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Fractional factorial design of water desalination by neutralization dialysis process: concentration, flow rate, and volume effects

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ABSTRACT

This work concerns the optimization of the neutralization dialysis process applied to water desalination. This process is used for salt removal from a NaCl model solution (initially 0.08 M) in a three-compartment membrane stack (acid, salt, and alkali compartments) with a cation-exchange membrane and an anion-exchange one, i.e. Neosepta® CMX and AMX, respectively. The desalination process has been investigated as a function of three main parameters, namely the initial concentration and the flow rate of acidic and alkaline solutions as well as the saline solution volume. A fractional factorial design, i.e. 2^{5-1}, has been employed to assess the parameters affecting the desalination efficiency, and specifically to study the ion-exchange of H^+ and Na^+ through CMX and that of OH^- and Cl^- through AMX. A linear theoretical model based on the experimental results has been carried out to account for the influence of the different parameters investigated and their relative interactions. The analysis of variance and student's *T*-tests have shown that the initial alkaline solution concentration and the saline solution volume are the most statistically significant parameters affecting the ion transport of H^+/Na^+ and OH^-/Cl^- through CMX and AMX, respectively, as well as the desalination duration.

Keywords: Neutralization dialysis; Desalination; Ion-exchange membranes; Fractional factorial design

1. Introduction

Worldwide water distribution is represented approximately by 97.4% of salty water, 1.7% of icecaps and glaciers, and only 0.8% of available fresh water [1]. In the last decade, many countries suffer from lack of natural fresh water. In fact, 2.3 billion people (which correspond to 41% of the world's population) live with a lack of water [2]. This phenomenon is more

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pronounced in North African countries and certain countries of the Mediterranean Basin. Indeed, the drinking water needs overtakes very widely the natural resources reserve. In Tunisia, for instance, the water resources represent only 460 m^3/year/inhabitant [3], which is under the shortage line of 500 m^3/year/inhabitant. By the end of 2025, the water resources (surface water and ground water) will reach quantitative saturation, while the qualitative deficit has already been real for a long time [4]. This water rarefaction can be explained by two important phenomena. The first one is global warming which causes 60% ground water diminution, 20% surface water evaporation, and 20% ground drought [5]. The second phenomenon is pollution which induces water contamination, due to industrial wastes [6], agriculture pollution [7], and domestic waste [8]. Accordingly, finding energy- and cost-saving process to produce drinking water has become a major challenge nowadays to satisfy the considerable water needs. The main drinking water resources are ground water and surface water. Surface water is the generic name for sea water and brackish water. The latter has a lower salinity than sea water (about 1–10 g L^{-1} of salt), it is an accessible and renewable resource but its salinity increases by overexploitation [9,10]. Thus, surface water desalination can be a viable solution to produce drinking water. To perform the water desalination, different process are used such as distillation [11], nanofiltration [12,13], electrodialysis (ED) [6,14], reverse osmosis [10,15,16], and neutralization dialysis (ND) [17,18]. The main desalination objective is currently to obtain a drinking water with low energy consumption and low operating cost. ND is a membrane process developed first by Igawa et al. in 1987 as a new water desalination method [18]. It is based on two Donnan dialysis operations [19–23] that use simultaneously anion- (AEM) and cation- (CEM) exchange membranes. ND can be a promising process in developing countries for water desalination. Indeed, ND is a low investment, low operational, and maintenance cost process [24]. It is also convenient to be used in isolated locations with a low technical requirement and staff qualification.

The ND process shows interesting ion transport phenomena, which makes membranes very promising separators in deionization [25], separation of weak acids and bases [26], as well as transport of glycine [27]. Desalination of aqueous solutions of carbohydrates and milk whey is developed as an industrial application [28]. In the process investigated here, the ND cell is composed of three compartments, i.e. A compartment contains HCl solution, B compartment contains NaOH solution, and C compartment contains a NaCl solution model. Moreover, the saline solution is placed between acidic and alkaline solutions separated, respectively, by a CEM and an AEM. The CEM (AEM) allows the substitution of cations (anions) of the treated solution by H^+ (OH^-) ions, leading to a decrease in the mineral charge of the treated solution.

A preliminary study has shown that main parameters affecting the ND desalination process are the initial acidic and alkaline solution concentrations and the flow rate [29]. Moreover, another study was carried out by ND process where the desalination kinetics of a model NaCl solution was treated, and the process feasibility was evaluated [24]. The ND cost was estimated to 1.38\$/m^3 for 0.02 M NaCl solution. The water desalination cost by ND process is relatively high compared to the ED (0.17–0.40\$/m^3) [30] and RO (0.10–1.00\$/m^3) [31] but lower compared to other using photovoltaic energy (4.60–6.80\$/m^3) [32]. However, the main advantage of de ND lies in the low technical requirement and low staff qualification for the material upkeep. Thus, this technique can be easily implemented in isolated areas. This is a great advantage for applications in the developed countries.

In order to get further insight into the ND process and to optimize the desalination duration, the initial concentration and flow rate of acidic and alkaline solutions as well as the saline solution volume are varied. In the present paper, 16 experiments have been carried out using a 2^{5-1} fractional factorial design to investigate the effect of the main parameter as well as the interaction between them. This study will allow us to find the optimal conditions to optimize the ND desalination process, particular by adjusting every parameter affecting desalination efficiency. These conditions will be applied in a future investigation of the use of this process for real surface water desalination.

2. Experimental

2.1. Membrane treatment

Two homogeneous membranes (Neosepta® CMX and AMX, ASTOM, Japan), known for their excellent permselectivity, were used for the ND process. These membranes were provided by Eurodia Industries S.A.

The cation-exchange membrane (CMX) and the anion-exchange membrane (AMX) were manufactured from the same polystyrene-DVB crosslinked polymer and contained PVC as a binder [33]. The only difference is that CMX contains sulfonate groups, whereas AMX contains quaternary ammonium groups.

Prior to any membrane process, CMX and AMX were treated following the French standard NF X 45-200 [34]. The main treatment objective is to remove manufacturing process impurities (organic molecules) and to allow for the stabilization of the physicochemical properties. Thus, the reproducible initial conditions were achieved.

AMX was immersed in 0.1 M HNO_3 for 1 h, then rinsed with distilled water and carefully dried with filter paper, then immersed in 0.1 M HCl for 1 h and rinsed by immersing in 0.1 M NaCl. However, CMX was pre-equilibrated with 0.1 M HCl for 1 h, then washed with distilled water, then immersed in 0.1 M NaOH for 1 h. The procedure was repeated twice for the two membranes. After these cycles, AMX and CMX were stored in 0.1 M HCl and 0.1 M NaCl, respectively.

2.2. Membrane characterization

For each membrane, the active surface area was equal to 64 cm^2. The main physicochemical characteristics of the cationic and anionic membranes are shown in Table 1.

The ion-exchange capacity was determined following the French standard NF X 45-200 [34].

The membrane samples, previously immersed in an appropriate stabilization solution, were placed between two foils of filter paper and pressed slightly in order to remove the excess of water. Then, the membranes were immediately placed in a HB43-S Mettler-Toledo moisture thermobalance in which they were heated to 140°C until their mass did not vary (at 0.001 g) for ten minutes. The water content (W) was determined by the mass differences between wet (W_a) and dried (W_b) samples as follows:

$$W\ (\text{wt}\%) = \frac{W_a - W_b}{W_a} \times 100 \tag{1}$$

The membrane thickness (T_m) was measured using a Käfer Thickness Dial Gauge. The membrane thickness value was averaged from measurements at 10 different locations on the effective surface region of the membrane.

Table 1
Main physicochemical properties of CMX and AMX membranes

Membranes	CMX	AMX
Ion-exchange capacity (meq g^{-1})	1.6	1.4
Water content (wt%)	25	26
Thickness (μm)	170	140
Electrical conductivity (mS cm^{-1})		
NaCl	2.1	1.2
HCl	16.0	–
NaOH	–	4.7

The electrical conductivity measurements were carried out using the differential method [35,36]. The experimental assembly used consisted a clip-type cell (10 kHz AC frequency) to measure the membrane conductivity, a conductivity meter CDM92 (RADIOMETER-TACUSSEL) and a water bath at 25 ± 0.2°C. The determination of membrane conductance G_m needed two measurements, one without the membrane (G_1) and another one with the membrane (G_2), to deduce the value:

$$G_m = \frac{G_1 G_2}{G_1 - G_2} \tag{2}$$

Using the value of the membrane thickness (T_m) value and the electrodes section area ($A = 1\ cm^2$), the membrane conductivity was calculated as:

$$k_m = G_m \times \frac{T_m}{A} \tag{3}$$

The electrical conductivity of CMX and AMX were measured in the HCl and NaOH solutions respectively as well as in the NaCl solution for both membranes at 0.05 mol L^{-1}.

2.3. ND process

In the ND process, the saline water is placed between an acidic and an alkaline solution separated, respectively, by CMX and AMX. The CMX enables the Na^+ ions substitution by H^+. In contrast, the AMX enables the Cl^- ions substitution by OH^- ions. Thus, the protons (H^+) and the hydroxide ions (OH^-) in the saline compartment are combined into water and the mineral charge decreases. Thus, the saline solution is treated under ideal conditions.

ND operations were performed using a PCCell ED 64-004 electrodialysis stack with three compartments to remove the salt from a model NaCl solution which corresponds to a surface water salinity of 5 g L^{-1} (i.e. 0.08 mol L^{-1}). The initial salt concentration used in this study is higher than that used in previous study. This higher salt concentration was chosen in order to improve the ND process. This choice was done because the salinity of the major part of the North Africa's brackish water does not exceed 5 g L^{-1} [37].

After each dialysis operations, the electrodialysis stack was washed with distilled water to remove any impurities in the spacers which come back from previous operations. For all experiments, the acid

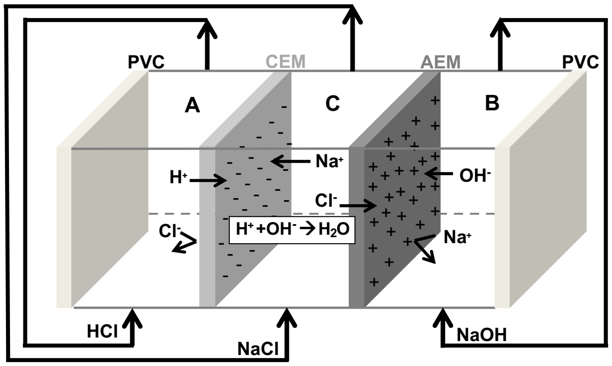

Fig. 1. Schematic representation of ND with a three-compartment membrane stack. CEM: cation-exchange membrane; AEM: anion-exchange membrane; A: acid solution; C: saline solution; B: base solution.

compartment (noted A) contains HCl solution, the base compartment (noted B) contains NaOH solution, and the saline compartment (noted C) contains NaCl solution. Fig. 1 shows the device used to study the desalination time by ND in a three-compartment membrane stack, the CMX membrane separates A and C compartments, while the AMX membrane separates B and C compartments.

The width of each compartment is 450 µm, corresponding to Nylon® spacer thickness, and the active area for each IEM is 64 cm^2. The fluid circulation through the three compartments is ensured by Masterflex® pumps. It is about a closed circuit. The temperature of A, B, and C solutions are maintained constant to 25.0℃ ± 0.1℃. Table 2 show the initial conditions of ND operations for each compartment.

During the desalination, pH and ionic conductivity were measured every two minutes in the saline compartment using a combined pH and conductivity meter (Jenway 3540). Its initial characteristics are as follows: a pH value of 5.5–6.0, and an ion conductivity of 8.48 mS cm^{-1} (corresponds to 5 g L^{-1}). The water desalination is achieved when the solution contain less than 0.5 g L^{-1} (which corresponds to 1 mS cm^{-1}) of salts or total dissolved solids [38–40].

2.4. Fractional factorial design

In order to obtain the optimal conditions for water salts removal by ND process, a factorial design was carried out. It is a multi-factor cross-group design which can test the differences between various levels of each factor and also test the interactions between the factors. If there is an interaction between two or more factors indicating that these factors are interdependent, when the level of a factor changes, the effect of one or several factors change accordingly. In contrast, if there is no interaction indicating the independence of various factors, changes in the level of a factor do not affect the effects of other factors.

To reduce the experiment number, a fractional factorial design at n^{k-q} is used. This type of factorial design permits the study of all the factors with two or four or 2^q minus times than a full factorial design. The system is composed of: n = number of levels, k = number of factors, and q = generators (here $n = 2$, $k = 5$, and $q = 1$). Thus, the total experiment number needed for the investigation is 2^{5-1} which corresponds to 16 experiments for 5 factors. The effect of factor i can be estimated from b_i (Eq. (4)):

$$b_i = \frac{\sum y_{t_i}^{+} - \sum y_{t_i}^{-}}{N} \tag{4}$$

where $\sum y_{t_i}^{+}$ is the sum of the responses for which factor i is on the high level $(+X_i = +1)$, $\sum y_{t_i}^{-}$ is the sum of the responses for which factor i is on low level $(-X_i = -1)$, and N is the number of experiments. The theoretical response at $X_i = 0$ can be estimated from b_0 (Eq. (5))

$$b_0 = \frac{\sum y_{t_i}^{+}}{N} \tag{5}$$

To increase the response, the factor i must be maintained [41]:

(1) at the high level (+) if b_i is positive.
(2) at the low level (−) if b_i is negative.

Table 2
Initial conditions of ND operations

Parameters	A compartment (HCl)	C compartment (NaCl)	B compartment (NaOH)
Initial concentration (mol L^{-1})	0.10–0.50	0.08	0.10–0.50
Flow rate (mL min^{-1})	50–100	75	50–100
Volume (L)	1.5	0.5–1.0	1.5

Table 3
Experimental ranges and level of factors studied in the factorial design

Factors	Coded symbol	Range and level	
		−1	+1
Initial acidic solution concentration ($[A]_0$, mol L^{-1})	A	0.1	0.5
Initial alkaline solution concentration ($[B]_0$, mol L^{-1})	B	0.1	0.5
Acidic flow rate (Q_A, mL min^{-1})	C	50	100
Alkaline flow rate (Q_B, mL min^{-1})	D	50	100
Saline solution volume (V_C, L)	E	0.5	1.0

Table 4
Studied parameters in their reduced and normal forms

Experiment	$[A]_0$	A	$[B]_0$	B	Q_A	C	Q_B	D	V_C	E	Y (min)
1	−1	0.1	−1	0.1	−1	50	−1	50	+1	1.0	1,246
2	+1	0.5	−1	0.1	−1	50	−1	50	−1	0.5	512
3	−1	0.1	+1	0.5	−1	50	−1	50	−1	0.5	508
4	+1	0.5	+1	0.5	−1	50	−1	50	+1	1.0	494
5	−1	0.1	−1	0.1	+1	100	−1	50	−1	0.5	368
6	+1	0.5	−1	0.1	+1	100	−1	50	+1	1.0	816
7	−1	0.1	+1	0.5	+1	100	−1	50	+1	1.0	410
8	+1	0.5	+1	0.5	+1	100	−1	50	−1	0.5	140
9	−1	0.1	−1	0.1	−1	50	+1	100	−1	0.5	446
10	+1	0.5	−1	0.1	−1	50	+1	100	+1	1.0	1,125
11	−1	0.1	+1	0.5	−1	50	+1	100	+1	1.0	363
12	+1	0.5	+1	0.5	−1	50	+1	100	−1	0.5	154
13	−1	0.1	−1	0.1	+1	100	+1	100	+1	1.0	1,065
14	+1	0.5	−1	0.1	+1	100	+1	100	−1	0.5	432
15	−1	0.1	+1	0.5	+1	100	+1	100	−1	0.5	581
16	+1	0.5	+1	0.5	+1	100	+1	100	+1	1.0	355

Table 3 shows the five factors (initial concentration and flow rate of acidic and alkaline solutions, saline solution volume) used in the fractional factorial design, as well as the low and high levels studied.

The 16 experiments that were carried out are presented in Table 4, as a screening approach to find the significant factors affecting the responses. If Y is the response variable of desalination duration when ionic conductivity achieved 1 mS cm^{-1}, then the regression equation with five parameters and their interactions is then given by Akhnazarova and Katarov Eq. (6) [42]:

$$Y = b_0 + b_1A + b_2B + b_3C + b_4D + b_5E + b_{12}AB + b_{13}AC + b_{14}AD + b_{15}AE + b_{23}BC + b_{24}BD + b_{25}BE + b_{34}CD + b_{35}CE + b_{45}DE \tag{6}$$

where b_0, b_1, b_2, b_3, b_4, and b_5 are the linear coefficients; b_{12}, b_{13}, b_{14}, b_{15}, b_{23}, b_{24}, b_{25}, b_{34}, b_{35}, and b_{45} are the second-order interaction terms; A, B, C, D, and E are the dimensionless coded factors of the following parameters studied: acidic and alkaline concentrations and flow rates as well as saline solution volume, respectively.

3. Results and discussion

3.1. Model equation for 2^{5-1} design

According to the fractional factorial design for desalination duration, effects, regression coefficients, standard errors, T and p-values (standardized effects) are calculated in Table 5.

By substituting the coefficients in Eq. (6) with their values from Table 5, a model equation relating the level of parameters to the desalination duration efficiency can be derived (Eq. (7)).

$$Y_t = 549.31 - 54.13A - 192.38B - 40.81C + 6.06D + 176.19E - 25.81AB - 22.25AC + 9.62AD + 315.88AE + 36.63BC - 20.37BD - 150.25BE + 82.69CD - 20.31CE - 11.06DE \tag{7}$$

Table 5
Statistical parameters and analysis of variance for a 2^{5-1} design

	Effect	Coefficient	Standard error	Df	SS	MS	T-value	p-value	F-value
Constant	–	549.31	6.27	–	–	–	87.68	0.000	
$[A]_0$	−108.25	−54.13	6.27	1	93,744	93,744	−8.64	0.000	74.63
$[B]_0$	−384.75	−192.38	6.27	1	1,184,260	1,184,260	30.70	0.000	942.79
Q_A	−81.63	−40.81	6.27	1	53,301	53,301	−6.51	0.000	42.43
Q_B	12.12	6.06	6.27	1	1,176	1,176	0.97	0.348	0.94
V_C	352.37	176.19	6.27	1	993,345	993,345	28.12	0.000	790.80
Main effects	–	–	–	**5**	**2,325,827**	**465,165**	–	–	**370.32**
$[A]_0 \times [B]_0$	−51.63	−25.81	6.27	1	21,321	21,321	−4.12	0.001	16.97
$[A]_0 \times Q_A$	−44.50	−22.25	6.27	1	15,842	15,842	−3.55	0.003	12.61
$[A]_0 \times Q_B$	19.25	9.62	6.27	1	2,964	2,964	1.54	0.144	2.36
$[A]_0 \times V_C$	31.75	15.88	6.27	1	8,065	8,065	2.53	0.022	6.42
$[B]_0 \times Q_A$	73.25	36.63	6.27	1	42,925	42,925	5.85	0.000	34.17
$[B]_0 \times Q_B$	−40.75	−20.37	6.27	1	13,284	13,284	−3.25	0.005	10.58
$[B]_0 \times V_C$	−300.50	−150.25	6.27	1	722,402	722,402	23.98	0.000	575.10
$Q_A \times Q_B$	165.38	82.69	6.27	1	218,791	218,791	13.20	0.000	174.18
$Q_A \times V_C$	−40.63	−20.31	6.27	1	13,203	13,203	−3.24	0.005	10.51
$Q_B \times V_C$	−22.13	−11.06	6.27	1	3,916	3,916	−1.77	0.097	3.12
Interaction	–	–	–	**10**	**1,062,713**	**106,271**	**84.60**	**0.000**	–
Residual	–	–	–	**16**	**20,098**	**1,256**	–	–	–
Total	–	–	–	**31**	**3,388,541**	–	–	–	–

3.2. Student's T-test

Student's T-test was carried out to determine whether the calculated main parameters and interaction effects were significantly different from zero. The main parameters represent the five different factors used in the fractional factorial design ($[A]_0$, $[B]_0$, Q_A, Q_B, and V_C) as well as the interaction effects of the combination that can exist between the different factors (10 interactions). Absolute values of main parameters and interaction factors are illustrated in a Pareto chart in the horizontal columns (Fig. 2). To indicate the minimum level, a vertical line is drawn in the Pareto chart. This value which corresponded to t-value was equal to 2.12 with a 95% confidence level and 16 degrees of freedom. In fact, the bars of B and E (that correspond to $[B]_0$ and V_C, respectively) which remained outside the reference line in the Pareto chart, showing that these terms are the most parameters affecting the desalination efficiency. However, DE, AD, and D (that correspond to $Q_B \times V_C$, $[A]_0 \times Q_B$, and Q_B, respectively) remained inside the reference line contributed the least to the prediction of salts removal efficiency. It is noticed that alkaline concentration and saline solution volume were the parameters most affecting the desalination efficiency.

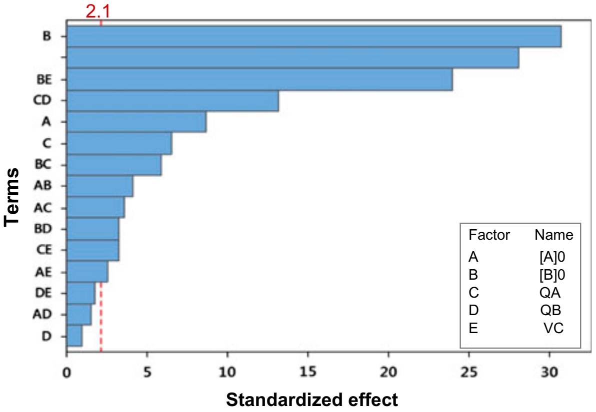

Fig. 2. Pareto chart for standardized effects.

3.3. Analysis of variance (ANOVA)

The significant main parameters and interaction effects of factors influencing the salts removal efficiency were determined by analysis of variance (ANOVA). Table 5 represents the sum of squares (SS) and mean square (MS) of each factor. The p- and F-values are defined as the ratio of the respective MS effect and the MS error. The importance of a parameter is evaluated by its F- and p-values. For 95% confidence level, 1 degree of freedom and 16 factorial tests, the percentage points of F Distribution ($F_{0.05,1,16}$) is equal to 4.49. Indeed, all the effects with F-values higher than 4.49 are significant. Concerning the main effects, only alkaline solution flow rate (Q_B) is not significant (i.e. F-value of Q_B is equal to 0.94). In the case of interaction effects, the interaction between $[A]_0$ and Q_B as well as Q_B and V_C are the less significant with F-values that correspond to 2.36 and 3.12, respectively. The p-value is the probability value used to determine the statistically significant effects in the desalination duration model. Values closer to zero imply higher significance. Thus, the p-values confirm the results obtained for F-values. Moreover, the results obtained from F- and p-values predict that initial alkaline solution concentration $[B]_0$ and saline solution volume are the factors most statistically significant with fast F-values of 942.79 and 790.80, respectively. Besides, the interactions between $[B]_0 \times V_C$ and $Q_A \times Q_B$ affect the desalination duration with F-values according to the data in Table 5. These results were positively associated with Student's T-test.

3.4. Main and interaction effects

3.4.1. Main effects for ND desalination

Fig. 3 represents the graph of main effects analyzing the coefficients of Eq. (7). Indeed, when the slope is important, the response is more significant. The positive slope is associated with a decrease in the ion-exchange kinetics which induces high desalination duration. However, the removal salt efficiency is high for a negative slope. It can be inferred that initial alkaline solution concentration and the saline solution volume are the most important parameters affecting desalination efficiency. These results are confirmed by the higher coefficients of $[B]_0$ and V_C in Eq. (7) which correspond to 192.38 and 176.19, respectively.

3.4.1.1. Acidic and alkaline solutions concentration effects. Concerning the concentration effects of acidic and alkaline solutions, it is noticed that initial alkaline solution concentration has a higher effect on the desalination duration than that of initial acidic solution concentration. This is due to the higher AMX electrical conductivity in the NaOH electrolyte (B compartment) compared to that of CMX in HCl electrolyte (A compartment) (Table 1). In addition, the Na^+ and H^+ transfer rate through CMX is not similar to than Cl^- and OH^- ions through AMX. In fact, at the beginning of desalination, the exchange flux through CMX (noted J^C) is more important than that the exchange flux through AMX (noted J^A) due to the faster decrease of Na^+ compared with Cl^- in the saline compartment and the higher H^+ transfer rate through CMX. Thus, the increase in the initial alkaline solution concentration leads to an increase in the J^A flux across AMX which induces a quasi-similar flux evolution and the desalination duration becomes even shorter.

3.4.1.2. Acidic and alkaline flow rate effects. The acidic and alkaline flow rates have moderately significant effects (Table 5) on the desalination efficiency, and especially the alkaline flow rate where the straight line is almost horizontal (its slope is almost zero) (Fig. 3, Q_B). The CMX is thicker than that the AMX, which induces a diffusion boundary layer (DBL) larger in the CMX side. Thus, the increase in acidic flow rate in the A compartment leads to thinner DBLs [43,44] through CMX which induces an acceleration of the desalination process. Indeed, the turbulences are created by spacers at the membrane–solution interface. These turbulences lead to thinner DBLs and more important exchange kinetics.

3.4.1.3. Saline solution volume effect. The saline solution volume has a very significant effect on the desalination duration. The volume decrease induces a low quantity of Na^+ and Cl^- in the saline compartment

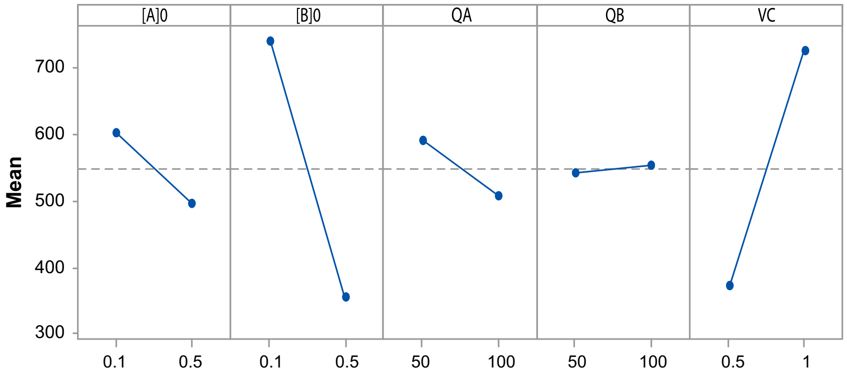

Fig. 3. Main effects plot for desalination by ND.

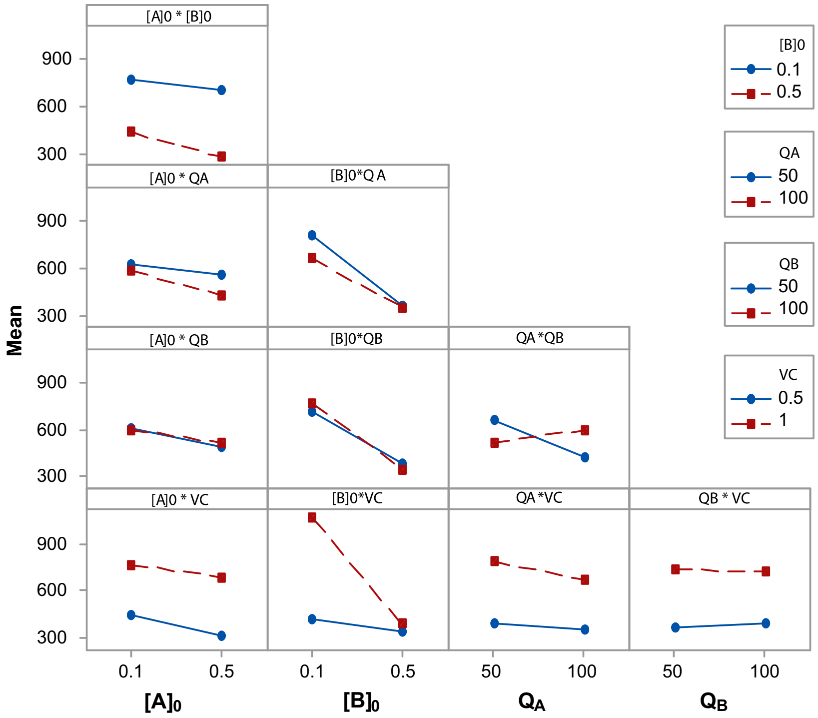

Fig. 4. Interaction effects plot for desalination by ND.

which accelerates the exchange flux through CMX and AMX, respectively. Hence, desalination duration becomes shorter.

3.4.2. Interaction effects for ND desalination

The interaction effects plots are shown in Fig. 4. The non-parallel lines in this figure indicate an interaction between the two factors. The graphs in Fig. 4 and coefficients of Eq. (4) show 6 negative interactions: initial acidic and alkaline solutions concentration ($[A]_0 \times [B]_0$), initial acidic solution concentration and alkaline flow rate ($[A]_0 \times Q_B$), initial alkaline solution concentration and alkaline flow rate ($[B]_0 \times Q_B$), initial alkaline solution concentration and saline solution volume ($[B]_0 \times V_C$), acidic flow rate and saline solution volume ($Q_A \times V_C$) as well as alkaline flow rate and saline solution volume ($Q_B \times V_C$). The 4 positive interactive effects were also observed: initial concentration and flow rate of acidic solution ($[A]_0 \times Q_A$), initial acidic concentration solution and saline solution volume ($[A]_0 \times V_C$), initial alkaline solution concentration and acidic flow rate ($[B]_0 \times Q_A$) as well as acidic and alkaline flow rates ($Q_A \times Q_B$).

In the negative interaction case, it is noticed that the interaction ($[B]_0 \times V_C$) is the most significant. It shows that at high initial alkaline solution concentration (0.5 mol L^{-1}) and at low saline solution volume (0.5 L), the desalination duration is improved.

In the positive interaction case, the acidic and alkaline flow rate interaction ($Q_A \times Q_B$) is the most influenced combination on the desalination efficiency. This interaction improves the ions exchange through anionic- and cationic-exchange membranes when acidic and alkaline flow rates correspond, respectively, to 100 and 50 mL min^{-1}. Thus, DBLs play a dominant role in the ND desalination process efficiency, mainly through CMX. Indeed, the CMX is thicker than the AMX, which leads to an important DBL thickness close to the CMX. Hence, the acidic flow rate acceleration decreases considerably the DBL thickness, contrary to the alkaline flow rate which has no effect.

3.5. Normal probability plot of residuals

The normality of the data can be checked by plotting a normal probability plot of residuals. If the data points on the plot fall fairly close to a straight line, it is considered that the data are normally distributed [45]. Fig. 5 shows a normal data distribution where the experimental points present a straight line.

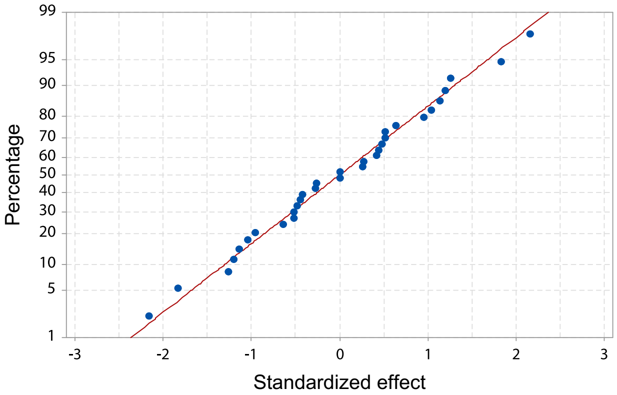

Fig. 5. Normal probability plot of residuals for ND desalination efficiency.

4. Conclusions

This study shows that the initial alkaline solution concentration and the saline solution volume values are the most significant parameters of the ND efficiency.

In the case of initial alkaline solution concentration effect, its increase leads to short desalination duration. This is can be explained by the higher electrical conductivity of AMX in the presence of sodium hydroxide electrolyte and the lower exchange transfer of chloride and hydroxide through AMX than the exchange transfer of sodium and proton through CMX at the beginning of the desalination.

On the other hand, the increase in saline solution volume leads to an important quantity of sodium and chloride ions in the saline compartment which induces a long duration for water desalination.

These results are confirmed by Student's *T*-test and ANOVA. *T* and *F*-tests show that both interactions of initial alkaline solution concentration and saline solution volume as well as acidic and alkaline flow rates are the most statistically significant combinations. Taking into account that the initial alkaline solution concentration and the saline solution volume are the parameters most affecting the desalination efficiency, it is obvious that the combination between these two factors (initial alkaline solution concentration and saline solution volume) will have an important effect on the desalination. The interaction between acidic and alkaline flow rates is also significant due to the DBL thickness through CMX and AMX that has a high impact influence on the ion-exchange kinetics. Moreover, it was found that the data have a normal distribution from the statistical analysis of the experimental data.

In order to accelerate the ions exchange of proton/sodium and hydroxide/chloride through CMX and AMX, respectively, during the ND desalination, it is recommended to use an initial acidic and alkaline concentration solution 6 times higher than the initial saline solution concentration, an acidic and alkaline solutions volume 2.5 times higher than the saline solution volume as well as an acidic flow rate faster than the alkaline flow. After investigating the optimal initial conditions, a real water treatment in South of Tunisia will be carried out by the ND process to estimate the performance of this process for such application.

Nomenclature

A — acidic compartment
AEM — anion-exchange membrane
$[A]_0$ — initial acidic solution concentration (mol L^{-1})
B — alkaline compartment
$[B]_0$ — initial alkaline solution concentration (mol L^{-1})
CS — aline compartment
CEM — cation-exchange membrane
DBL — diffusion boundary layer
DD — Donnan dialysis
ED — electrodialysis
IEM — ion-exchange membrane
K_m — electrical membrane conductivity (mS cm^{-1})
V_C — saline solution volume (L)
ND — neutralization dialysis
Q_A — acidic flow rate (mL min^{-1})
Q_B — alkaline flow rate (mL min^{-1})
Q_C — saline solution flow rate (mL min^{-1})
RO — reverses Osmosis
$b_0, b_1, b_2, b_3, b_4, b_5$ — linear coefficients
$b_{12}, b_{13}, b_{14}, b_{15}, b_{23}, b_{24}, b_{25}, b_{34}, b_{35}, b_{45}$ — second-order interaction terms
A, B, C, D, E — dimensionless coded factors of parameters
Y — responses of desalination duration

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