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Arsenic removal from drinking water by reverse osmosis: Minimization of costs and energy consumption



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ABSTRACT

Arsenic is one of the most serious inorganic contaminants in drinking water on a worldwide scale. To comply with the MCL (maximum contaminant level, $10 \mu g/l$ arsenic in drinking water) established by the World Health Organization, numerous techniques have been studied, such as ion exchange, coagulation and flocculation, precipitation, adsorption and membrane technologies. Among the available technologies applicable to water treatment, membrane filtration has been identified as a promising technology to remove arsenic from water.

The goal of this study is to demonstrate the technical and economic viability of removing arsenic (V) using an optimized reverse osmosis process, with minimization of the total cost as the objective of the optimization strategy. The optimization results showed that the total costs of a two-stage membrane cascade used for the removal of arsenic (V) from drinking water for a population of 20,000 inhabitants were 1041 \$/d and 0.52 \$/m³ of drinking water produced. Energy consumption was the most relevant cost, corresponding to 35% of the total cost. Sensitivity analysis was performed to determine the total costs of the installation for different scenarios in terms of drinking water production: (i) 0.44–0.56 \$/m³ for electricity prices of 0.05–0.10 \$/KW h; (ii) 0.88–0.45 \$/m³ for populations ranging from 5000 to 50,000 inhabitants; and (iii) 0.52–0.61 \$/m³ when the membrane lifetime was reduced from 3 to 1.5 years. The multiobjective optimization solutions, which consider the best compromises among the quality and cost objectives, indicated that the concentration of As (V) in the permeate water can be reduced to 0.5 μ g/l at a feasible cost.

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1. Introduction

Arsenic, which typically exists in natural waters in oxidation states of As(III) and As (V), is currently recognized as one of the most serious inorganic contaminants in drinking water on a worldwide scale. A long-term intake of arsenic causes serious chronic symptoms; therefore, the World Health Organization (WHO) has established a maximum contaminant level (MCL) of 10 μ g/l [18].

Arsenic is released from a variety of natural and anthropogenic sources. Arsenic occurs naturally in over 200 different mineral forms, of which approximately 60% are arsenates, 20% are sulphides and sulphosalts, and the remaining 20% includes arsenides, arsenites, oxides, silicates and elemental arsenic [7]. The primary anthropogenic sources of arsenic compounds are the mining industry, the pharmaceutical sector, enterprises producing glass and ceramics, pesticides, herbicides, dyes, woodworking enterprises, oil refineries and the metal smelting and melted alloy industries [10].

Note that for humans, exposure to arsenic through food or drink is more important than through the skin; the latter can be considered non-existent. The symptoms of arsenic poisoning caused by the consumption of drinking water are typically revealed over a period of 5–20 years, and certain consequences of arsenic exposure are irreversible [10]. The only solution to arsenic poisoning is to stop drinking the contaminated water. Diseases related to arsenic contamination are broad, ranging from a dry throat to cancer of the skin, lungs, urinary bladder, liver and kidney [5]. Therefore, arsenic is classified as a Group 1 carcinogenic substance to humans.

Currently, to comply with the maximum contaminant level, numerous techniques have been studied, such as ion exchange, coagulation and flocculation, precipitation, adsorption and membrane technologies [5]. Among the available technologies applicable to water treatment, membrane filtration has been identified as a promising technology to remove arsenic from water [15]. In addition, this technology can eliminate other types of ions or molecules. Membrane separation is addressed as a pressure-driven process, and

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operate costs (\$/d)

Nomenclature

A(i)	membrane area of the <i>i</i> stage (m^2)
AC	capital costs attributable to analysis (\$/d)
CC	capital costs (\$/d)
CCinst	capital costs attributable to installation (\$/d)
CCmemb	capital costs attributable to membranes (\$/d)
CF	concentration of arsenic in the feed stream (ppb)
Cp	concentration of arsenic in the permeate stream (ppb)
$C_{\rm F(i)}$	concentration of arsenic in the i feed stream (ppb)
$C_{P(i)}$	concentration of arsenic in the i permeate stream (ppb)
$C_{\rm R(i)}$	concentration of the arsenic in the i retentate stream
	(ppb)
$(C_{\rm S})_{\rm ln}$	logarithmic average solute concentration across the
	membrane (mol/m ³), defined by $\Delta C_s / \Delta (\ln C_s)$
C _{WHO}	maximum allowed concentration of arsenic by the
	World Health Organization (ppb)
F	initial feed flow (m ³ /d)
F(i)	feed flow of the i stage (m^3/d)
J_{V}	permeate flux (m/s)
$J_{V(i)}$	permeate flux of the i stage (m/d)
Js	flux of the solute due to the gradient of chemical poten-
	tial (mol/m ² s)
K _{memb}	ratio of the membrane capital costs to total capital costs
$L_{\rm P}$	hydraulic permeability coefficient (m/s bar)
LT _{memb}	membrane lifetime (d)
LT _{inst}	installation lifetime (d)
OC	operation costs (\$/d)

it is classified into four categories: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) [17]. Among these categories, the applications of the latter two have proven to be reliable in removing arsenic from water [16].

In recent years, substantial research efforts have been conducted to identify arsenic removal technologies that can be applied in rural areas. Membrane technologies are considered the best option for application in point-of-use filters or household filters to evaluate the arsenic levels in drinking water to ensure that they meet the MCL in rural areas or in certain arsenic-contaminated areas. Oh et al. [11] developed the concept of passing the fluid through RO and NF membranes using a bicycle pumping system, which is ideal for rural areas without electricity.

Different commercial membranes for nanofiltration and reverse osmosis systems have been reported in the literature, and the following studies are noted:

- Fang et al. ([6] evaluated the removal of arsenate using nanofiltration with DK and DL (GE Osmonics) membranes.
- Chang et al. [4] employed a nanofiltration membrane (Desal HL for General Electric Co.) and a low-pressure reverse osmosis membrane (Desal AK for General Electric Co.).
- Akin et al. [3] studied the removal of arsenic using a reverse osmosis technique with SWHR and BW-30 (Filmtec DOW) membranes.
- Uddin et al. [15] investigated the removal of arsenic from drinking water by the nanofiltration membranes NF-90 and NF-200 (Filmtec Dow).
- Saitúa et al. [12] studied arsenic removal from synthetic waters and surface water using the nanofiltration membrane 192-NF300 (Osmonics).
- Kosutic et al. [9] investigated the removal of arsenic and pesticides from natural groundwater by nanofiltration membranes NF270 and NFc (Filmtec DOW) and a reverse osmosis membrane CPA2 (Hydranautics); and

ocen	chergy costs (\$/u)
OC _{lab}	labour costs (\$/d)
OCm	maintenance costs (\$/d)
OC _{pt}	pretreatment costs (\$/d)
Р	permeate flow of the final stage (m^3/d)
P(i)	permeate flow of the <i>i</i> stage (m^3/d)
R	rejection coefficient
$R_{(i)}^{As}$	rejection coefficient of arsenic in the <i>i</i> stage
R(i)	retentate flow of the <i>i</i> stage (m^3/d)
Rec _(i)	recovery rate of the <i>i</i> stage
SF	safety factor
TC	total costs (\$/d)
Yelec	electricity price (\$/kW h)
Y_{lab}	salary (\$/h)
Ymemb	price of reverse osmosis membranes (\$/m ²)
Greek syr	nbols
ΔP	pressure difference across the membrane (bar)
$\Delta P_{(i)}$	pressure difference across the membrane in the <i>i</i> stage
()	(bar)
$\Delta \Pi$	osmotic pressure difference across the membrane (bar)
η	pump efficiency
σ	reflection coefficient
ω	coefficient of solute permeability (m/s)
ω'	modified coefficient of solute permeability (m/s)

 Oh et al. [11] tested the membrane HR3155 (Toyobo) for reverse osmosis and the membranes ES-10 (Nitro Denko) and HS5110 (Toyobo) for nanofiltration.

The effects of pH and arsenic concentration in the feed water, as well as the ionic strength and operating pressure on the rejection of arsenate and arsenite, were examined in these studies. Certain points to highlight include the following: (i) high pressure, high pH and low temperature favoured a more efficient removal of arsenic, whereas an increase in ionic strength reduced the removal of arsenate; (ii) the removal of arsenate is more complete than the removal of arsenite because at the pH that is typically used, arsenate exists in an anionic form, whereas arsenite is present in a neutral molecular form in aqueous solution, which complicates its rejection; and (iii) for the nanofiltration process, pre-oxidation of arsenite to arsenate can be required to obtain drinking water because the removal of arsenic is low, whereas better results were obtained with the reverse osmosis process, in which the pre-oxidation step was not required.

The use of an oxidizing agent, such as chlorine, was indicated for the improvement of the arsenic removal rate when arsenic in the source water is primarily present as As(III). However, oxidation is not a simple method to improve the efficiency because the oxidant could damage the membrane. Certain microorganisms can transform the arsenic oxidation state without the addition of an oxidant, providing a possible method to improve the efficiency of arsenite removal by combining membrane processes and biooxidation [13].

Considering that the referenced literature does not address the optimization of the membrane system for arsenic removal, the goal of this study is to use the process system engineering approach for the removal of arsenic (V) in aqueous solution by reverse osmosis to design and optimize the operation of this system under specific conditions. Experimental data for arsenic rejection and water permeate flux were obtained for a set of selected membranes to calculate the transport parameters. The transport equations (Kedem–Katchalsky model) and the mass balances (overall and component) used to describe the multistage membrane cascade were introduced into the optimization task, which was formulated in economic terms to calculate the process conditions for the minimum total costs and to evaluate the relevance of the energy consumption and the population in the process.

2. Experimental

2.1. Chemicals

A 100 ppb arsenate solution was prepared in ultrapure water by diluting the chemical compound Na₂HAsO₄7H₂O (PA-ACS for Panreac). Ultrapure water (18.2 M Ω cm resistivity) was obtained using a Milli-Q Element (Millipore).

2.2. Installation and reverse osmosis membranes

A lab-scale cross-flow flat-sheet configuration test unit (SEPA CF II from Osmonics) was purchased for the reverse osmosis experiments. The membrane cell can accommodate any 19 cm \times 14 cm flat-sheet membranes, resulting in 140 cm² of effective membrane area. Diverse commercially available polymeric flat-sheet RO membranes from different manufacturers were preselected for this study. The characteristics of the preselected membranes are summarized in Table 1.

The membrane cell was fed by a Hydra-Cell G-03 (Wanner Engineering) diaphragm pump equipped with a digital variable frequency drive to adjust the flowrate. The component materials were chosen to minimize the possible contamination of circulating water through the installation. HP PFA tubing was selected for the entire installation, except for the tube joining the pump and the cell (which worked under pressure), for which a PTFE tube enhanced with braided stainless steel was preferred, and the feed tank, which was constructed of polyethylene (PE).

2.3. Reverse osmosis experiments

Different tests were performed with the prepared solution. A constant feed flow of 3.4 l/min was maintained at a frequency of 22.9 Hz. Different applied pressures were maintained by the high-pressure concentrate control valve supplied with the membrane cell. The experiments were performed at room temperature and in total recycling mode, indicating that the permeate and retentate streams were continuously recycled to the feed tank, which assured constant characteristics in the feed stream during the entire experiment. The concentration of arsenic (V) in the feed tank was 100 μ g/l, and the target value for the permeate was 10 μ g/l (maximum contaminant level established by WHO).

As a first step prior to the experiments, the membranes were placed in ultrapure water to soak for at least 12 h. Then, each membrane was flushed with the feed liquid for 3.5 h at a feed pressure of 40 bar to ensure compaction of the membranes. In the experiments, the applied pressure in the system ranged from 10 to 40 bar. After sufficient time had passed to reach steady state conditions, the permeate flux was measured (weighing each sample and determining the time), and samples were obtained to determine the arsenate concentrations. Flux measurements and sample acquisition were performed in triplicate at 10-min intervals. All of the samples were analysed using inductively coupled plasma mass spectrometry (ICP-MS) with an Agilent 7500 ce ICP-MS system.

3. Results and discussion

3.1. Experimental results

The preselected membranes were used with doped water containing 100 µg/l arsenic (V) to compare and select the most appropriate membrane for the removal of arsenic. The applied pressure ranged from 10 to 40 bar, with measurements of the permeate flux and the arsenic rejection at each pressure point. From the experimental results, the dependence of the permeate flux (J_v) on the applied pressure (ΔP) was established. As shown in Fig. 1, the water fluxes increased with increasing pressure, displaying a linear relationship with high correlation coefficients. The linear evolution of fluxes shows that Darcy's law is verified. The slope of this straight line is the solvent permeability L_p , which is defined as J_v divided by ΔP .

A comparison of the permeate productions revealed that the BE membrane manufactured by Woongjin Chemical Co. was superior to the other membranes. The other membranes showed similar L_p values, but that of the BE membrane was more than twice those values, as shown in Table 2.

The determination of the efficiency of the preselected membranes for arsenic removal was also studied. The rejection coefficients (R) were defined by the following equation:



Fig. 1. Determination of the solvent permeability of the membranes for an aqueous solution with 100 μ g/l arsenic (V).

Table 2

Comparison of membrane solvent permeability for an aqueous solution with 100 $\mu g/l$ arsenic (V).

Membrane	Experimental L _p (m/s bar)
BE	5.0E-07
AD	1.2E-07
SW	2.0E-07
UTC	2.1E-07

Table 1

Primary characteristics of the selected flat-sheet membranes from the suppliers. Temperature 25 °C.

Designation	Manufacturer	Material	Permeate flow $(m^3/m^2 day)$	Rejection (%)	NaCl (mg/l)	Pressure (bar)
AD	GE Osmonics	Polyamide	0.61	99.5	32,000	55
BE	Woongjin Chemical	Polyamide	1.12	99.5	2000	15
SW30HR	Filmtec	Polyamide	0.66	99.7	32,000	55
UTC 80 B	Toray	Polyamide	0.57	99.75	32,000	55



Fig. 2. Arsenic rejection coefficients of the membranes for the pressure range of 10–40 bar. Points: experimental data. Lines: values from the application of the K–K model.

$$R = \frac{C_F - C_P}{C_F} \tag{1}$$

where $C_{\rm F}$ and $C_{\rm P}$ represent the arsenic concentrations measured in the feed and permeate streams, respectively.

As shown in Fig. 2, for all of the preselected membranes, the experimental rejection coefficients of arsenic are higher than 0.90 for the operational pressure range of 10–40 bar, and the values reach 0.98 at 40 bar in the cases of the BE and UTC membranes.

From the permeate flux results and the arsenic rejection coefficients of the different membranes, the BE membrane was considered the most promising membrane for arsenic removal.

3.2. Transport equations

The Kedem–Katchalsky model (K–K model) was selected to relate the permeate fluxes and rejection coefficients with applied pressure [14], as follows:

$$J_{\rm V} = L_{\rm P}(\Delta P - \sigma \Delta \Pi) \tag{2}$$

$$J_{\rm S} = \omega \Delta \Pi + (1 - \sigma) J_{\rm V}(C_{\rm S})_{\rm ln} \tag{3}$$

$$\frac{1}{R} = \frac{1}{\sigma} + \frac{\omega'}{\sigma} \frac{1}{J_{\rm V}} \tag{4}$$

Using the experimentally obtained values for permeate flux (J_v) and arsenic rejection (R) as functions of applied pressure, the estimation of the parameters in the Kedem–Katchalsky model was

 Table 3

 Estimated parameters from the Kedem–Katchalsky model for different membranes, including the overall explained data (%).

Membrane	σ	ω (m/s)	$L_{\rm p}~({\rm m/s~bar})$	Overall
BE	1	5.37E-07	5.04E-07	97
AD	0.972	4.23E-08	1.23E-07	83
SW	0.962	1.48E-07	2.03E-07	90
UTC	0.978	6.03E-08	2.07E-07	90

performed by a software tool (Aspen Custom Modeler). The osmotic pressure-related term in the equations describing the solvent flux was considered negligible. The obtained results for the estimation of parameters $L_{\rm p}$, ω and σ are given in Table 3. Fig. 2 shows the calculated rejection coefficient trends from the K–K model for the pressure range of 10–40 bar.

For the transport parameters in the Kedem–Katchalsky model (Eqs. (2)–(4)), three practical parameters are required to characterize each membrane + solution system: L_p is the hydraulic permeability of the membrane; ω is the solute mobility (or solute permeability); and σ is the reflection coefficient. The parameters L_p and ω strongly depend on the thickness of the selective membrane, and using these correlations between the practical parameters to compare different membranes is valid only when the results are normalized to layers of the same thickness. Moreover, σ and ω strongly depend on the nature of each solute, and they can be used to correlate the separation ability relative to the solute or solutes (also expressed in the values of rejection coefficients, *R*).

Eq. (4), which relates the rejection coefficient of each solute, *R*, to the transport parameters, can be rearranged as follows:

$$R = \frac{\sigma J_{\rm V}}{J_{\rm V} + \omega'} \tag{5}$$

Eq. (5) shows that the rejection increases with the increase of solution flux and reaches a limiting value σ at infinitely high J_v . Because the diffusive flux of the solute can be neglected in the range of high solution flux (operating at high pressures), the reflection coefficient (σ) is characteristic of the convective transport of each solute. A value of one indicates that no transport by convection occurs, which may be the case for ideal RO membranes that have a dense structure and no pores available for convective transport. As shown in Table 3, the reflection coefficient of arsenic for the BE membrane reaches a value of one, indicating that this component undergoes diffusive transport.

3.3. Optimization

The optimization task was applied to an *n*-stage BE membrane cascade, taking as a case study a water feed with an initial arsenic concentration of $2000 \ \mu g/l$ (contaminated water; [15,16]) and a final permeate volume flow of $2000 \ m^3/day$ considering a population of 20,000 inhabitants and a minimum quantity of 100 l per inhabitant per day.

Fig. 3 shows the scheme of the *n*-stage integrated countercurrent membranes; this configuration has also been used in previous publications by this research group [1,2].

The proposed optimization model is based on an economic model for the total cost minimization, including capital and operating costs. The objective function is subject to linear (overall and component material balances) and nonlinear equations (Kedem–Katchalsky transport equations) applied to each stage of the membrane cascade.

The variables introduced in the material balances are the feed, permeate and retentate volume flows of the streams involved in each i membrane stage (represented by F(i), P(i) and R(i),



Fig. 3. General scheme of an integrated countercurrent membrane cascade comprising n stages.

respectively), the corresponding arsenic concentrations, given as $C_{F(i)}$, $C_{P(i)}$ and $C_{R(i)}$, as well as *F* and *P*, which are related to the initial feed and final permeate streams, respectively.

The transport parameters estimated for the BE membrane were used for the optimization task. The characteristic variables that describe the performance of the i reverse osmosis stage, namely, the membrane-specific permeate flux ($J_{V(i)}$) and the rejection coefficient of arsenic ($R_{(i)}^{As}$), can be defined by the direct application of the Kedem–Katchalsky equations. Once the membrane transport is defined, the characteristics of the permeate streams (flow and arsenic concentrations) can be calculated as a function of the membrane area of the corresponding *i* stage ($A_{(i)}$), as follows:

$$P(i) = A_{(i)}J_{V(i)} \tag{6}$$

$$C_{P(i)} = (1 - R_{(i)}^{As})C_{F(i)}$$
(7)

The recovery ratio of each module $(Rec_{(i)})$ is defined as

$$\operatorname{Rec}_{(i)} = \frac{P(i)}{F(i)} \tag{8}$$

The total daily costs (TC), the formulated objective function to minimize, were defined as the addition of the capital costs (CC) and the operational costs (OC). The capital costs attributable to the membrane or to the remainder of the installation were differentiated, while the operational costs were itemized into pretreatment, labour, energy and maintenance costs, as follows:

$$TC = CC + OC \tag{9}$$

$$CC = CC_{memb} + CC_{ins}$$
(10)

$$OC = OC_{pt} + OC_{lab} + OC_{en} + OC_m$$
(11)

The capital costs of the membrane modules considering a straight-line depreciation were expressed as a function of the total membrane area of the installation:

$$CC_{memb} = \frac{Y_{memb} \sum A_i}{LT_{memb}}$$
(12)

The efficient lifetime of polyamide membranes ranges from 3 to 5 years (Kipper da Silva et al. [8]; thus, the value for the efficient lifetime of the membrane was fixed to 3 years with the intention of representing the most disadvantageous scenario.

Once the membrane costs were defined, the capital costs corresponding to the remaining installation were related using a coefficient (K_{memb}) that expressed the contribution of the investment attributable to the membranes. The term AC was added to include the costs of analysis, and its value was determined according to the research group's experience in analytical equipment management.

$$CC_{ins} = CC_{memb} \frac{(1 - K_{memb})}{K_{memb}} \frac{LT_{memb}}{LT_{inst}} + AC$$
(13)

The operational costs are essentially based on the consumption of the corresponding resource, except for cases of maintenance and pretreatment, which are a function of the total capital costs and the initial feed flow, respectively. The installation was designed to be fully managed by a single worker.

$$OC_{lab} = 24Y_{lab} \tag{14}$$

$$OC_{en} = \frac{\sum (F(i)\Delta P_i)}{36\eta} Y_{elec}$$
(15)

$$OC_m = 0.05 CC$$
 (16)

$$OC_{pt} = 0.035 F$$
 (17)

Table 4

Unit	Value
(\$/m ²)	50
(d)	1095
(d)	3650
	0.25
(\$/h)	7
(\$/KW h)	0.08
	0.70
(\$/d)	65.75
	Unit (\$/m ²) (d) (d) (d) (\$/h) (\$/KW h) (\$/d)

Table 5

Optimization problem formulation.

Objective function	TC
Optimization target	Minimization
Independent variables	$\operatorname{Rec}_{(i)}, \Delta P_{(i)}$
Constraints of independent variables	$0.3 < \text{Rec}_{(i)} < 0.9$
	$10 < \Delta P_{(i)} < 40$
Other constraints	Conc Prod < Conc WHO

The values for all of the parameters appearing in the economic model can be observed in Table 4.

All of the model variables are expressed in terms of the independent design and operational variables, namely, the recovery rates (Rec_i) and applied pressures (ΔP_i). Constraints for the independent variables were established, as shown in Table 5. The valid interval of the recovery rate was defined as 0.3–0.9. Recovery rates close to one may cause polarization problems, and recovery rate values close to zero imply low production rates. The pressure range was chosen for safe operating conditions considering the maximum pressure recommended by the manufacturer of the BE membranes.

GAMS software was used as an optimization tool to manage the presented nonlinear programming (NLP) model using the BARON solver. The General Algebraic Modeling System (GAMS) is a highlevel modelling system for mathematical programming and optimization, and it consists of a language compiler and a number of integrated high-performance solvers.

The optimization results to achieve the minimum cost of a membrane system for the treatment of drinking water are compiled in Table 6. The installation includes two stages of reverse osmosis membrane modules, producing a final permeate stream with an arsenic concentration of $3.3 \mu g/l$, which is less than the maximum contaminant level of $10 \mu g/l$ [18].

The optimum applied pressure for both of the stages was 24.4 bar, and both of the optimum recovery rates were 0.9, which is the upper boundary restriction for these variables.

Under these conditions, the minimum total cost for the membrane system is 1041/d, which corresponds to a cost of 0.52/m³ of drinking water produced for a population of 20,000 inhabitants.

As shown in Fig. 4, the item with the highest cost is related to electricity consumption, which was 35% of the total cost for the membrane system. Considering these results, the relevance of the energy cost is discussed in the following section.

3.4. Influence of the price of electricity on the optimization model

A sensibility analysis was performed to study the influence of the price of electricity on the principal variables of a two-stage integrated optimized installation. Fig. 5 shows the influence of the price of the electricity on the capital costs and the operating costs, resulting from the total cost minimization. Table 7 shows

Table 6

Optimization results: Minimum costs of the membrane system for arsenic removal of drinking water. Population: 20,000 inhabitants.

$\Delta P_{(i)} (bar)^{a}$ Rec _(i) ^a	24.4 0.9	
$C_P (ppb)$ $\Sigma A (m^2)$	3.3 3984	
Economic terms TC CC CC _{memb} CC _{inst}	(\$/d) 1041 411 182 229	(\$/m ³ water) 0.52 0.20 0.09 0.11
$\begin{array}{c} OC\\ OC_{pt}\\ OC_{lab}\\ OC_{en}\\ OC_{m} \end{array}$	630 79 168 363 20	0.32 0.04 0.09 0.18 0.01

^a i = 1, 2.



Fig. 4. Cost breakdown for the optimum configuration of the membrane system.



Fig. 5a. Influence of the price of electricity on the capital costs.

the daily total results and the energy cost contribution when the price ranged from 0.05 to 0.10 /KW h.

The total cost of the installation in terms of the drinking water production significantly increases from 0.44 to 0.56 m^3 (for 0.05–0.10 KW h electricity prices).

Table 7 also includes the changes of the process conditions, such as the applied pressure, the concentration of the permeate stream and the total area. Lower operating pressures are calculated when the total costs are minimized under higher electricity prices, resulting in a water permeate stream of lower quality (Cp, arsenic concentration from 2 to 4 ppb, which is less than the WHO concentration).



Fig. 5b. Influence of the price of electricity on the operating costs.







Fig. 6b. Influence of the number of inhabitants on the operating costs.

3.5. Influence of the population size on the optimization model

A sensibility analysis was also performed to study the influence of the population size (number of inhabitants) on the principal variables of a two-stage integrated optimized installation. The population size varied over a range of 5000–50,000 inhabitants, which are small populations of great interest for the membrane systems.

Fig. 6 shows the capital costs and the operating costs resulting from the total cost minimization. The results conclude that the critical costs of the reverse osmosis installation are the membrane modules (CC_{memb}) and energy consumption (OC_{en}), which increase significantly with the population size using the membrane system for arsenic removal from drinking water.

As shown in Table 8, the optimization results of the total costs are 440–2250 \$/d over a range of 5000–50,000 inhabitants, with the increase of the cost related to the total area of the membrane

Table 7

Optimization results of total cost minimization of the membrane system for different prices of electricity. Population: 20,000 inhabitants.

Yelec (\$/KW h)	0.05	0.06	0.07	0.08	0.09	0.10
$\Delta P_{(i)} (bar)^{a}$ $Rec_{(i)}^{a}$ $C_{P} (ppb)$ $\Sigma A (m^{2})$	30.8	28.1	26.1	24.4	22.9	21.8
	0.9	0.9	0.9	0.9	0.9	0.9
	2.1	2.5	2.9	3.3	3.7	4.1
	3149	3450	3726	3984	4225	4454
TC (\$/d)	889	944	994	1041	1085	1127
(\$/m ³ water)	0.44	0.47	0.5	0.52	0.54	0.56
OC _{en} (\$/d)	287	314	339	363	385	406

^a i = 1, 2.

Table 8

Optimization results of total cost minimization of the membrane system for different populations.

Inhabitants	5000	20,000	50,000
$\Delta P_{(i)} (bar)^{a}$	24.4	24.4	24.4
Rec _(i) ^a	0.9	0.9	0.9
$C_{\rm P}$ (ppb)	3.3	3.3	3.3
$\Sigma A(m^2)$	996	3984	9959
TC (\$/d)	438	1041	2248
(\$/m ³ water)	0.88	0.52	0.45
OC_{en} (\$/d)	91	363	907

^a i = 1, 2.



Fig. 7a. Influence of membrane life on the capital costs.

system. The total costs per volume of drinking water are calculated as $0.88-0.45 \text{ }/\text{m}^3$ for the population range.

3.6. Influence of the effective membrane life on the optimization model

A sensibility analysis was performed to study the influence of the effective membrane life on the principal variables of a twostage integrated optimized installation and to quantify the effect of possible fouling in a long-term RO operation. The membrane life ranged from 1.5 to 3 years (547–1095 days).

As shown in Fig. 7, the longer the membrane life, the lower the capital costs and the operating costs, with the exception of capital costs due to installation, which increase.

Table 9 presents the changes in the optimization results when the membrane life is altered. The results show that the cost per unit of water volume increases from 0.52 to $0.61 \text{ }^{\text{s}}/\text{m}^{3}$ (17% increase) due to a 50% reduction in the membrane lifetime.

3.7. Multiobjective optimization: Quality versus economic criteria

The quality is formulated as a dimensionless safety factor, SF, defined as the quotient between the limit concentration, $C_{\rm WHO}$



Fig. 7b. Influence of membrane life on the operating costs.

Table 9

Optimization results of total cost minimization of the membrane system for different membrane lifetimes.

Membrane life (d)	547	657	766	876	985	1095
$ \begin{aligned} & \Delta P_{(i)} \ (bar)^a \\ & \text{Rec}_{(i)}^a \\ & C_P \ (ppb) \\ & \Sigma A \ (m^2) \\ & \text{TC} \ (\$/d) \end{aligned} $	30.1	28.3	27	25.9	25.1	24.4
	0.9	0.9	0.9	0.9	0.9	0.9
	2.2	2.5	2.7	2.9	3.1	3.3
	3224	3427	3598	3745	3872	3984
	1212	1159	1119	1088	1062	1041
(\$/m ³ water)	0.61	0.58	0.56	0.54	0.53	0.52
OC _{en} (\$/d)	448	422	402	386	373	363

^a *i* = 1, 2.

(fixed by WHO), and the concentration of arsenic in the permeate stream, C_P :

$$SF = \frac{C_{WHO}}{C_{P}} = \frac{Failure \ concentration}{Design \ concentration}$$
(18)

The multiobjective optimization implies the evaluation of all of the feasible solutions to the problem, which consider the best compromises among the objectives. These solutions are those in which an improvement in one objective can only be achieved by accepting a deterioration in at least one other objective. The respective solutions, which are not dominated by any other solutions, are called Pareto optimal points, and the set comprising all of these solutions is the Pareto set, also called the Pareto frontier because it lies on the border between the dominated and not dominated solutions.

The Pareto set of the problem resulting from the simultaneous minimization of total costs and maximization of the quality of the obtained solution is depicted in Fig. 8. The solutions were obtained via the epsilon constraint method, which addresses the



Fig. 8. Pareto set for a two-stage cascade for arsenic removal from drinking water.

multiobjective optimization problems by solving the corresponding series of single objective subproblems in which all of the objectives, except for one, are transformed into constraints.

An analysis of the results shown in Fig. 8 reveals that two different zones can be distinguished: a slowly increasing curve from the minimum total costs (TC = 1041 \$/d, 3.3 ppb As in the permeate water) to the point corresponding to an SF value of seven and a steeper linear zone, which continued until reaching the maximum quality solution corresponding to an SF value of 19.6 (0.5 ppb As in the permeate water) and total costs of 2550 \$/d.

4. Conclusions

Four polyamide reverse osmosis membranes, AD (GE Osmonics), BE (Woongjin Chemical), SW30HR (Filmtec) and UTC 80B (Toray), were evaluated to calculate the transport parameters required for the process simulation and optimization tasks. The BE membrane provides the best results for the removal of arsenic (V) from an aqueous solution, as well as the highest solvent permeability and rejection coefficient values.

The Kedem–Katchalsky model can be considered a suitable representative for the removal of arsenic (V), as demonstrated by its agreement with the experimental results.

The optimization results showed the total costs (1041 \$/d; 0.52 \$/m³ drinking water produced) of a two-stage membrane cascade used for the removal of arsenic (V) from drinking water for a population of 20,000 inhabitants. The energy consumption corresponds to the most relevant cost, representing 35% of the total cost.

The variability in the price of electricity results in changes in the process conditions, such as the applied pressure, the concentration of the permeate stream and the total area required for the membrane system. Lower operating pressures are calculated when the total costs are minimized under higher electricity prices. The total costs of the installation in terms of the drinking water production increase significantly from 0.44 to 0.56 \$/m³ (for 0.05–0.10 \$/KW h electricity prices).

According to the process conditions, the number of inhabitants only influences the total area for the membrane system when the total costs are minimized. The total costs of the installation varied from 440 to 2250 \$/d, indicating a cost of 0.88–0.45 \$/m³ for this population range in terms of drinking water production.

The effect of possible fouling in a long-term RO operation was quantified by varying the effective membrane life. The cost per unit of water volume increased from 0.52 to $0.61 \text{ }/\text{m}^3$ (17% increase) when the membrane lifetime was reduced from 3 to 1.5 years.

The multiobjective optimization solutions, which consider the best compromises among the quality and cost objectives, indicated that the concentration of As (V) in the permeate water can be reduced to 0.5 ppb at a feasible cost.

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