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Determination of the Working Optimum Parameters for an Electrodialysis Reversal Pilot Plant

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ABSTRACT

Electrodialysis employs specially prepared membranes which are semipermeable to ions based on their charge, and it employs electrical current to reduce the ionic content of water in one of the most widely used desalting processes. In this paper we determine the parameters which characterize the working optimum of an electrodialysis reversal pilot plant, i.e., the values of the applied voltage (V) used in the electrical stages and the pressure of the feedwater (P). These values correspond to the maximum separation percentage ($\%S$) and are obtained by plotting $\%S$ against V and P.

Key Words. Ion-exchange membranes; Electrodialysis reversal; Optimization

INTRODUCTION

Electrodialysis (ED) is a membrane separation process based on the selective migration of aqueous ions through ion-exchange membranes as a result of an electrical driving force. This technique represents one of the most important methods for desalting solutions (1). It is used to produce drinking water

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from sea and brackish water and to concentrate seawater prior to evaporation (2–4), to remove salts and acids from pharmaceutical solutions and food products (5, 6), to recover water and valuable metal ions from industrial wastewater (3, 4), to produce ultrapure water, to concentrate dilute solutions, to separate electrolytes and nonelectrolytes, and to produce acids and alkalies from their salts (7–9). As regard the desalination of brackish water, ED is a well-proven technology with a multitude of systems operating worldwide (10).

In the ED process, only the dissolved solids move through the membranes and not the solvent, so that practical concentrations or depletions of electrolyte solutions is possible. The transport direction and the transport rate for each ion depend on its charge and mobility, solution conductivity, relative concentrations, applied voltage, etc., and the ion separation is closely related to the characteristics of the ion-exchange membranes, especially the permselectivity in the system being used (11).

The stack configuration and the ED process are outlined in Fig. 1. Two main streams flow in parallel through the membrane stack: one is progressively desalted and is referred to as the product stream, while the other main stream is the concentrate stream, a fraction of which can be recirculated to reduce the quantity of wastewater. In this way this stream increases in concentration and the addition of acid and conditioning chemicals may be required to prevent membrane stack scaling.

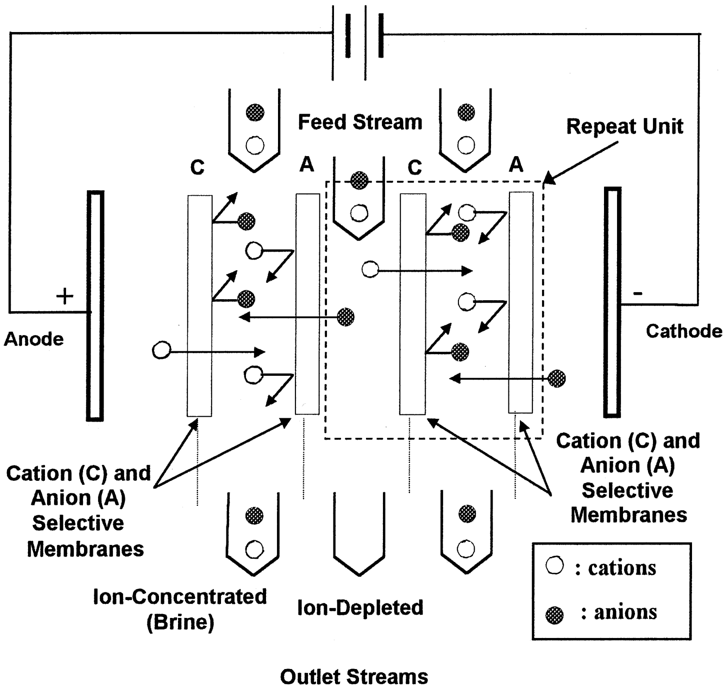

FIG. 1 Principle of electrodialysis. Scheme of an electrodialyzer with two cell pairs. Migration of ions is caused by the action of an electric field.

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The aim of this study was to investigate some aspects related to the working of an ED pilot plant by determining the most favorable experimental conditions for salt extraction by using voltage sweeps at different working pressures. To do this, a graphic relationship among separation percentage, applied voltage, and pressure of feed water was studied.

EXPERIMENTAL

Materials

The basic building block of an ED system is a membrane stack (12) in which parallel diluate and concentrate compartments, separated by anion and cation flat sheet membranes, are arranged alternately to form a setup similar to a conventional filter press. These membranes are highly selective for the respective ions and are composed of a strongly acidic (cation exchange) and strongly basic (anion exchange) resin containing a reinforced fabric. The membranes are semirigid, have a long life, and show the physical appearance of a plastic sheet. They are essentially impermeable to water under pressure, insoluble in aqueous solutions, and highly resistant to changes in pH (from 1 to 10), osmotic swelling, and fouling. The membranes also have a low electrical resistance.

Plastic separators are placed between the membranes to form the flow paths of the diluate and concentrate streams. These spacers are designed to minimize boundary layer effects and are arranged in the stack so that all the diluate and concentrate streams are manifolded separately. In this way a repeating section called a cell pair is formed; it consists of a cation-exchange membrane, a diluate flow spacer, an anion-exchange membrane, and a concentrate flow spacer. Up to several hundred of these repeat units are used in commercial systems.

Hydraulic and Electrical Staging

The manner in which the membrane stack array is arranged is called the staging. The aim of this staging is to provide a sufficient membrane area and sufficiently long retention time to remove a specified fraction of salt from the diluate stream.

In each hydraulic stage the feed solution entering the stack passes across the membrane surfaces between one pair of electrodes and exits. This represents one hydraulic stage. To increase the amount of salt removed, the exiting concentrate and diluate streams must again pass across another hydraulic stage. So by incorporating additional hydraulics stages in series, water of a specified purity can be obtained. When hydraulic stages are added within a single membrane stack, interstage membranes must be used. These membranes are thick cation membranes, twice as thick as the normal cation membrane in order to withstand a greater hydraulic pressure differential, and they form part of some cell pairs since they perform the same ion transfer function as the regular cation membranes. In this way a hydraulic stage is formed by placing the interstage membrane at an appropriate place in the membrane stack.

In a ED unit the ions are transferred through membranes from a less concentrated to a more concentrated solution as a result of the flow of direct electric current. The electric field generating this current arises from the application of a potential difference between two electrodes. The current is generated by ions migrating through the membranes: cations migrating to the cathode through the cation-exchange membranes and anions migrating to the anode through the anion-exchange membranes. In this way the feed solution is desalted since the salt is transported to the concentrate compartments.

Electrode compartments made up of an electrode, a flow spacer, and a thick cation-exchange membrane are placed at the end of the membrane stack. The thickness of the electrode spacer is two to six times greater than that of the membrane spacer, which permits a greater flow of water across the electrodes in order to reduce electrode scaling and fouling. The thick cation membrane is used to withstand the slight pressure difference ($\cong$ 2 psi) which is maintained between the electrode stream and the main flow stream in the membrane stack. Water from the electrode compartments is passed through a degasifier to vent off reaction gases.

Electrical staging is accomplished by inserting additional electrode pairs into the membrane stack. Each electrical stage allows the use of an independently controlled current to the cell pairs within the stage, providing maximum salt removal rates while avoiding polarization and hydraulic limitations.

Figure 2 outlines the staging in our ED pilot plant. There are two electrical stages, each with 50 cell pairs grouped into three hydraulic stages. The dashed lines show the two streams inside the stack.

Recent developments have improved the efficiency of ED by reversing the polarity of the electrodes periodically (3 or 4 times each hour). This is called electrodialysis reversal (EDR) (13). The polarity change reverses the direction of the movement of ions within the membrane stack, and the diluate stream becomes the concentrate stream and vice versa. For this, a device consisting of a set of electrovalves or motorized valves automatically commutes the inlets and outlets of the diluate and concentrate streams so that the incoming feedwater flows into the new product compartments and the concentrate stream flows into the new concentrate compartments. In this way the EDR system reduces the membrane scaling and fouling problems, and water is desalted without the constant need of adding chemicals during normal operation.

ED System and Experimental Details

The experiments were carried out using an Ionics Aquamite I EDR stack ($23 \times 25.5 \times 50$ cm^3) with two electrical stages, each with three hydraulic

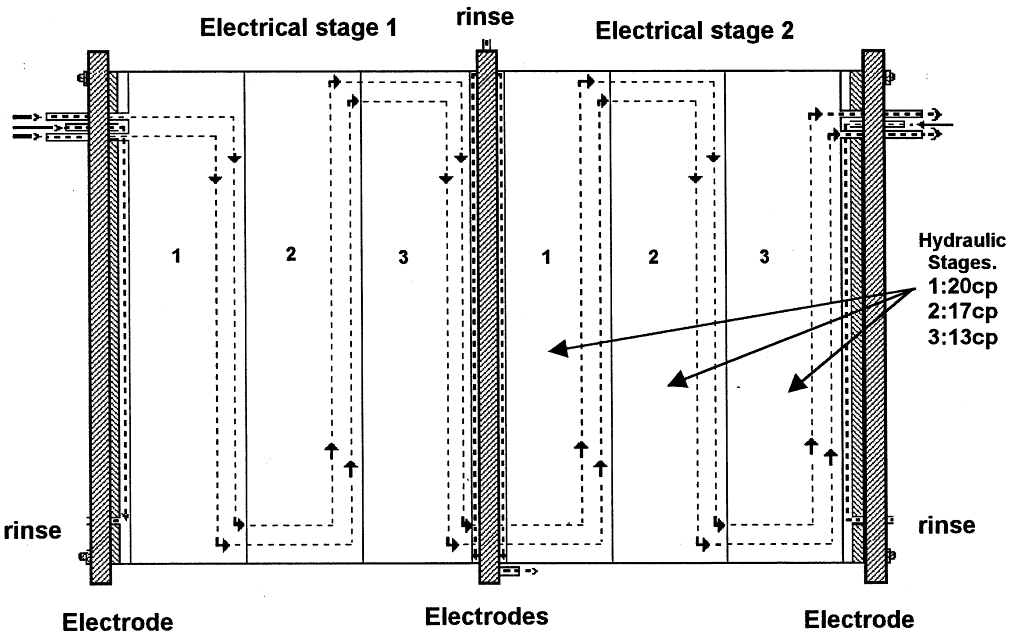

FIG. 2 Hydraulic and electrical staging in our ED pilot plant.

stages (Fig. 2), giving a total membrane area of 58.65 m^2 (cross-sectional membrane area of 23 × 25.5 cm^2; 50 cell pairs per 2 stages). Cation- and anion-exchange membranes were CATION-CR67-HMR-412 and ANION-204-SXZL-386 from Ionics, with a thickness of 0.5 mm; the cation membranes separating hydraulic stages were 1 mm thick. The fixed negative charges of the cation membranes are sulfonate groups which repel negative ions and allow positive ions to pass through; the charge of the anion membranes is positive because quaternary ammonium ions repel the positive ions and allow transport of the negative ions.

The separators are MKI Ionics tortuous path spacers, 1 mm thick, in which the solution flow path is a long narrow channel which makes several 180° bends between the entrance and exit ports of a compartment (Fig. 3). The left half of the spacer outlined in Fig. 3 shows the individual narrow solution channels and the cross-straps used to promote turbulence, while these straps are omitted in the right half of the figure so the flow path can be better depicted. Although at first sight the array of cross-straps seems to block the flow of water, they are only 1-ply thick and thus force water to flow in an over-and-under manner (Fig. 4). This type of flow causes turbulence in the stream and allows a higher electric current flow per unit area, therefore making more efficient use of the membrane area in the stack. Turbulence in the stream also promotes mixing, which aids the transfer of ions to the surface of the membranes.

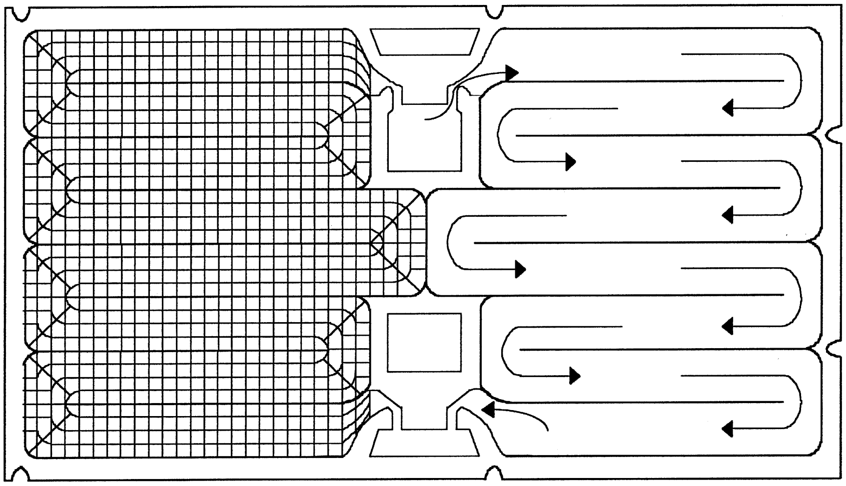

FIG. 3 Diagram of the tortuous-path spacer for an ED stack.

The stack has three stainless steel blocks, two at the ends and another in a central position, secured by means of four transverse bars with nuts which allow hermetic closing of the stack. Lateral filtration is thus avoided, and the streams only flow through the spacers.

The specifications of our EDR stack are given in Table 1. The flow rate of water in a stack corresponds to the mean value for concentrated and diluted streams when the plant operates without recirculation. These values were calculated by using the following conversion equation (13) for spacer MKI:

$$V = 1035 \times \text{gpm/cp}$$

where V = velocity in cm/s and gpm = demineralized inlet flow rate in US gallons per minute per cell pair (cp). In our case this flow was over the 20–80 L/h range and cp = 50.

The total electric current ranged from 2 to 9 mA/cm^2 at the first electrical stage and 2 to 5 mA/cm^2 at the second. These values are below the critical current–density where boundary layers effects become important (14).

ED is carried as outlined in Fig. 5, which permits the operation in a batch-type or continuous ED mode. In the first case the product stream and the con-

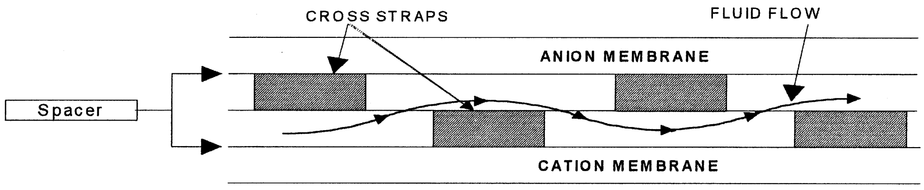

FIG. 4 Cross section of a tortuous-path spacer.

TABLE 1
Specifications of the EDR Stack

Type	Filter press	Ionics aquamite I
ED stack	Number	1
	Ion-exchange membranes	100 cell pairs/stack
	Size of membranes	$23 \times 25.5\ \mathrm{cm}^2$
	Spacer MK I	Thickness: 1 mm Flow path length: 348 cm
	Effective area of membrane	220 cm^2
	Electric stages	2
	Hydraulic stages	3/electric stage
Operating conditions	Flow rate of water in stack	2–7 cm/s
	Current density ($\mathrm{mA/cm}^2$)	Stage I: 2–9 Stage II: 2–5
	Pressure range	0.4–2.0 atm
	Voltage range (each stage)	30–110 V
Standard feed water	Salt	Sodium chloride
	Concentration	Low salinity: 3330 mg/L Mean salinity: 7190 mg/L
	Temperature:	Inlet: 20°C Outlet: 20–25°C

centrate stream are joined to be used again as feedwater and to prolong the characterization experiments with no need for a new supply of feedwater. In the second case the product stream exits while the concentrate stream is partially recycled to reduce waste by being injected into the feed concentrate stream with acid to avoid scaling in the stack or (if necessary) blended with the product water. The recycled stream may also be used to rinse the electrode compartments.

Given that all the electrodes are in the anode mode at some time, they were constructed of platinized titanium. They have an effective area of 550 cm^2 and are continually rinsed with a flow of 6 L/h.

The solution temperatures rose slightly during each experiment from room temperature (20°C) to about 25°C at maximum, mainly due to heat transfer from the centrifugal pumps.

The plant works in an automated manner, while the data acquisition of pressures, currents, and voltages is computerized. The pH of the concentrated stream was continually monitored to avoid scaling problems.

RESULTS AND DISCUSSION

Our EDR stack was characterized over a range of low salinity (<6000 mg/L) by desalting a standard aqueous solution prepared with degassed,

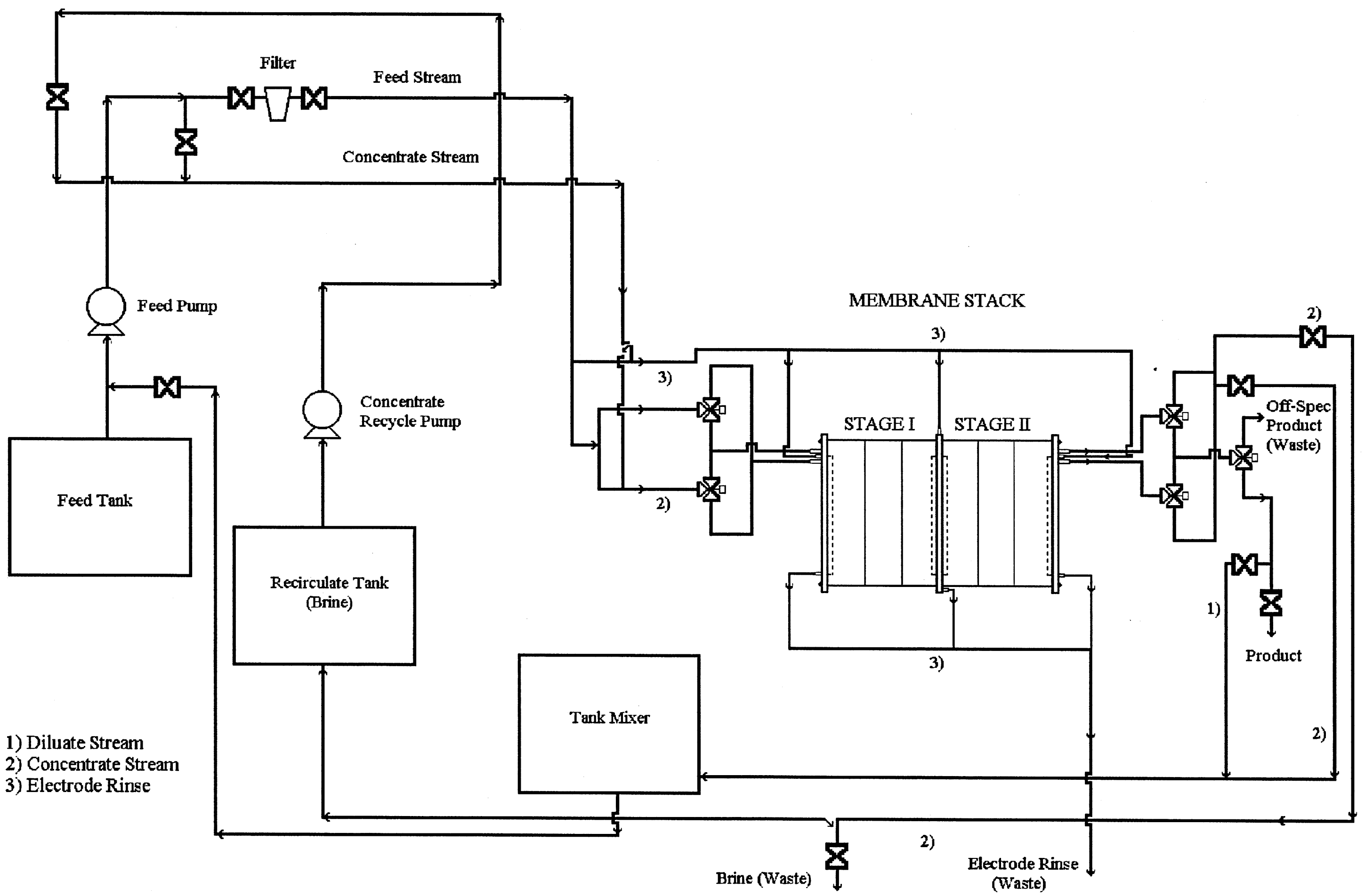

FIG. 5 Flow diagram for our EDR pilot plant.

deionized water and sodium chloride with a conductivity of 5200 μS/cm (≅ 3330 mg/L) and a pH of 5.7. The salt used was of analytical grade.

With the same fixed working voltage for both electrical stages, a pressure sweep was carried out over the 0.4 to 1.8 atm range. The voltage was varied between 30 and 110 V with increments of 10 V. These values were chosen to avoid exceeding the maximum values recommended by the manufacturer for the electric current (limiting current density) and flows. The current intensity was read at each stage for every value of the electric voltage and feed pressure. Also, the flow values in the feed, product, and waste streams were determined together with their respective conductivities, which are directly related with their respective saline concentrations. If the values of these concentrations are C_f (feed), C_p (product), and C_b (brine) and the corresponding flows are Q_f, Q_p, and Q_b, mass conservation requires that

$$C_f Q_f = C_p Q_p + C_b Q_b \tag{1}$$

as was verified. The separation percentage or extraction percentage ($\%S$) is defined as

$$\%S = 100\,\frac{C_f Q_f - C_p Q_p}{C_f Q_f} = 100\left[1 - \frac{C_p Q_p}{C_f Q_f}\right] \tag{2}$$

which represents the loss of saline mass in the dilute stream with respect to the feed stream.

In this way, for every value of the applied voltage, V, different values of the applied pressure were considered, together with the values of the current at each stage (I_1, I_2) and the concentrations and flows of each stream. The power consumed in each stage was also determined ($P_i = I_i V_i$, $i = 1, 2$), and the stack was characterized by reference to the mean power for both stages. The polarity change occurring every 15 minutes generates slight changes in the voltage and current strengths, and therefore in the consumed power, so the characterization also refers to the mean values of both polarities.

The influence of applied voltages and feedwater pressures on the separation percentages measured is illustrated in Fig. 6. At each pressure the separation percentage increased quickly with an increase of voltage, giving the surface shown, whose study reveals the existence of a relative maximum for $\%S$ at $V = 70$ V and $P = 1, 2$ atm (inlet flow: 100 L/h), which correspond to the working optimum.

The dependence of consumed power on applied voltages and feedwater pressures is shown in Fig. 7. The power increases with the pressure at each voltage, while the power increases at each pressure when the voltage ranges up to 90 V, showing an important change in the slope starting at about 70 V. For this voltage and at 1.2 atm (working optimum), the power was 84.79

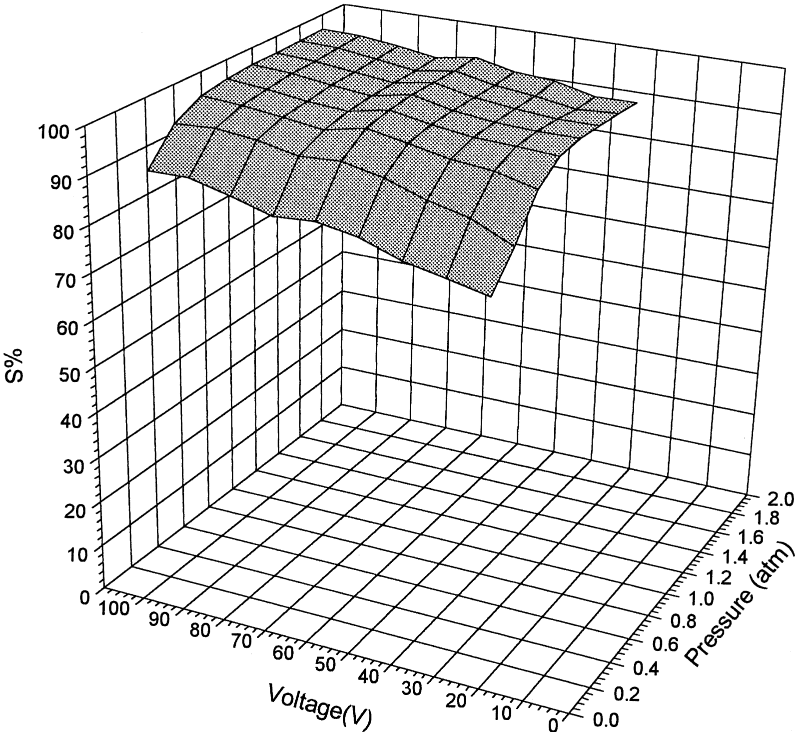

FIG. 6 Separation percentage (%S) versus applied electric voltage (V) and feedwater pressure (P, atm). The standard employed was an aqueous solution of sodium chloride with a conductivity of 5200 μS/cm.

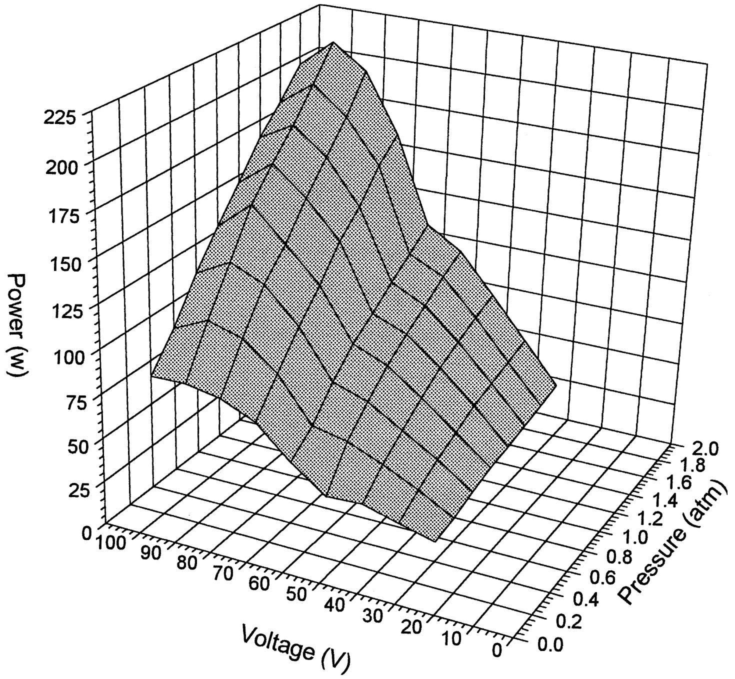

FIG. 7 Consumed power against electric voltage and feedwater pressure for low salinity.

W/m^3. Relating these values to a point on the surface in Fig. 7 gives the energy consumption at optimum operative conditions.

At the optimum voltage (70 V) and assuming an electricity cost of 0.09 \$/kWh, the values of the concentration of the diluted stream (C_p) are represented versus product flow (Q) and operating cost per m^3 from the surface given in Fig. 8. The working optimum corresponds to the minimum C_p, for which the cost per m^3 is \$0.17, although this does not include the cost of energy pumping. The cost of maintenance and investment must be added to obtain an estimate of the total cost.

In the same way as described above, a mean salinity range (6000 to 8000 mg/L) was characterized and optimized, and the surfaces shown in Figs. 9, 10, and 11 were obtained. An aqueous solution of sodium chloride was used as standard, but with a conductivity of 11,230 μS/cm (≅ 7190 mg/L). The values for V and P in the working optimum are 60 V and 1.2 atm, respectively, with a consumed power of 120 W/m^3 and a cost per m^3 of \$0.25. For salinities above 8000 mg/L, ED is not competitive compared with other desalting procedures, such as reverse osmosis, due to the electrical energy required (15).

After characterizing an EDR plant over a determined salinity range by obtaining the working optimum, brackish water with a total salt concentration in

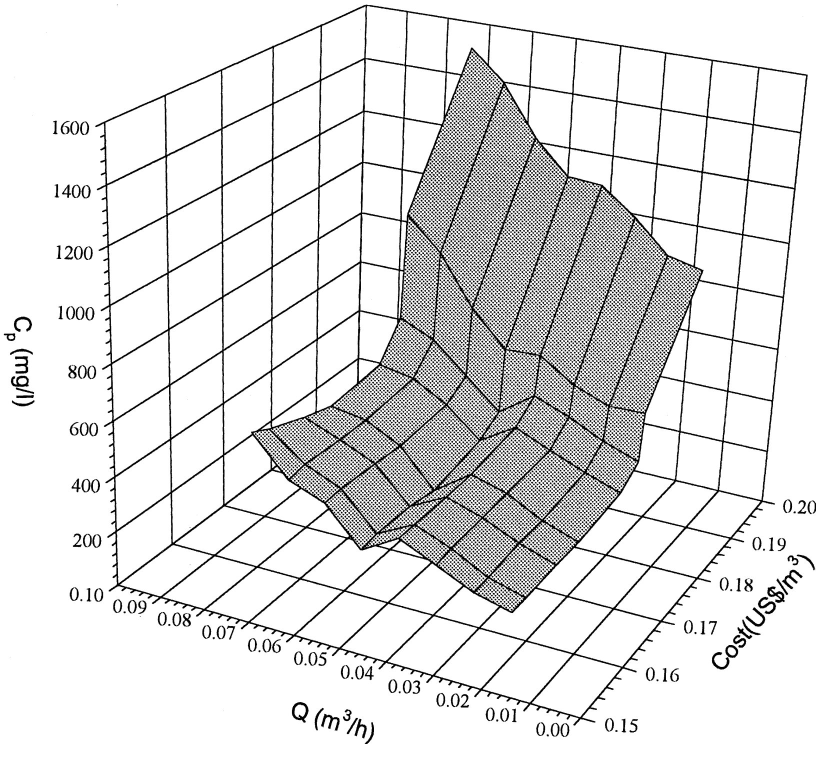

FIG. 8 Concentration in the diluted stream (C_p, mg/L) versus product flow (Q_p) and operating cost (US\$/m^3 product) at the optimum voltage (70 V) for low salinity.

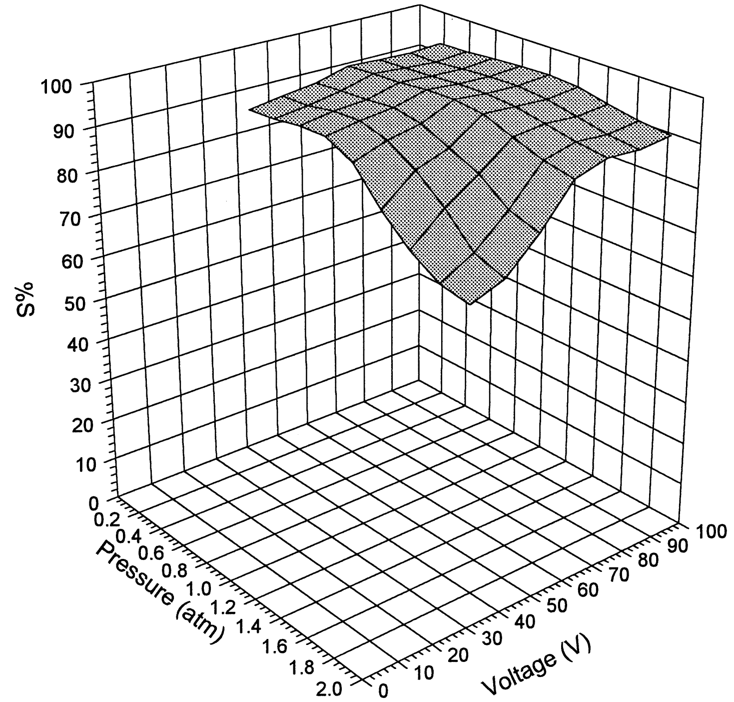

FIG. 9 Separation percentage (%S) versus applied electric voltage (V) and feedwater pressure (P, atm). The standard employed was an aqueous solution of sodium chloride with a conductivity of 11,230 μS/cm.

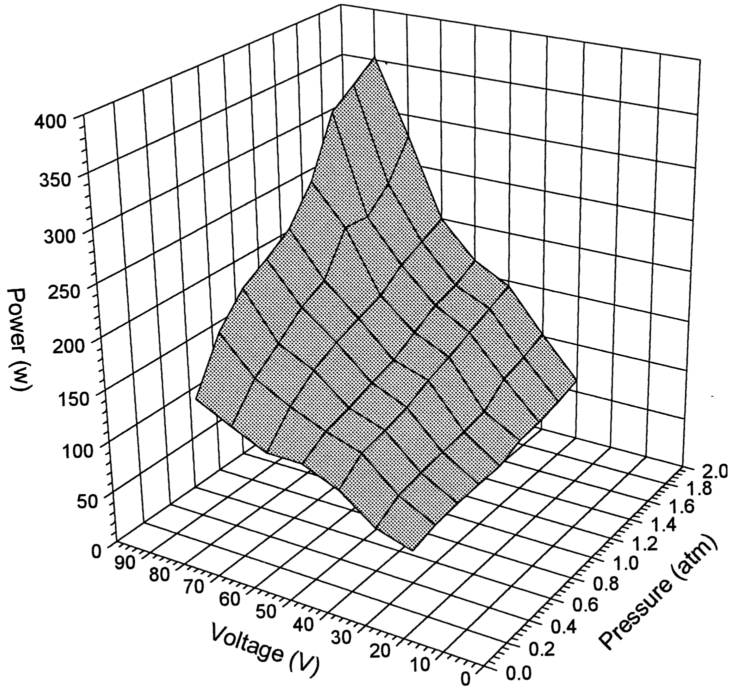

FIG. 10 Consumed power against electric voltage and feedwater pressure for mean salinity.

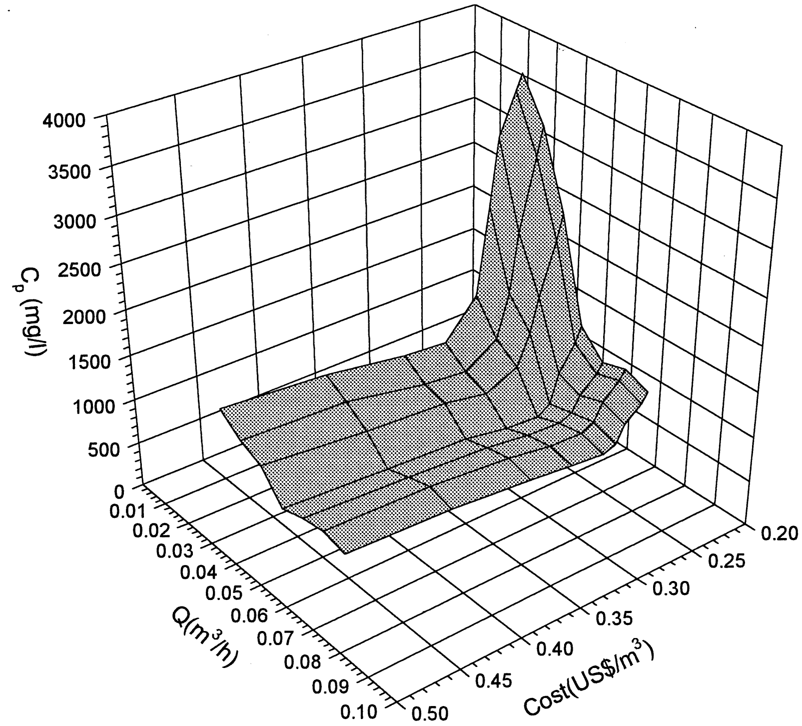

FIG. 11 Concentration in the diluted stream (C_p, mg/L) versus product flow (Q_p) and the operating cost (US\$/m^3 product) at the optimum voltage (60 V) for mean salinity.

TABLE 2
Ion Concentrations in Feedwater

	mg/L (ppm)	meq/L	% meq/L
Anions:			
Chloride (Cl^-)	1,485	41.9	48.3
Sulfate (SO_4^{2-})	1,814	37.8	43.5
Bicarbonate (HCO_3^-)	394	6.5	7.4
Nitrate (NO_3^-)	39	0.6	0.7
Cations:			
Sodium (Na^+)	635	27.6	30.2
Calcium (Ca^{2+})	816	40.7	44.6
Magnesium (Mg^{2+})	248	20.4	22.3
Potassium (K^+)	101	2.6	2.8
Other Parameters			
Electrical conductivity (at 20°C)	11,430 μS/cm		
Total dissolved solids (TDS)	5,532 mg/L		

TABLE 3
Ion Concentrations in Product Water

	mg/L (ppm)	meq/L	% meq/L
Anions:			
Chloride (Cl^-)	288	8.1	54.5
Sulfate (SO_4^{2-})	168	3.5	23.5
Bicarbonate (HCO_3^-)	189	3.1	20.8
Nitrate (NO_3^-)	11	0.2	1.2
Cations:			
Sodium (Na^+)	149	6.5	41.4
Calcium (Ca^{2+})	133	6.6	42.4
Magnesium (Mg^{2+})	18	1.5	9.5
Potassium (K^+)	41	1.0	6.7
Other Parameters			
Electrical conductivity (at 20°C)	2030 μS/cm		
Total dissolved solids (TDS)	997 mg/L		

TABLE 4
Ion Concentrations in Brine

	mg/L (ppm)	meq/L	% meq/L
Anions:			
Chloride (Cl^-)	3,332	94.0	51.0
Sulfate (SO_4^{2-})	3,911	81.4	44.2
Bicarbonate (HCO_3^-)	392	6.4	3.5
Nitrate (NO_3^-)	152	2.5	1.3
Cations:			
Sodium (Na^+)	1,197	52.1	29.6
Calcium (Ca^{2+})	1,445	72.1	41.0
Magnesium (Mg^{2+})	569	46.8	26.6
Potassium (K^+)	188	4.8	2.7
Other Parameters			
Electrical conductivity (at 20°C)	18,390 μS/cm		
Total dissolved solids (TDS)	11,186 mg/L		

TABLE 5
Dilution of Feedwater by Ion-Exchange Electrodialysis Reversal (EDR)

Stream	TDS (mg/L)	Flow (L/h)	TDS × flow (mg/h)
Feed	5,532	90	497,880
Product	997	50	49,850
Brine	11,186	40	447,440

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TABLE 6
Separation Percentage of Ions in Feedwater

	(%*S*)
Anions:	
Cl^-	80.6
SO_4^{2-}	90.7
HCO_3^-	52.0
NO_3^-	71.8
Cations:	
Na^+	76.5
Ca^{2+}	83.7
Mg^{2+}	92.7
K^+	59.4

the considered range can be treated in the most suitable operating conditions. In our case the feedwater had a saline content of 5532 mg/L. The most favorable degree of desalination was obtained without recirculation with a separation percentage of 82%, leaving a salt concentration of 997 mg/L and a recovery of 55.56%. This recovery can be increased up to 80% with a recirculation rate of 65% over the concentrated stream, although this results in a certain loss of quality in the product water.

Tables 2, 3 and 4 show the concentrations of different ions corresponding to the feedwater, product water, and concentrated stream, respectively, as determined by capillary electrophoresis ion analysis. Table 5 illustrates the results for the demineralization of feedwater by EDR. Table 6 gives the separation percentages corresponding to different ions in feedwater.

CONCLUSIONS

1. The characterization of an EDR plant by means of the determination of the working optimum allows us to operate in the most suitable conditions with maximum efficiency.
2. This characterization can be effected by surfaces corresponding to separation percentage and consumed power versus applied voltage and pressure, whose study reveals the existence of a maximum for the separation percentage and minimum energy consumption, which also correspond to the minimum cost.
3. In the case of our pilot plant Aquamite I, the characterizations were accomplished by the use of standard aqueous solutions of NaCl. For a low salinity range (<6000 ppm) and the mean salinity range (6000–8000

ppm), the optimum voltages were at 60 and 70 V, respectively, with a feedwater pressure of 1.2 atm in both cases, and a cost of \$0.17 and \$0.25/m^3 product water.

4. A problem of low salinity brackish water was treated by EDR under optimum conditions and gave a separation percentage of 82% without recirculation.

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