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# Modeling and analysis of concentration polarization and degree of rejection of boric acid, borates and monovalent cations in low-pressure RO

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**ABSTRACT**

The model for analysis of concentration polarization and the degree of rejection of boric acid, borate anions and monovalent cations has been proposed. The model is based on the following physical assumptions: (1) boron exists in the form of boric acid and deprotonated borate ions; (2) thermodynamic equilibrium between boric acid and borate ions takes place, it was expressed through the dissociation constant; (3) transverse transport is based on the mechanisms of convection due to pressure difference and back diffusion owing to concentration gradient; (4) the governing equation for transverse (convective–diffusion) transport contains six terms such as convective transport of boric acid and borate toward membrane, back diffusion of both components and their transport in permeate. Integrating this equation over the control volume between membrane surface and the upper boundary of diffusion layer has been carried out. It was shown that the conjugated behavior of boric acid, deprotonated borate and monovalent cations takes place. It was demonstrated that the growth of pH is accompanied by the decrease of boric acid along with the increase of borate ions and monovalent cations at the membrane surface and in permeate. The calculations based on this model match the experimental data namely the degree of rejection of boron and monovalent cations (and conductivity) remains almost pH-invariant within the acid and neutral range of pH (at pH < 8.6) whereas it changes sharply within alkaline domain. In particular, the boron rejection (being at the level ~30%–33% at 26 pH < 8.6) goes up to 90% whereas the rejection of monovalent cations (and conductivity) reveals opposite behavior: it goes down from 95% to 70%. Average deviation between calculated and pilot data on the rejection of boron and monovalent cations do not exceed 15% and 9%, respectively. This model can be used for estimating transmembrane fluxes of both components and analyzing the permeate characteristics at different operating conditions. The model can be applied for quantitative analysis of the concentration polarization conjugated for monovalent ions, boric acid and deprotonated borate. This model permits analyzing the influence of transmembrane flux, bulk concentration, physical properties, membrane rejection, channel geometry and pH on the degree of concentration polarization and surface concentration. The presented solution can be segmented and built into a complex algorithm for further modeling and analysis.

*Keywords:* Desalination; Reverse osmosis, Boron removal; Concentration polarization of boric acid and borates; Degree of rejection; Modeling and analysis

## 1. Introduction

Boron is the one of seven essential micronutrients, also called trace elements, required for normal biosynthesis and cell metabolism [1]. This element is affecting the plant and agricultural crops development. Boron is widespread both in the hydrosphere and lithosphere. In nature, boron is never found elemental but always in compounded forms. The concentration of boron in groundwater ranges from 0.3 to 100 mg/L (commonly it is below 2 mg/L). The concentration in seawater is 4.5–5 mg/L although it can reach up to 9.6 mg/L [2–5]. It was revealed that boron is essential element in cell wall formation; in particular, in creation of structural component of primary cell walls. It is involved into the structural integrity of cell walls providing the stability of cell wall matrix. Boron plays an important role in the phenomena of lignification, membrane transports, enzyme interactions, nucleic acid synthesis, phenol and carbohydrate metabolism, transport of sugar, polyol, hydroxy acid, etc., but it is getting toxic when the concentration exceeds its critical limit [6–9]. The range between permissible and harmful level of boron is appeared to be extremely narrow. The main aspects of this problem were intensively scrutinized in different studies [5,10–12].

An excess of boron can be toxic for all the biological species: plants, agricultural crops, animals and human beings. A negative impact of elevated boron concentration for the first time was observed in Israel in 1997 after commissioning seawater reverse osmosis (SWRO) plant in Eilat [5]. Farmers using desalted water for irrigation noticed poisoning of crops and partly discolored leaves. Later, boron was identified as the toxic element responsible for this effect. Since that time different post-treatment methods for boron removal after SWRO have been proposed and developed [13]. The physiological adverse effect of boron involves the reduction of root cell division, retarded shoot and root growth, inhibition of photosynthesis, deposition of lignin and suberin, decrease in leaf chlorophyll, etc. [14,15]. In most crops, the symptoms of boron toxicity are shown as burned edges on the older leaves, yellowing of the leaf tips, accelerated decay and ultimately plant expiration [16].

For many years boron was not considered as a toxic element. In 1958, 1963 and 1971 there was no even mentioning boron in the World Health Organization (WHO) standards. A limit of 0.3 mg/L for boron in drinking water was recommended by WHO only in 1993. This limit was considered as a provisional value due to the lack of a comprehensive toxicological impact assessment and unavailability of reliable technology for boron removal and this value remained as a guideline for many years. Later that limit was re-evaluated and the value of 0.5 mg/L was recommended for drinking water sources (including seawater) it was set by the agricultural sector focusing mainly on the unlimited irrigation of boron-sensitive crops. Recently, WHO recommended to elevate the limit for boron in drinking water up to 2.4 mg/L, at the same time EU still suggests the maximum concentration of boron in drinking water at the level of 1.0 mg/L. However, some utilities may still impose seawater desalination limits as low as 0.3–0.5 mg/L focusing mainly on the agriculture-related aspects [5]. Currently, the national guideline values are still randomized: some countries do not have federal regulations on boron; many of them assume the maximum boron concentration much higher than the WHO guidelines, etc. [5,17–19].

### *1.1. Technologies for boron removal*

Lack of reliable technologies for boron removal and methods of their analysis still remain the hampering factors for successful implementation of membrane desalination. It is well-known that boron is effectively removed by thermal desalination but the level of removing by reverse osmosis (RO) was appeared to be insufficient. Since membrane technology is increasing over the last years, the boron-related issue is becoming challenging aspect nowadays. There is a wide spectrum of technological concepts for boron elimination proposed over the last years. Conditionally they can be subdivided into two broad groups such as: (A) the group covering the schemes with multipass and cascade membrane post-treatment and (B) so-called "hybrid" group, the group including combination of membrane processes with conventional operations such as ion exchange, adsorption, electrocoagulation, electrodialysis, complexation on the stage of pretreatment, etc. [20–34].

According to Faigon and Hefer [23] cascade design allows adjusting the operating conditions (since the membrane removal of boron is dependent upon different operating conditions). According to Oo and Song [24] boron rejection increases from around 50%–75% at pH 7–8 to over 95% at pH 10.5 and is mainly due to the growth of the proportion of borate ions caused by the pH rises. According to Prats et al. [25], Koseoglu et al. [26] and Cengeloglu et al. [27], the pressure growth tends to increase the boron rejection. The removal of boron, however, was decreased by the increase in feed pressure from 700 to 800 psi where Filmtec SW30HR membranes were used [26]. Complexion reactions can also be used in order to increase boron rejection and this is achieved by the addition of Fe cations or other complexing agent which cause the formation of boron containing complexes (Qin et al. [28] and Geffen et al. [29]).

Technological concepts considered by Redondo et al. [21] are based on hybridization of the double pass schemes with cascade and recycle. Research done by Taniguchi et al. [22] was focused on combining SWRO, brackish water reverse osmosis (BWRO) and ion exchange using boron-selective resins (BSR). The review of data was presented by Bodzek [20] where different processes such as ion exchange, adsorption based on BSR; RO, multistage configurations with RO, hybrid systems combining sorption with membrane filtration, polymer-enhanced ultrafiltration, etc. were considered.

Minif et al. [30] studied coupling of nanofiltration (NF) with RO for boron removal. The pilot test was conducted on the spiral modules by Osmonics equipped by AG 2514 TF and HL 2514T membranes. The results indicated that boron rejection mostly depends upon membrane type, pH level, and to a lesser extent on the recovery. The study done by Tu et al. [31] investigated coupling effect of feed pH and ionic strength on the rejection of boron by the combined NF/RO system. NF membranes (namely NF270 and NF90) and RO membranes (BW30, SW30 and UTC80) were used. Boron rejections by the NF90 and the NF270 membranes were only 10% and 30% lower than those by the other three RO membranes. On the other hand, the permeability of the NF membranes was 3–11 times higher than those of the RO membranes. The reported data suggest a possibility of using NF membranes for the second pass in seawater desalination applications to avoid over demineralization of the final product water. It was revealed that the growth of ionic strength (up to 42.5 mM) is accompanied by considerable increase in boron rejection by both the NF270 and the BW30 membranes. Results reported in reference [31] suggest that the rejection of boron on the second pass could be further optimized by increasing the salt passage after the first pass. The review published by Tu et al. [32] provides a summary on the removal of boron in seawater desalination based on NF/RO. It was highlighted that the mutually conjugated behavior of the process parameters such as feed temperature, ionic strength, pH and the rejection of boron represent a potential for further optimization.

The effects of membrane type on the effectiveness of rejection of boric acid, monoborate and selected boron–polyol complexes by selected RO membranes were examined. Boron complexes with d-mannitol, sodium d-gluconate as well as *N*-methyl d-glucamine were considered by Dydo et al. [33]. Based on the laboratory testing, the mass transport coefficients, including the permeability and reflection coefficients, specific for each molecular species, were determined. The boric acid rejection was descending through the following membrane order: SW30 > BW30 > TW30 > XLE, whereas at low permeate flux rates the XLE membrane was slightly more efficient for boric acid rejection than the TW30 membrane [33]. The review by Akerman et al. [17] covers the main processes to be used for born eliminations. The treatment of irrigating water based on the hybrid membrane-based systems was considered in reference [17] where the processes of electrocoagulation, electrodialysis and adsorption were tested. Final boron concentration from 1.5 to 0.5 mg/L was achieved. Boron removal in the process of desalination of geothermal water was studied by Tomaszewska and Bodzek [34]. The tests were conducted on hybrid UF–RO system. The system was equipped by Dow Filmtec BW30HR-440i spiral elements based on polyamide thin-film composite membranes. The permeate production was 1 $m^3$/h. It was found that the rate of boron removal in acidified range of feedwater depended on the boron concentration. The highest removal rate at 56% was obtained at 2.5 mg B/L; 48% in water with 8.98 mg B/L and 12% in the water with concentration of 96.73 mg B/L. The highest boron removal rate being equal to 96%–97% was obtained in water with pH 10–11 containing 10 mg B/L, regardless of the feedwater uptake ratio. Feedwaters with a high boron concentration of 100 mg/L and pH 10 yielded 66% of boron removal. Efficient and stable performance of the desalination system equipped by BWRO membranes was achieved using geothermal waters containing 7 g/L total dissolved solids and boron concentration of up to 10 mg/L. Taking into account that the low pressure was applied in the RO process 1.1 MPa, the rate of rejection in terms of conductivity was rather high (~96%–97%).

### 1.2. Existing methods of process analysis

Physical behavior of multicomponent solution has to be taken into account in the development of design and operation software. The degree of boron rejection is strongly influenced not only by pH, but by ionic strength, temperature, system recovery, etc. as well, that's why oversimplified correlations imbedded into existing software are responsible for certain inaccuracy of the results that makes their applicability very limited.

An impact of pH on boron rejection has been analyzed by many authors; in particular, the experimental data by Hung et al. [12] showed that boron rejection increases as feed pH goes up due to the growth of borate fraction. It was increased from 70%–85% at pH 7.5 to 90%–98% at pH 10. Experimental data [23] demonstrated the growth of boron rejection from 78% (at pH 8) to 88% (at pH 8.55). According to the data submitted by Glueckstein and Priel [35], boron rejection goes from 75% (pH 7) to 97% (pH 11) and from 87% (pH 7) to 94% (pH 9) for low-pressure BWRO and seawater RO, respectively. Therefore, these published data demonstrates the similar shape of the rejection curve where the digital values are dependent upon the type of membrane; operating temperature, process characteristics, etc. The degree of boron rejection remains pH invariant within the acidic and neutral range of pH (normally at pH < 8.6) while it changes sharply within an alkaline domain. An attempt to establish the correlation between boron and salt rejection using an irreversible thermodynamic model was done by Choi et al. [36]. It was stated that the rejection of boron follows a different mechanism from those of other ionic solutes and could not be correlated with ion rejection [36].

Although there is a wide set of data on boron rejection, however, the examination of existing published data has revealed some disputable statements, such as a lack of meaningful relationship between the transport of boric acid or borates (see Kim et al. [37]). The study published by Tu et al. [38] comprises another questionable conclusion that the values of boron rejection were not correlated with sodium rejection, indicating that the boron and sodium are rejected by different mechanisms. According to Hyung and Kim [10] boron rejection was dependent upon pH while the rejection of other ionic species is not indicative to boron rejection. According to the study by Choi et al. [36] boron rejection is not proportional salt rejection.

Unlike the prevalent published data, our previous pilot study [39] indicated an obvious relationship between the rejection of monovalent ions $[Na]^{-1}$, boric acid $[H_3BO_3]$ and negatively charged borate ion $[B(OH)_4]^{-1}$. Those pilot data were used for verification and validation of the model.

In spite of the difference in underlying assumptions the existing methods of analysis use similar techniques for evaluation of mass-transfer coefficients. They are based on the film theory and applicable for monocomponent systems. Since the considered cases are characterized by multifold difference between the concentration of target component (e.g., boron) and component that controls the level of salinity and ionic strength (e.g., monovalent ions), it should be noted that the existing methods suitable for analysis of monocomponent concentration polarization (CP) cannot be applied in those cases. For that reason proposed study focuses on modeling and development of relationship between rejection of monovalent ions, boric acid and negatively charged borate ions. The model proposed in this study can be used for further quantitative analysis of CP for conjugated multicomponent systems.

## 2. The main assumptions and premises of the model

### 2.1. Physical behavior and governing equation

Boron exists in the form of boric acid $[H_3BO_3]$ and negatively charged borate ion $[B(OH)_4]^{-1}$. Boric acid behaves as a weak Lewis acid as follows:

$$H_3BO_3 + H_2O = B(OH)_4^{-1} + H^{+1} \tag{1}$$

The main components are boric acid $[H_3BO_3]$, negatively charged borate ion $[B(OH)_4]^{-1}$ and monovalent ions characterizing the level of ionic strength. Since the concentration of boron in seawater is around 4.8 mg/L, it was accepted that only mononuclear species $[H_3BO_3]$ and $[B(OH)_4]^{-1}$ exist in seawater [40]. The distribution between boric acid and borate ion can be expressed through the apparent first acid constant ($K_{a1}$) that in turn depends on temperature, pressure, pH and ionic strength. It is defined as:

$$K_{a1} = \frac{\left[\mathrm{B\,(OH)_4^{-1}}\right]\{\mathrm{H}^+\}}{\left[\mathrm{H_3BO_3}\right]} \tag{2}$$

The first acid dissociation constant p$K_{a1}$ = 9.14 (at $t$ = 25°C) in a low ionic strength solution, and p$K_{a1}$ = 8.68 (at $t$ = 25°C) in high ionic strength solutions such as seawater. The equilibrium constant is defined using the concentration of a boron species and the activity of proton. Since boric acid is weak, the majority exists as uncharged boric acid $[H_3BO_3]$ in the natural pH range. However, as pH increases, the fraction of negatively charged borate $[B(OH)_4]^{-1}$ increases and becomes the dominant. Assuming the second and third acid constants ($K_{a2}$ and $K_{a3}$) of boric acid are negligible, the total amount of boron $[B_\Sigma]$ is equal to the sum of the forms of boric acid and borate anion $[B_\Sigma] = [H_3BO_3] + [B(OH)_4]^{-1}$. Based on Eq. (2), we can express the concentration of boric acid $[H_3BO_3]$ and negatively charged borate $[B(OH)_4]^{-1}$ as follows:

$$\left[\mathrm{H_3BO_3}\right] = f_{\mathrm{B1}}\left[\mathrm{B}_\Sigma\right] \tag{3}$$

$$\left[\mathrm{B(OH)_4}\right]^{-1} = f_{\mathrm{B2}}\left[\mathrm{B}_\Sigma\right] \tag{4}$$

where $f_{B1}$ and $f_{B2}$ are equilibrium fractions of boric acid and borate, respectively.

$$f_{\mathrm{B1}} = \frac{\{\mathrm{H}^+\}}{\{\mathrm{H}^+\} + K_{a1}} \tag{5}$$

$$f_{\mathrm{B2}} = \frac{K_{a1}}{\{\mathrm{H}^+\} + K_{a1}} \tag{6}$$

In terms of these variables the concentration of boric acid and borate can be expressed as $C_{B1} = f_{B1}C_{B\Sigma}$ and $C_{B2} = f_{B2}C_{B\Sigma}$ where $C_{B1} = [H_3BO_3]$; $C_{B2} = [B(OH)_4]^{-1}$ and $C_{B\Sigma} = [H_3BO_3] + [B(OH)_4]^{-1}$.

The fluid is assumed to be incompressible, continuous and isothermal with uniform density field under the steady-state (time independent) conditions. Transverse transport is based on the following mechanisms: convection due to pressure difference and back diffusion owing to concentration gradient. Since there are boric acid $[H_3BO_3]$ and negatively charged borate ions $[B(OH)_4]^{-1}$ so the governing equation for transverse transport contains six terms:

$$\begin{aligned} VC_{\mathrm{B1}}(z) + VC_{\mathrm{B2}}(z) - D_{\mathrm{B1}}\frac{dC_{\mathrm{B1}}(z)}{dz} - D_{\mathrm{B2}}\frac{dC_{\mathrm{B2}}(z)}{dz} \\ = VC_{\mathrm{PERMEATE\_B1}} + VC_{\mathrm{PERMEATE\_B2}} \end{aligned} \tag{7}$$

where $V$ is transverse flux; $D$ is diffusivity and $C$ is local concentration. The first and second terms on the left-hand side represent convection of boric acid and borate toward membrane; the third and fourth terms represent back diffusion from the membrane surface; the right-hand side terms represent convective transport in permeate. All the characteristics related to boric acid or borate are marked by the subscripts B1 or B2, respectively. All the transport constituents are illustrated in Fig. 1.

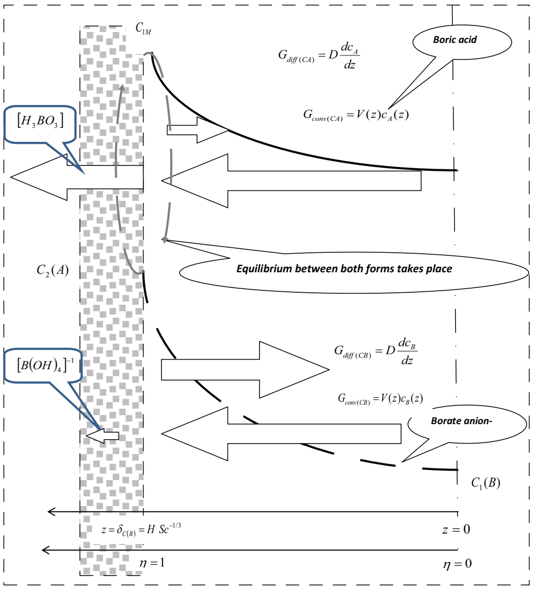


Fig. 1. Illustration of transport mechanisms.

### 2.2. Auxiliary dimensionless variables

For the further mathematical treatment to be simplified some auxiliary non-dimensional variables were introduced. In particular, the variable η can be used for modeling within diffusion layer.

$$\eta = z/\delta_C \tag{8}$$

This variable ranges from $\eta = 0$ at the upper boundary of diffusion layer ($z = 0$ and $c_{z=0} = C_1$) to $\eta = 1$ at membrane surface ($z = \delta_C$ and $c_{z=\delta} = C_{1M}$). According to [41] the ratio of the thickness of diffusion layer to the viscous one can be assumed as:

$$\delta_C/\delta_W \approx Sc^{-1/3} \tag{9}$$

Symmetric channel of plate and frame type was considered in this model (the control volume ranges from membrane surface to the channel centerline). In this case, the dynamic (viscous) layer $\delta_W$ can be assumed to be equal to the half height of the channel, thus $H \approx \delta_W$.

Replacing $\delta_W$ in Eq. (9) by $H$ we get:

$$\delta_C \approx H\ Sc^{-1/3} \tag{10}$$

Combining Eq. (10) with Eq. (8), we get the following relation between physical and non-dimensional variables $z$ and η.

$$z = H\ Sc^{-1/3}\eta \tag{11}$$

$$dz = H\ Sc^{-1/3}d\eta \tag{12}$$

Auxiliary variables such as the degree of membrane rejection, degree of CP (or polarization module $\alpha$) can be used for linking the concentration in bulk, at membrane and in permeate. Membrane rejection expressed through the concentration in bulk and permeate is referred to as observed (or apparent) degree of rejection whereas the degree of rejection expressed through the concentration at membrane surface is referred to as true (or intrinsic) degree of rejection.

$$R_{\mathrm{APPARENT}} = 1 - C_{\mathrm{PERMEATE}} / C_{\mathrm{BULK}} \tag{13}$$

$$R_{\mathrm{TRUE}} = 1 - C_{\mathrm{PERMEATE}} / C_{\mathrm{MEMBRANE}} \tag{14}$$

The polarization module $\alpha$ can be assumed as a quantitative measure of CP.

$$\alpha = C_{\mathrm{MEMBRANE}} / C_{\mathrm{BULK}} \tag{15}$$

It can be used to link the bulk and permeate values of concentration.

$$C_{\mathrm{PERMEATE}} = C_{\mathrm{BULK}}\ \alpha\ (1 - R_{\mathrm{TRUE}}) \tag{16}$$

For the first approximation to be done the degree of rejection can be assumed based on experimental data at so-called "boundary conditions", at pH 7 (dominant boric acid) and at pH 10 (dominant borate ion). The estimation is based on the following assumptions: (1) an equilibrium between boric acid and borate ions takes place; (2) the true degree of rejection of sodium, boric acid and borate does not depend upon pH and (3) Sherwood-based correlation can be used for the estimation of CP degree at the boundary conditions. Relying upon the above-mentioned premises and assumptions the true degree of rejection for sodium, boric acid and borate were estimated to be $R_{\mathrm{TRUE\_Na}} = 0.94$, $R_{\mathrm{TRUE\_B1}} = 0.25$ and $R_{\mathrm{TRUE\_B2}} = 0.97$.

Assuming an equilibrium between protonated and deprotonated forms, the concentration at membrane surface can be expressed through the equilibrium fractions of boric acid and borate (see Eqs. (5) and (6) for $f_{\mathrm{B1}}$ and $f_{\mathrm{B2}}$, respectively). Implying that both forms are characterized by different rejection the true degree of rejection (overall value) can be expressed in terms of boric acid and borate as follows:

$$R_{\mathrm{TRUE\_\_B1\ \&\ B2}} = 1 - f_{\mathrm{B1}}(1 - R_{\mathrm{TRUE\_\_B1}}) - f_{\mathrm{B2}}(1 - R_{\mathrm{TRUE\_\_B2}}) \tag{17}$$

## 3. Modeling

### 3.1. Polarization and rejection of boric acid and borate

Further analysis is based on the rearrangement of governing equation, see Eq. (7). Since an equilibrium between boric acid and borate at any longitudinal control section takes place, the concentration of boric acid and borate ion can be expressed as $C_{\mathrm{B1}} = f_{\mathrm{B1}} C_{\mathrm{B\Sigma}}$ and $C_{\mathrm{B2}} = f_{\mathrm{B2}} C_{\mathrm{B\Sigma}}$, where $C_{\mathrm{B\Sigma}}$ overall concentration; $C_{\mathrm{B1}}$ boric acid concentration, $C_{\mathrm{B2}}$ borate concentration; $f_{\mathrm{B1}}$ and $f_{\mathrm{B2}}$ pH-dependent fractions of boric acid and negatively charged borate at the state of equilibrium (Eqs. (5) and (6)). Therefore, the convective terms on the left-hand side of Eq. (7) can be rewritten as:

$$V\,C_{\mathrm{B1}}(z) = V\,C_{\mathrm{B\Sigma}}(z)\ f_{\mathrm{B1}} \tag{18}$$

$$V\,C_{\mathrm{B2}}(z) = V\,C_{\mathrm{B\Sigma}}(z)\ f_{\mathrm{B2}} \tag{19}$$

In the same way, replacing $C_{\mathrm{B1}}$ and $C_{\mathrm{B2}}$ in the derivatives on the left-hand side of Eq. (7), the diffusive terms on the left-hand side of Eq. (7) can be rewritten as:

$$D_{\mathrm{B1}} \frac{dC_{\mathrm{B1}}(z)}{dz} = D_{\mathrm{B1}} f_{\mathrm{B1}} \frac{d\,C_{\mathrm{B\Sigma}}(z)}{dz} \tag{20}$$

$$D_{\mathrm{B2}} \frac{dC_{\mathrm{B2}}(z)}{dz} = D_{\mathrm{B2}} f_{\mathrm{B2}} \frac{d\,C_{\mathrm{B\Sigma}}(z)}{dz} \tag{21}$$

The terms on the right-hand side of Eqs. (18) and (19) can be rewritten in terms of CP module $\alpha$ and the degree of rejection $R$ as follows:

$$VC_{\mathrm{PERMEATE\_B1}} = VC_{\mathrm{B1}}\alpha(1 - R_{\mathrm{TRUE\_B1}}) \tag{22}$$

$$VC_{\mathrm{PERMEATE\_B2}} = VC_{\mathrm{B2}}\alpha\left(1 - R_{\mathrm{TRUE\_B2}}\right) \tag{23}$$

where $R_{\mathrm{TRUE\_B1}}$ and $R_{\mathrm{TRUE\_B2}}$ are the values of the true degree of rejection of boric acid [$H_3BO_3$] and negatively charged borate [$B(OH)_4$]$^{-1}$. Combining Eqs. (17) and (18)–(21) we get:

$$\begin{aligned} &V\,(f_{\mathrm{B1}} + f_{\mathrm{B2}})\ C_{\mathrm{B\Sigma}}(z) - (D_{\mathrm{B1}} f_{\mathrm{B1}} + D_{\mathrm{B2}} f_{\mathrm{B2}}) \frac{dC_{\mathrm{B\Sigma}}(z)}{dz} \\ &\quad = V\,C_{\mathrm{B\Sigma}}(z)\ \alpha\ [f_{\mathrm{B1}}(1 - R_{\mathrm{TRUE\_B1}}) + f_{\mathrm{B2}}(1 - R_{\mathrm{TRUE\_B2}})] \end{aligned} \tag{24}$$

Further rearrangement and separation of the variables yields:

$$\frac{dC_{\mathrm{B\Sigma}}}{C_{\mathrm{B\Sigma}}} = \frac{1 - \alpha\ [f_{\mathrm{B1}}(1 - R_{\mathrm{TRUE\_B1}}) + f_{\mathrm{B2}}(1 - R_{\mathrm{TRUE\_B2}})]}{(D_{\mathrm{B1}} f_{\mathrm{B1}} + D_{\mathrm{B2}} f_{\mathrm{B2}})}\ V\,dz \tag{25}$$

Introducing an auxiliary variable $\Psi$ we get:

$$\frac{dC_{\mathrm{B\Sigma}}}{C_{\mathrm{B\Sigma}}} = \Psi\ dz \tag{26}$$

where

$$\Psi = \frac{1 - \alpha\ [f_{\mathrm{B1}}(1 - R_{\mathrm{TRUE\_B1}}) + f_{\mathrm{B2}}(1 - R_{\mathrm{TRUE\_B2}})]}{(D_{\mathrm{B1}} f_{\mathrm{B1}} + D_{\mathrm{B2}} f_{\mathrm{B2}})}\ V \tag{27}$$

For the further mathematical treatment to be simplified the dimensionless variable, $z = H\ \mathrm{Sc}^{-1/3}\ \eta$ can be used (see Eq. (11)). Inserting it into Eq. (26) gives:

$$\frac{dC_{\mathrm{B\Sigma}}}{C_{\mathrm{B\Sigma}}} = \Psi\ H\ Sc^{-1/3} d\eta \tag{28}$$

Further integration gives:

$$\ln C_{B\Sigma} = \Psi\ H\ Sc^{-1/3}\eta + \mathrm{const} \tag{29}$$

For the constant of integration const to be evaluated, the conditions at the upper boundary of diffusion layer ($z = 0$, $c_{z=0} = C_1$ and $\eta = 0$) were used. Therefore:

$$\text{const} = \ln C_{\mathrm{B}\Sigma(\mathrm{BULK})} \tag{30}$$

$$\ln C_{\mathrm{B}\Sigma} = \Psi \; H \; Sc^{-1/3}\eta + \ln C_{\mathrm{B}\Sigma(\mathrm{BULK})} \tag{31}$$

$$\ln \frac{C_{\mathrm{B}\Sigma}(\eta)}{C_{\mathrm{B}\Sigma(\mathrm{BULK})}} = \frac{1-\alpha \; [f_{\mathrm{B1}}(1-R_{\mathrm{TRUE\_B1}}) + f_{\mathrm{B2}}(1-R_{\mathrm{TRUE\_B2}})]}{(D_{B1}f_{B1} + D_{B2}f_{B2})} \; V \; H \; Sc^{-1/3}\eta \tag{32}$$

At $\eta = 1$, the numerator on the left-hand side $C_{\mathrm{B}\Sigma}(\eta)$ gives the surface concentration $C_{\mathrm{B}\Sigma(\eta=1)}$, therefore, this ratio can be replaced by the module of CP (CP module), $\alpha = C_{\mathrm{B}\Sigma(\eta=1)}/C_{\mathrm{B}\Sigma(\mathrm{BULK})}$. Thus, Eq. (30) can be rewritten in terms of CP module as follows:

$$\ln \alpha = \frac{1-\alpha \; [f_{\mathrm{B1}}(1-R_{\mathrm{TRUE\_B1}}) + f_{\mathrm{B2}}(1-R_{\mathrm{TRUE\_B2}})]}{(D_{\mathrm{B1}}f_{\mathrm{B1}} + D_{\mathrm{B2}}f_{\mathrm{B2}})} \; V \; H \; Sc^{-1/3} \tag{33}$$

This equation represents a relation between CP module $\alpha$, physical properties and process parameters. Solving Eq. (33) for CP module gives a set of calculated projections at different transverse velocity (see Fig. 2).

Assuming an equilibrium between both forms of boron (boric acid and borate) the concentration at membrane surface can be expressed as follows:

$$C_{\mathrm{B1(MEMBRANE)}} = f_{\mathrm{B1}} \; \alpha \; C_{\mathrm{B}\Sigma(\mathrm{BULK})} \tag{34}$$

$$C_{\mathrm{B2(MEMBRANE)}} = f_{\mathrm{B2}} \; \alpha \; C_{\mathrm{B}\Sigma(\mathrm{BULK})} \tag{35}$$

where $\alpha$ is CP module (Eq. (33)); $f_{\mathrm{B1}}$ and $f_{\mathrm{B2}}$ are equilibrium fractions of boric acid and borate ion, respectively (Eqs. (5) and (6)). Calculated profiles are shown in Fig. 3.

Since an experimental characterization of both bulk and permeate samples was based on inductively coupled plasma mass spectrometry (ICP-MS) method, therefore, it gave us an overall concentration of boric acid and borate. In this regard for the results to be compared the permeate concentration and apparent degree of rejection was estimated as:

$$\begin{aligned} C_{\mathrm{B1\&B2(PERMEATE)}} &= f_{\mathrm{B1}} \; \alpha \; C_{\mathrm{B}\Sigma(\mathrm{BULK})}\left[1-R_{\mathrm{B1}}\right] \\ &\quad + f_{\mathrm{B2}} \; \alpha \; C_{\mathrm{B}\Sigma(\mathrm{BULK})}\left[1-R_{\mathrm{B2}}\right] \end{aligned} \tag{36}$$

$$R_{\mathrm{APPARENT\_B1\&B2}} = 1 - C_{\mathrm{B1\&B2(PERMEATE)}}/C_{\mathrm{B1\&B2(BULK)}} \tag{37}$$

### *3.2. Polarization and rejection of the cations of the first group*

Due to the difference in transmembrane transport of boric acid and borate ion, selective accumulation of negatively charged borates within the boundary layer takes place this phenomenon, in turn, enhances the accumulation of cations in order to maintain electroneutrality of the layer.

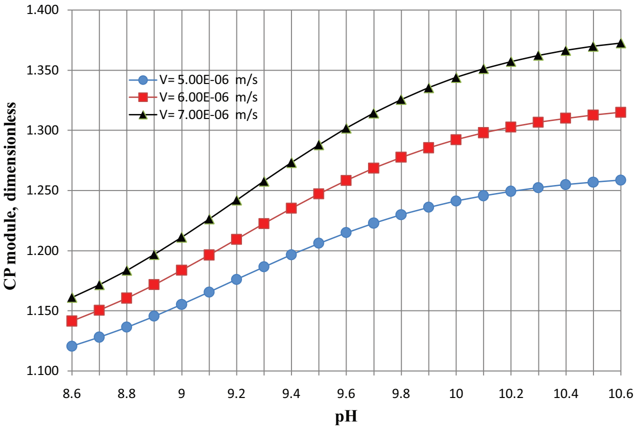


Fig. 2. CP module for boric acid and deprotonated borate ion (calculated profiles are based on Eq. (33)). Input data: $D_{\mathrm{B1}} = 2 \times 10^{-9}$ m/s; $D_{\mathrm{B2}} = 1.5 \times 10^{-9}$ m/s; $V = 7.00 \times 10^{-6}$ m/s; $H = 2.0 \times 10^{-3}$ m; Sc = 800 ($Sc^{-1/3} = 0.10772$); $R_{\mathrm{TRUE\_B1}} = 0.25$; $R_{\mathrm{TRUE\_B2}} = 0.97$; $R_{\mathrm{TRUE\_Na}} = 0.94$.

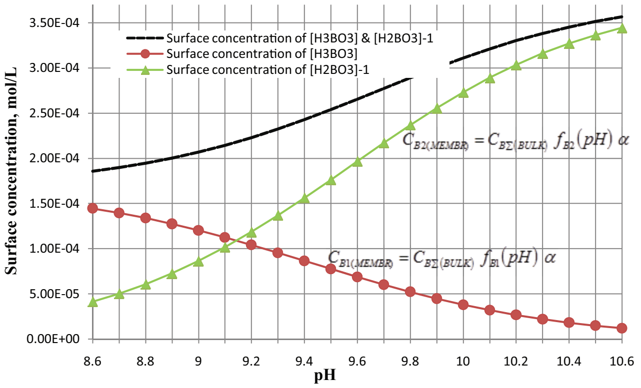


Fig. 3. Concentration of boric acid and deprotonated borate at the membrane surface (see Eqs. (34) and (35)). Input data: $D_{\mathrm{B1}} = 2 \times 10^{-9}$ m$^2$/s; $D_{\mathrm{B2}} = 1.5 \times 10^{-9}$ m$^2$/s; $V = 7.0 \times 10^{-6}$ m/s; $H = 2.0 \times 10^{-3}$ m; Sc = 800 ($Sc^{-1/3} = 0.10772$); $R_{\mathrm{TRUE\_B1}} = 0.25$; $R_{\mathrm{TRUE\_B2}} = 0.97$; $R_{\mathrm{TRUE\_Na}} = 0.94$.

Boron in seawater usually exists as weak boric acid [21,34]. In acidic and near neutral conditions, it is boric acid commonly written as $H_3BO_3$. It does not dissociate in aqueous solution, but it is acidic due to its interaction with water molecule, forming tetrahydroborate (see Eq. (1)). At pH 7–10 polymeric anions containing structural OH groups (polyhydroxoborates) can be formed (if the concentration of boron is higher than 0.025 mol/L). In considered case the concentration of boron is almost two orders lower than the level required for the formation of polymeric anions (polyhydroxoborates). At elevated pH the accumulation of negatively charged borates takes place that makes the neutralization reaction possible. Different forms of negatively charged borates can be formed in that case. For the calculations to be simplified the formation of fictitious deprotonation ion series can be assumed [42]; in particular, dihydrogen borate $[H_2BO_3]^{-1}$, hydrogen borate $[HBO_3]^{-2}$ and borate $[BO_3]^{-3}$. The reaction of neutralisation can be illustrated as follows:

$$\begin{aligned} &H_3BO_3 + 3Na^+ \Leftrightarrow NaH_2BO_3 + 2Na^+ + H \\ &\quad \Leftrightarrow Na_2HBO_3 + 2H^+ + Na^+ \Leftrightarrow Na_3BO_3 + 3H^+ \end{aligned} \tag{38}$$

Since boric acid is a tribasic one, the growth of pH can be accompanied by selective accumulation of sodium cations at membrane as $[BO_3]^{-3} \approx 3[Na]^+$. Therefore, the concentration of the cations of the first group at the membrane surface is expected to be linked with the surface concentration of borate ions. Based on the calculated concentration of borate at the surface $C_{\mathrm{B2(MEMBRANE)}}$, see Eq. (35), and assuming that $C_{\mathrm{Na(MEMBRANE)}} \approx k\, C_{\mathrm{B2(MEMBRANE)}}$ we can estimate the sodium concentration at the membrane surface $C_{\mathrm{Na(MEMBRANE)}}$ and in permeate along with an apparent degree of rejection.

$$C^{\mathrm{Calculated}}_{\mathrm{PERMEATE(Na)}} = C^{\mathrm{Calculated}}_{\mathrm{MEMBRANE(Na)}} \left[1 - R_{\mathrm{TRUE(Na)}}\right] \tag{39}$$

$$R^{\mathrm{Calculated}}_{\mathrm{APPARENT(Na)}} = 1 - C^{\mathrm{Calculated}}_{\mathrm{PERMEATE(Na)}} \Big/ C_{\mathrm{BULK\ (Na)}} \tag{40}$$

The curve based on Eqs. (39) and (40) are shown in Figs. 6 and 7.

## 4. Experimental part

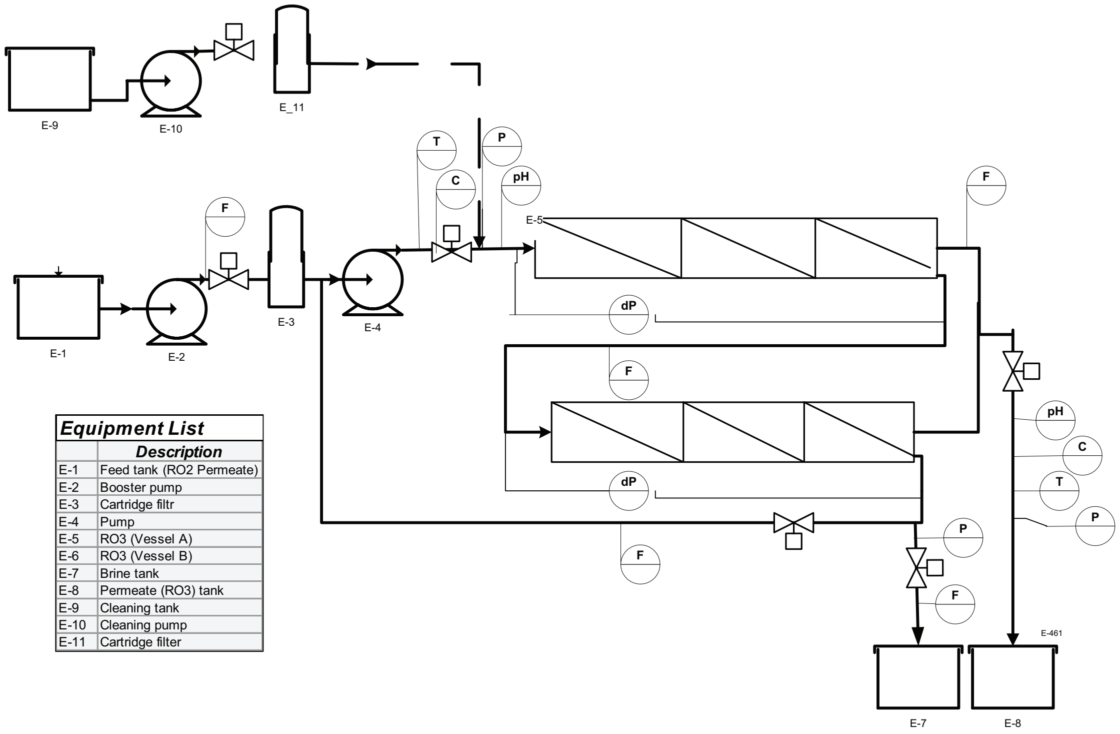
### 4.1. Pilot scheme (low-pressure RO post-treatment for boron elimination)

Experimental data gathered through the pilot study were used for verification of the model. The pilot system was equipped by spiral element, Woongjin Chemical RE4040-BE, which was installed on the third pass RO where thin-film composite polyamide membranes were used, see CSM RO Catalogue, [43]. Total membrane area is 47.4 m$^2$ where six elements with effective membrane area 7.9 m$^2$ per element were assembled within the pressure vessel. Normalized permeability ranges from 3.4 to 5.6 m$^3$/m$^2$ h bar; operating pressure varies from 5.3 to 8.5 bar. Permeate after the second RO pass entered the third stage after pH adjustment. Permeate after the second RO pass was characterized by the following values: conductivity within the range from 53 to 160 μS/cm (salinity equivalent to the estimated conductivity was ranged from $3.27 \times 10^{-4}$ to $1.12 \times 10^{-4}$ mol/L): concentration of sodium ranges from $3.7 \times 10^{-4}$ to $1.52 \times 10^{-4}$ mol/L. The pilot system was located at Al-Mirfa site (UAE). Experimental data were presented in reference [39]. Simplified fragment of the flow diagram of the pilot plant was shown in Fig. 4.

### 4.2. Water sample characterization

The primary experimental data on boron, salinity, conductivity and pH were received during the pilot study. The salinity was estimated based on both electrical conductivity and sodium concentration. Their values vs. pH revealed similar behavior in both cases. It was shown that the degree of rejection of boron and salinity remains pH independent in acidic and neutral range of pH while it changes sharply within alkaline domain. The salinity was expressed though the equivalency to conductivity and sodium concentration.

Kmiecik et al. [44] gave comparison between analytical methods used for boron characterization such as ICP-MS and inductively coupled plasma optical emission spectroscopy. The results obtained were compared and correlated. In our study, ICP-MS technique was used for boron analysis. This technique gives overall concentration for both boric acid and negatively charged borate $[\mathrm{B(OH)_4}]^{-1}$. Some analytical techniques for quantitative characterization of boron are presented in references [44,45]. An experimental relation between the degree of observed rejection of boron and salinity vs. pH, see Fig. 5 (for pilot data see [39]).



**Equipment List**

| | Description |
|---|---|
| E-1 | Feed tank (RO2 Permeate) |
| E-2 | Booster pump |
| E-3 | Cartridge filtr |
| E-4 | Pump |
| E-5 | RO3 (Vessel A) |
| E-6 | RO3 (Vessel B) |
| E-7 | Brine tank |
| E-8 | Permeate (RO3) tank |
| E-9 | Cleaning tank |
| E-10 | Cleaning pump |
| E-11 | Cartridge filter |

Fig. 4. The low pressure section of the pilot plant. The system was equipped by spiral element based on film composite polyamide with total membrane area of 47.4 m$^2$. Manufacturer: Woongjin Chemical RE4040-BE [43].

## 5. Comparison of calculated results with pilot data and verification of the model

The pilot data on permeate concentration and the degree of rejection were used for the verification of the model (for pilot data see Fig. 5). The model based on Eqs. (33)–(37) gives the concentration of boron at membrane and in permeate (both in the form of boric acid and deprotonated borate). Comparison of experimental and calculated data is shown in Figs. 6 and 7. It was shown that the growth of pH is accompanied by the decrease of boric acid along with the increase of monovalent cations and borate ions at the membrane surface and in permeate. In particular, the boron rejection (being at the level ~30%–33% at pH < 8.6) goes up to 90% whereas the rejection of monovalent cations (and conductivity) reveals opposite behavior: it goes down from 95% to 70 %. The calculations based on this model match the experimental data on boron and monovalent cations namely average deviation between calculated and pilot data on the rejection of boron and monovalent cations do not exceed 15% and 9%, respectively.

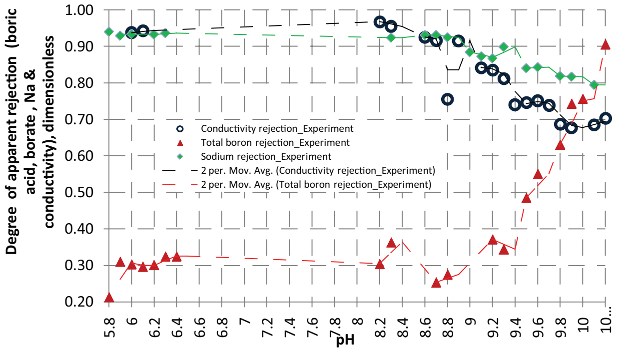


Fig. 5. The degree of observed rejection of boron and conductivity vs. pH. Experimental data received during the pilot study [39].

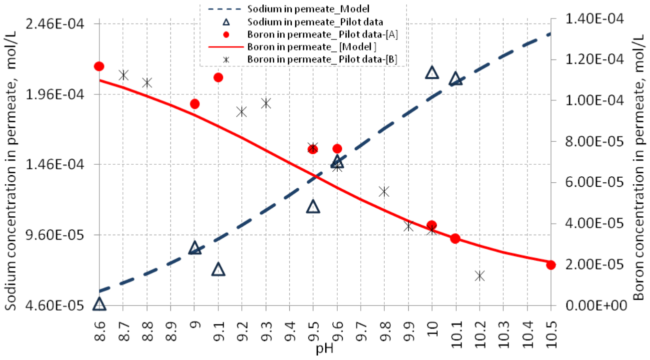


Fig. 6. Experimental and calculated concentration of boron and sodium in permeate (calculations are based on Eqs. (36) and (39)). Input data: $D_{B1} = 2 \times 10^{-9}$ m$^2$/s; $D_{B2} = 1.5 \times 10^{-9}$ m$^2$/s; $V = 7.0 \times 10^{-6}$ m/s; $H = 2.0 \times 10^{-3}$ m; Sc(B) = 800 ($\mathrm{Sc(B)}^{-1/3} = 0.10772$); $R_{\mathrm{TRUE\_B1}} = 0.25$; $R_{\mathrm{TRUE\_B2}} = 0.97$; $R_{\mathrm{TRUE\_Na}} = 0.94$.

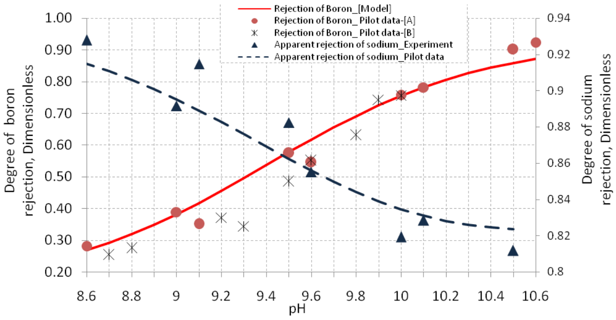


Fig. 7. The degree of apparent rejection (experimental and calculated) of boron and sodium (calculations are based on Eq. (13)). Input data: $D_{B1} = 2 \times 10^{-9}$ m$^2$/s; $D_{B2} = 1.5 \times 10^{-9}$ m$^2$/s; $V = 7.00 \times 10^{-6}$ m/s; $H = 2.00 \times 10^{-3}$ m; Sc(B) = 800; [$\mathrm{Sc(B)}^{-1/3} = 0.10772$]; $R_{\mathrm{TRUE\_B1}} = 0.25$; $R_{\mathrm{TRUE\_B2}} = 0.97$; $R_{\mathrm{TRUE\_Na}} = 0.94$.

## 6. Conclusions

The model for analysis of CP and the degree of rejection of boric acid, borate anions and monovalent cations at different pH has been developed. Unlike the prevalent published studies saying that the rejection of boron and sodium is not correlated, the proposed paper contains the model for conjugated behavior of the rejection for monovalent ions [Na], boric acid [$H_3BO_3$] and negatively charged borate [$B(OH)_4$]$^{-1}$. It was shown that the growth of pH is accompanied by the decrease of boric acid along with the increase of borate ions and monovalent cations at the membrane surface and in permeate. The calculations based on this model match the experimental data namely the degree of rejection of boron and monovalent cations (and conductivity) remains almost pH invariant within the acid and neutral range of pH (at pH < 8.6) whereas it changes sharply within alkaline domain.

This model can be used for estimation of transmembrane passage of both forms and it allows analyzing the permeate characteristics at different operating conditions. The model can be applied for quantitative analysis of the CP conjugated for monovalent ions, boric acid and borate ions. This model permits analyzing the influence of transmembrane flux, bulk concentration, physical properties, membrane rejection, channel geometry and pH on the degree of CP and surface concentration. This model can be applied when the multifold difference between the concentration of target component (e.g., boron) and the component that controls the level of salinity and ionic strength (e.g., monovalent ions) takes place, whereas the existing methods of analysis can be applicable mainly for monocomponent systems. The developed equations can be segmented into the algorithm for characterization of conjugated behavior of boron, borate anion and monovalent cations as well. The model can be built into the target function for solving different optimization problems. They can be used in the software for optimization of operating regimes or in the synthesis of optimal flow-diagram software for the process design and operation.

## Acknowledgment

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## Symbols

$C$ — Concentration, mol/m$^3$; kg/m$^3$
$C_{B\Sigma(\eta=1)}$ — Surface concentration
$D$ — Diffusivity coefficient, m$^2$/s
$f_{B1}$, $f_{B2}$ — The equilibrium fractions of boric acid and borate, respectively
$H$ — Half height of the channel, m
$R_{TRUE}$ — True degree of rejection, $R_{TRUE} = 1 - C_2/C_{1M}$, dimensionless
$R_{OBSERVED}$ — Observed degree of rejection, $R_{OBSERVED} = 1 - C_2/C_1$, dimensionless
$V$ — Transverse velocity, m/s
$z$, $Z$ — Transverse coordinate
$\alpha$ — Modulus of concentration polarization, dimensionless, $\alpha = (C_{B\Sigma(\eta=1)}/C_{B\Sigma(BULK)}) = C_{1M}/C_1$
$\delta_C$ — Thickness of diffusion layer, m
$\delta_W$ — Thickness of viscous layer, m
$\eta$ — Auxiliary coordinate (dimensionless thickness of diffusion layer)
Re — The Reynolds number, dimensionless, Re = $ud\varrho/\mu$
Sc — The Schmidt number, dimensionless, Sc = $\mu/D\varrho$

*Indexes*

B1 — Characteristics related to boric acid
B2 — Characteristics related to borate
B$\Sigma$ — Characteristics corresponding to overall amount of boric acid and borate
PERMEATE — Characteristics corresponding to permeate
BULK — Characteristics corresponding to bulk

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