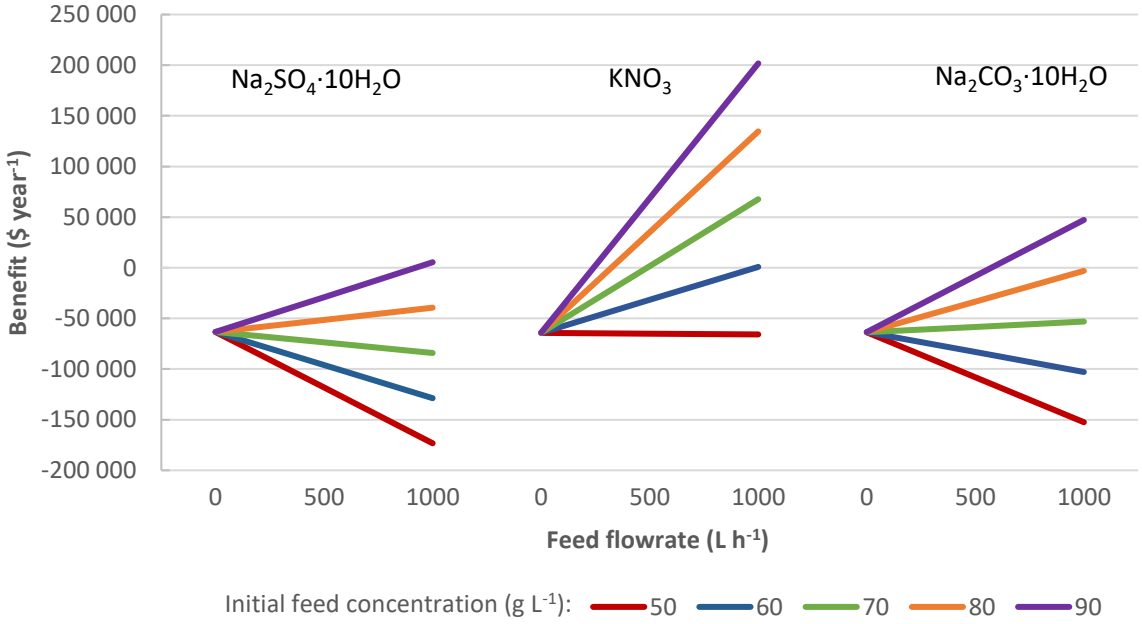


Figure 3: Benefits generated by the crystallization of the 3 salts as function of the initial feed flow rate and the initial feed concentration. Initial osmotic concentration and flow rate are 350 g L^{-1} and 27 L h^{-1} respectively.

Figure 4 illustrates the influence of the initial osmotic concentration (ranging from 150 to 350 g L^{-1}) on the economics of the process for the three salts, with initial feed concentration and flow rate of 100 g L^{-1} and 500 L h^{-1} respectively and initial osmotic flow rate of 27 L h^{-1}. It can be clearly seen that the osmotic concentration has a major impact on the economic viability of the process. Increasing this concentration leads to a higher driving force, leading to a higher flux, less membrane area and lower

costs. The authors recommend using the highest concentration possible (limited by the solubility of the salt i.e. 358 g L^{-1} at room temperature for NaCl) because of the marginal costs of the osmotic solution.

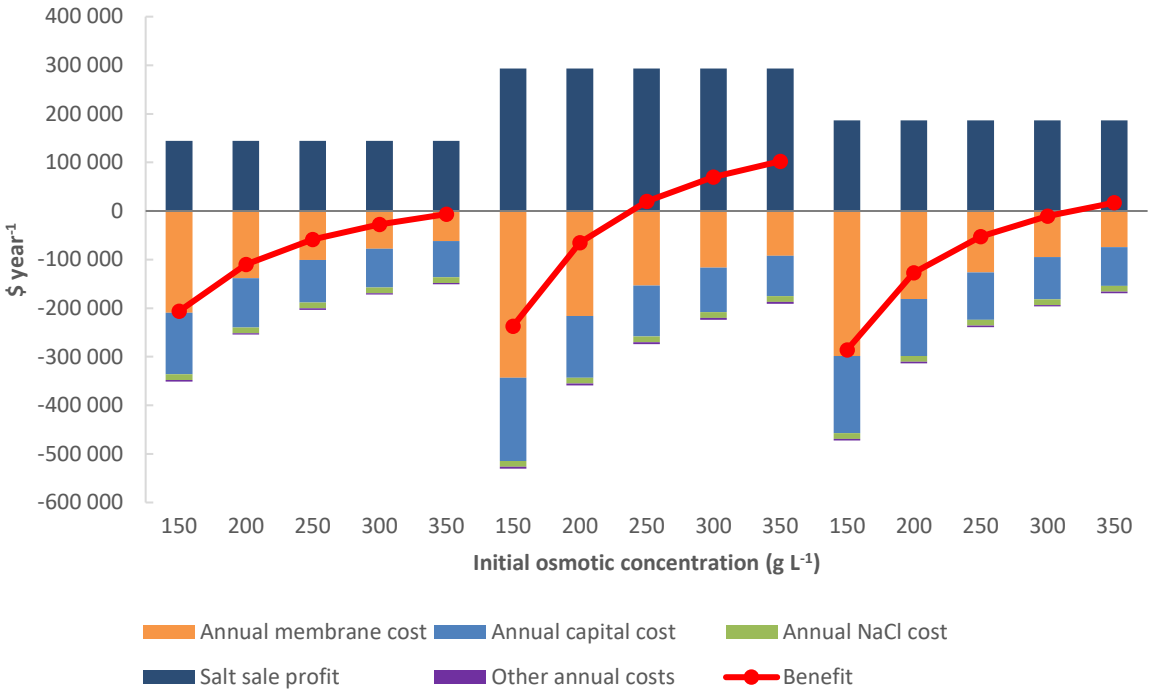
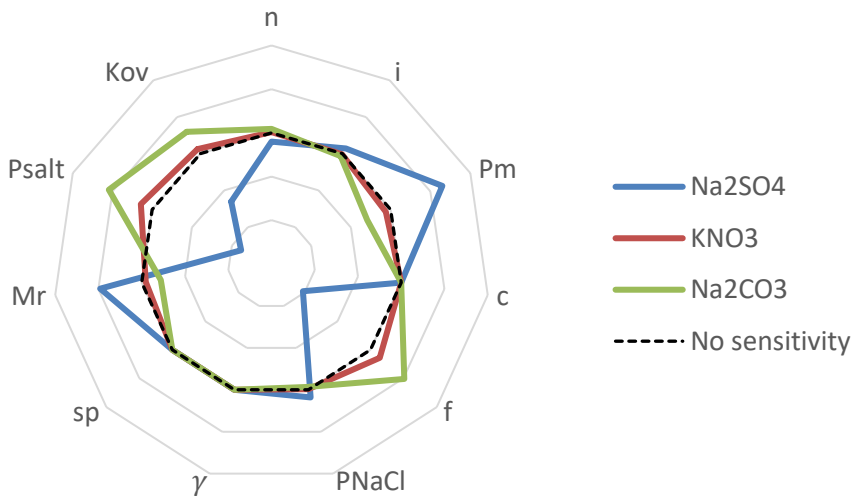


Figure 4: Costs, salt sale profit and benefit for the crystallization of the 3 salts. Different initial osmotic concentrations are illustrated. Initial feed concentration and flow rate are 100 g L^{-1} and 500 L h^{-1} respectively and osmotic flow rate is 27 L h^{-1}.

The contribution of the different costs is displayed in detail for a deeper analysis. Maintenance, labor, electricity, cleaning and disposal costs are gathered under the name of "Other annual costs" and can be considered negligible as they represent only 1 to 2% of the total expenses. This is due to several factors: the simplified equations used to do the calculation, the easy functioning of membrane which cuts down the maintenance and labor costs, the low feed streams studied (the maximum flow rate considered corresponds to 1 m^3 h^{-1}) and the low energy consumption of membranes due to the fact that only electrical energy is needed and no heating is required.

On the other hand, three main factors with a strong influence on the economic viability can be extracted from Figure 4: the salt sale profit, the annual membrane cost and the annual capital cost (purchase of equipment and contingency costs). In order to determine which economic parameters affect these factors, a sensitivity analysis has been performed according to section 2.4. The partial derivatives with respect to each economic parameter can be easily calculated thanks to the analytic formula for benefits given in Table 2. Figure 5 illustrates and sums up the sensitivity coefficients of all the parameters that were used for the economic analysis, for initial feed concentration and flow rate of 100 g L^{-1} and 500 L h^{-1} and initial osmotic concentration and flow rate of 350 g L^{-1} and 27 L h^{-1}.



	S_n	S_i	S_{P_m}	S_c	S_f	$S_{P_{NaCl}}$	S_γ	S_{sp}	S_{M_r}	$S_{P_{salt}}$	$S_{K_{ov}}$
$Na_2SO_4 \cdot 10H_2O$	-2.00	1.44	13.0	0.00	-20.5	1.83	0.01	0.01	9.60	-22.4	-13.0
KNO_3	0.19	-0.14	-1.22	-0.00	2.75	-0.12	-0.00	-0.00	-0.90	2.87	1.22
$Na_2CO_3 \cdot 10H_2O$	0.91	-0.66	-5.92	-0.00	10.2	-0.69	-0.01	-0.00	-4.38	10.9	5.92

Figure 5 : Sensitivity coefficients (S_x) of parameter x at room temperature (20°C) for initial feed concentration and flow rate of 100 g L^{-1} and 500 L h^{-1} and initial osmotic concentration and flow rate of 350 g L^{-1} and 27 L h^{-1}. n: plant life, i: interest rate, P_m : membrane price, c: electric cost, f: plant availability, P_{NaCl} : NaCl price, γ: specific cost, sp: spare parts, M_r : membrane replacement rate, P_{salt} : salt price, K_{ov} : overall mass transfer coefficient. The effect of the mass transfer coefficient of the membrane on the benefits is also studied here because it is the major parameter characterizing membrane performance.

Some parameters such as electric cost, specific cost and spare parts exhibit very small sensitivity coefficients, which confirms the previous conclusions about their negligible impact. The parameters with the highest degree of influence on the benefits are the salt price, plant availability and membrane cost per area. The extent of their influence depends on the salt studied. As can be seen in Figure 4, sodium sulfate does not generate benefits in the studied conditions, contrarily to the two other salts. Therefore, the sensitivity results displayed on Figure 5 display an opposite trend, but general observations and conclusions can be drawn for the three salts in these operating conditions:

1. The salt price always exhibits the highest (absolute) sensitivity coefficient in the studied operating conditions. This demonstrates that the salt price is a determining parameter for the economic viability of the process because it has a direct impact on the profit that can be generated. Therefore, salt recovery from wastewater via crystallization will be cost-effective only if the salt is valuable enough. This also suggests that the market price evolution must be monitored.
2. Plant availability (i.e., the actual amount of time the plant is able to run compared to ideal nonstop performance) shows a similar order of magnitude as the salt price. Because plant availability intervenes both in the process profit and costs, the sign of the sensitivity coefficients (negative for sodium sulfate and positive for the other salts) suggests that its influence on the profit generated is greater than on the costs incurred.
3. Membrane cost per area and overall mass transfer coefficient come in third place. Membrane cost per area is particularly interesting because it is the economic parameter with the greatest negative (resp. positive) coefficient for sodium carbonate and potassium nitrate (resp. sodium sulfate). It affects both the annual membrane cost (membrane replacement) and the annual capital cost (amortization), which are the highest costs (see Figure 4). This suggests that

reducing the membrane cost per area is the easiest way to limit the costs. The overall mass transfer coefficient has the same sensitivity coefficient as the membrane cost per area in absolute terms, which means that it can increase the salt sale profit as easily as the membrane cost can limit the costs. This study has been performed using the overall mass transfer coefficient of a particular membrane (i.e., the contactor used in [18]) but another membrane choice would have changed the results. Therefore, intensified research dedicated to low-cost and high-performance membranes could lead to a process substantially more performant and cost-effective than with the actual membrane prices and overall mass transfer coefficients.

4.3. Case study example

In order to show an example at industrial scale, the basic case of a plant able to process 500 L h^{-1} of 100 g L^{-1} initial feed concentration is studied in terms of membrane area, number of modules, module volume, crystallizer volume, unit crystal production cost and payback time required.

For such a plant, 11, 16 and 13 membrane modules would be required, corresponding to 1.32, 1.92 and 1.56 m^3 for sodium sulphate decahydrate, potassium nitrate and sodium carbonate decahydrate, respectively. The volume of crystallizer body with a retention time t_r can be estimated as expressed in equation (7) and would be around 1.30, 0.79 and 1.16 m^2 for a retention time of 5 h.

$$\text{Volume of crystallizer body } [\text{m}^3] = \frac{Q_{ff}\, t_r}{1000}, \tag{7}$$

The payback time is defined as the time needed to recoup the investment costs:

$$\text{Payback time [years]} = \frac{\text{D}_{\text{CC}}+\text{I}_{\text{CC}}}{\text{P}_{\text{ss}}-\text{A}_{\text{OC}}}, \tag{8}$$

The payback time is 14, 6 and 10 years for sodium sulphate decahydrate, potassium nitrate and sodium carbonate decahydrate, respectively. Additionally, the unit crystal production costs can be calculated in order to compare this process for resource recovery with market values. In these conditions, the unit crystal production of the plant would be 144.8, 410.4 and 127.5 \$ ton^{-1}. Comparison with the market prices (154, 700 and 156 \$ ton^{-1}, respectively [40], [45]) shows that recovery can be competitive. Table 4 summarizes the results obtained for the example at industrial scale:

Salt	Na_2SO_4	KNO_3	Na_2CO_3
Membrane area [m^2]	2068	3077	2489
Number of modules [-]	11	16	13
Membrane volume [m^3]	1.32	1.92	1.56
Crystallizer volume [m^3]	1.30	0.79	1.16
Unit crystal production cost [\$ ton^{-1}]	144.8	410.4	127.5
Payback time [years]	14	6	10

Table 4: Plant considerations for the base case with initial feed concentration and flow rate of 100 g L^{-1} and 500 L h^{-1} and osmotic concentration of 350 g L^{-1}.

5. Conclusions

The economic evaluation of an osmotic membrane distillation system for crystallization of inorganic salts present in wastewater streams has been performed. First, the membrane area requirements have been investigated in detail because the membrane price has an important impact

on the process costs. Then, an economic analysis has been performed in order to determine the viability of the process and highlight the main economic parameters affecting the potential benefits.

It was found that, under the studied conditions, the membrane area requirements are strongly dependent on the initial osmotic concentration and on the operating temperature, but less dependent on initial feed concentration. The total membrane area needed for crystallization turned out to be rather high, but not in terms of volume thanks to the high area per volume ratio of membrane contactors compared to conventional crystallization.

The economic analysis revealed that the three salts can generate profits depending on the operating parameters. Within the parameters range studied in this work, it can be concluded that: i) increasing the initial feed concentration leads to better economic results. This is because less water needs to be evaporated if the feed concentration is higher hence the expenses decrease. A pre-concentration step may be needed to reach the minimum profitable feed concentration; ii) low feed flow rates invariably lead to unprofitable results and increasing the feed flow rate leads to better economic results only if the initial feed concentration is high enough. This suggests that the initial feed concentration is of uttermost importance for the economic viability of the system; iii) higher osmotic concentration leads to better economic results because of the lower membrane area requirements. Because it does not affect the salt sale profit, the authors recommend to use the highest osmotic concentration possible (limited by the solubility of the salt) if the osmotic agent is cheap.

The sensitivity analysis showed the significant influence of salt price, plant availability, membrane cost per area and overall mass transfer coefficient on the benefits. Therefore, it can be expected that intensified research dedicated to low-cost and/or high-performance membranes could lead to a process even more cost-effective than with the actual membrane prices and overall mass transfer coefficients.

In conclusion, the economic viability of membrane crystallization via osmotic membrane distillation has been demonstrated, being the salt price, plant availability, membrane cost per area and overall mass transfer coefficient the main economic factors that affect the overall benefit.

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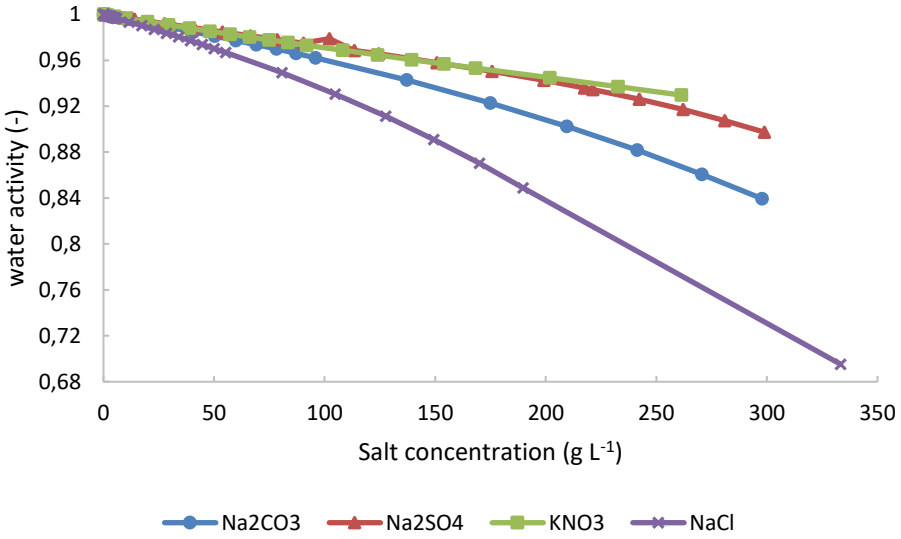
Appendix A



412

413 *Figure A.1: Water activities in function of the concentration. Reprinted and modified from* [18].

414 **Appendix B**

415 Experimental transmembrane fluxes *J* can be used to calculate the overall mass transfer coefficient K_{ov}
416 thanks to the following relation:

$$J = K_{ov}\left(p_f^* a_f - p_o^* a_o\right), \tag{B.1}$$

417 with *p** and *a* the vapor pressure and activity coefficient of the feed (*f*) and osmotic (*o*) side, which are
418 computed following the procedure described by Hamer *et al.,* [51] and by Sandler [52] when the value
419 of the osmotic coefficients were not found in the literature. The vapor pressure (mmHg) is given by
420 Antoine's equation, with temperature T given in °C:

$$p^* = 10^{8.07131-\frac{1730.63}{233.426+T}}, \tag{B.2}$$

421 The modular nature of membrane contactors makes the scale-up straightforward. The use of several
422 modules in parallel ensures similar fluido-dynamics and thermodynamics and enables the use of fresh
423 (non-diluted) osmotic solution in each module.

424 **Appendix C**

425 The electric power consumption of the plant is mostly governed by the recirculation, feed and osmotic
426 pumps. The recirculation pump is responsible for the mixing in the crystallizer (forced-circulation
427 crystallizer). Their efficiency is considered to be 0.7 [53] and the pressures at feed inlet (P_f) and
428 osmotic solution inlet (P_o) are fixed at 1.5 bar. The total electric power consumption (w) is calculated
429 as [54]:

430 $$w = w_f + w_o + w_{recirc}$$

431 $$w_f = \frac{P_f Q_{f0}}{36\,\eta\, Q_{evap}}$$

432 $$w_o = \frac{P_o Q_{o0}}{36\,\eta\, Q_{evap}}$$

433 $$w_{recirc} = \frac{9.81\ \rho\ h\ Q_{ff}}{3.6\ 10^6\ \eta\ Q_{evap}}$$

434 Where ρ is the liquid density at saturation and h is the differential head, estimated to be 7 m.

435 **Competing interests**

436 The authors have no competing interests to declare.

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