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Low pressure RO for boron elimination: impact of pH on the degree of rejection of boron and monovalent ions

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ABSTRACT

Pilot data on the degree of rejection of boron, sodium and conductivity at different operating pH have been collected. The study is based on the analysis of the third stage of the RO pilot system for seawater desalination. The system was equipped by spiral element on a thin film composite polyamide membrane *Woongjin Chemical RE4040-BE*. The study covered the case when the target component (e.g. boron) was characterized by the lower value of concentration compared to the component (monovalent ions) that controls the level of salinity and ionic strength. It was shown that the degree of rejection of boron and the degree of rejection of conductivity remained almost pH invariant within the acid and neutral range of pH (at pH < 8.6), whereas it changed sharply within an alkaline domain. In particular the degree of boron rejection, being at the level ~30–33% at pH < 8.6 increased to 90%, and conductivity rejection revealed the opposite behavior: it decreased from 95 to 70%. It was shown that the transmembrane transport and rejection of monovalent ions, boric acid and deprotonated form of borate are closely conjugated. It was confirmed that the design of the process, analysis and modeling the rejection performance of boric acid and deprotonated borate must be coupled with quantitative estimation of the ions characterizing the level of ionic strength. It was revealed that the low-pressure RO membrane used in this study can be recommended for boron elimination at the stage of post-treatment in seawater desalination. Pilot data on the degree of observed rejection of boron, conductivity and monovalent ions vs. pH are attached.

Keywords: Desalination; Reverse osmosis; Post-treatment; Boron removal; Rejection of monovalent ions

1. Introduction

Boron is an important element affecting biosynthesis and cell metabolism; however, it is toxic and harmful when it exceeds certain critical limits. The main aspects of this problem were scrutinized in different studies [1–10]. The 1958, 1963, and 1971 WHO International Standards for Drinking Water did not contain any reference to boron. The first mention of boron was encountered in 1984, but it was not considered harmful at that time. Since then, it has been shown that boron can induce several harmful effects on animals in laboratory studies. Provisional guidelines for boron in drinking water were first introduced by WHO in 1993, where the guideline value, being

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equal to 0.3 mg/L, was recommended. This value was based on NOAEL (No Observed Adverse Effect Level). In the guidelines published in 1998, this value was increased to 0.5 mg/L. Today, 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 [11,12].

Technological aspects of this problem were included into many research programs and studied by different authors. It is well known that boron is effectively removed by thermal desalination, but the level of removal using RO appears to be insufficient. Since membrane technology is increasing, the boron-related issue is becoming challenging aspect. Engineering and economic characteristics are essential in design and analysis of the processes to be used in desalination technology. Different technological concepts, such as hybridization of the double-pass schemes with cascade and recycle were considered by Redondo et al. [13]. Research done by Taniguchi et al. [14] focused on combining SWRO, BWRO, and BSR. According to Faigon and Hefer [15] cascade design allows adjusting operating conditions. Since the membrane removal of boron is influenced by the factors such as, temperature, pressure, pH, feed flow rate, ionic strength, initial concentration, etc., the process design requires comprehensive input data. According to Oo and Song [3], 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. [4], Koseoglu et al. [5] and Cengeloglu et al. [6], the pressure growth tends to increase 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 [5]. Complexion reactions can also be used to increase boron rejection and this is achieved by the addition of Fe cations or other complex agents, which cause the formation of boron-containing complexes [7,8].

Existing methods of analysis, in spite of the difference in underlying assumptions, are based on the similar methods of evaluations of mass-transfer coefficients. They are based on the film theory and applicable for mono-component systems. An impact of pH on boron rejection has been considered by different authors, but examining existing published data have revealed some disputable statements, such as the lack of meaningful relationship between transport of boric acid or borate [16]. The study published by Tu et al. [17] comprises another questionable conclusion that the values of boron rejection were not correlated with sodium rejection, indicating that boron and sodium are rejected by different mechanisms. According to Hyung and Kim [1], boron rejection was dependent upon pH, while the rejection of other ionic species is not indicative to boron rejection.

These cases are characterized by multifold difference between the concentration of target component (boron) and the component (NaCl) that controls the level of salinity and ionic strength: the concentration of boron is lower than monovalent ions. Existing methods of process analysis and modeling concentration polarization cannot be applied in those cases. For that reason, the proposed study focuses on the relationship between rejection of monovalent ions and boron in protonated and deprotonated forms. In this study, the degree of rejection of boron, sodium, and conductivity has to be compared at different operating values of pH.

2. Experimental part

The pilot system was equipped by spiral element, *Woongjin Chemical RE4040-BE*, which was installed on the third-pass RO, where thin films of composite polyamide membranes were used (CSM RO Catalogue [19]). Total membrane area as 47.4 m^2 with six elements and with effective membrane area 7.9 m^2 per element were assembled within the pressure vessel. Normalized permeability ranged from 3.4 to 5.6 m^3/m^2-h-bars; operating pressure varied from 5.3 to 8.5 bars. Permeate after the second RO pass entered the third one after pH adjustment. The permeate, after the second RO pass, was characterized by the following values, conductivity within the range from 53 to 160 μS/cm, (salinity based on the evaluation of conductivity ranges from 3.27 E−4 to 1.12 E−4 mol/l): Concentration of sodium ranges from 3.7 E−4 to 1.52 E−4 mol/l (see Table 1 for experimental data). The pilot system was located at Al-Mirfa site (UAE). Simplified fragment of the flow diagram of the pilot plant was shown in Fig. 1.

The primary experimental curves of boron, salinity, and pH were received during the pilot study (see Fig. 2). ICP technique was used for analysis of samples.

3. Discussion of the results and comparison with existing published data

Boron exists in the form of boric acid [H_3BO_3] and deprotonated borate ion $[H_2BO_3]^{-1}$. Boric acid behaves as a weak Lewis acid according to the following equation.

Table 1
Experimental data on boron, sodium concentration, conductivity, pH, and the degree of rejection based on the reading from the low-pressure RO3 section of the pilot system. The pilot system was equipped by spiral element based on film composite polyamide with total membrane area–47.4 m^2. Membrane manufacturer: *Woongjin Chemical RE4040-BE* (see CSM RO Membrane Catalogue [19])

	pH	Boron			Conductivity			Salinity based on conductivity			Sodium concentration		
Number of samples	Feed for RO3	Feed for RO3 mol/l	Permeate of RO3 mol/l	Degree of rejection [–]	Feed for RO3 µs/cm	Permeate of RO3 µs/cm	Degree of rejection [–]	Feed for RO3 mol/l	Permeate of RO3 mol/l	Degree of rejection [–]	Feed for RO3 mol/l	Permeate of RO3 mol/l	Degree of rejection [–]
1	8.52	1.65E−04	1.19E−04	0.281	5.70E+01	1.50E+01	0.737	3.53E−04	8.27E−05	0.766	8.26E−04	6.09E−05	0.926
2	8.52	1.66E−04	1.18E−04	0.291	5.70E+01	1.50E+01	0.737	3.53E−04	8.27E−05	0.766	8.17E−04	5.65E−05	0.931
3	8.54	1.64E−04	1.19E−04	0.271	5.30E+01	1.10E+01	0.792	3.27E−04	5.90E−05	0.819	7.22E−04	6.09E−05	0.916
4	–	–	–	–	–	–	–	–	–	–	–	–	–
5	9.07	1.69E−04	1.13E−04	0.333	8.75E+01	1.57E+01	0.821	5.63E−04	8.69E−05	0.846	7.13E−04	7.83E−05	0.890
6	9.07	1.72E−04	1.13E−04	0.344	8.75E+01	1.57E+01	0.821	5.63E−04	8.69E−05	0.846	7.22E−04	6.96E−05	0.904
7	9.06	1.72E−04	1.11E−04	0.355	8.55E+01	1.75E+01	0.795	5.49E−04	9.78E−05	0.822	7.13E−04	6.96E−05	0.902
8	–	–	–	–	–	–	–	–	–	–	–	–	–
9	9.49	1.68E−04	8.24E−05	0.508	9.60E+01	2.43E+01	0.747	6.23E−04	1.40E−04	0.776	9.13E−04	1.35E−04	0.852
10	9.49	1.65E−04	8.15E−05	0.506	9.60E+01	2.43E+01	0.747	6.23E−04	1.40E−04	0.776	8.70E−04	1.35E−04	0.845
11	9.50	1.66E−04	8.89E−05	0.464	1.02E+02	2.64E+01	0.740	6.62E−04	1.53E−04	0.769	8.70E−04	1.52E−04	0.825
12	–	–	–	–	–	–	–	–	–	–	–	–	–
13	9.99	1.64E−04	4.44E−05	0.729	1.50E+02	4.78E+01	0.680	1.01E−03	2.92E−04	0.711	1.26E−03	2.22E−04	0.824
14	9.99	1.67E−04	4.17E−05	0.750	1.50E+02	4.78E+01	0.680	1.01E−03	2.92E−04	0.711	1.52E−03	2.22E−04	0.854
15	10.02	1.65E−04	4.17E−05	0.747	1.55E+02	5.09E+01	0.671	1.05E−03	3.13E−04	0.702	1.30E−03	2.30E−04	0.823
16	–	–	–	–	–	–	–	–	–	–	–	–	–
17	8.51	1.64E−04	1.29E−04	0.215	7.43E+01	6.22E+00	0.916	4.72E−04	3.18E−05	0.933	8.26E−04	4.78E−05	0.942
18	8.51	1.64E−04	1.25E−04	0.237	7.43E+01	6.22E+00	0.916	4.72E−04	3.18E−05	0.933	6.52E−04	4.78E−05	0.927
19	8.52	1.62E−04	1.24E−04	0.234	7.55E+01	7.15E+00	0.905	4.80E−04	3.69E−05	0.923	7.83E−04	5.65E−05	0.928
20	–	–	–	–	–	–	–	–	–	–	–	–	–
21	9.04	1.72E−04	1.13E−04	0.344	8.21E+01	1.30E+01	0.842	5.26E−04	7.08E−05	0.865	7.39E−04	9.13E−05	0.876
22	9.04	1.72E−04	1.12E−04	0.349	8.21E+01	1.30E+01	0.842	5.26E−04	7.08E−05	0.865	6.96E−04	9.13E−05	0.869
23	9.02	1.72E−04	1.18E−04	0.317	8.22E+01	1.35E+01	0.836	5.26E−04	7.38E−05	0.860	7.39E−04	8.70E−05	0.882
24	–	–	–	–	–	–	–	–	–	–	–	–	–
25	9.54	1.72E−04	8.89E−05	0.484	1.08E+02	2.78E+01	0.744	7.11E−04	1.62E−04	0.772	9.13E−04	1.48E−04	0.838
26	9.54	1.72E−04	8.70E−05	0.495	1.08E+02	2.78E+01	0.744	7.11E−04	1.62E−04	0.772	8.70E−04	1.48E−04	0.830
27	9.55	1.76E−04	8.24E−05	0.532	1.11E+02	3.03E+01	0.727	7.30E−04	1.78E−04	0.756	9.13E−04	1.65E−04	0.819
28	–	–	–	–	–	–	–	–	–	–	–	–	–
29	10.04	1.72E−04	3.89E−05	0.774	1.57E+02	4.78E+01	0.695	1.06E−03	2.92E−04	0.725	1.13E−03	2.22E−04	0.804
30	10.04	1.80E−04	3.89E−05	0.784	1.57E+02	4.78E+01	0.695	1.06E−03	2.92E−04	0.725	1.13E−03	2.22E−04	0.804
31	10.03	1.75E−04	4.26E−05	0.757	1.52E+02	4.99E+01	0.673	1.03E−03	3.06E−04	0.703	1.09E−03	2.26E−04	0.792
32	–	–	–	–	–	–	–	–	–	–	–	–	–
33	8.53	1.55E−04	1.09E−04	0.293	8.50E+01	6.70E+00	0.921	5.46E−04	3.44E−05	0.937	6.96E−04	5.22E−05	0.925

(*Continued*)

Table 1 (*Continued*)

Number of samples	pH Feed for RO3	Boron			Conductivity			Salinity based on conductivity			Sodium concentration		
		Feed for RO3 mol/l	Permeate of RO3 mol/l	Degree of rejection [–]	Feed for RO3 µs/cm	Permeate of RO3 µs/cm	Degree of rejection [–]	Feed for RO3 mol/l	Permeate of RO3 mol/l	Degree of rejection [–]	Feed for RO3 mol/l	Permeate of RO3 mol/l	Degree of rejection [–]
34	8.53	1.56E−04	1.19E-04	0.238	8.50E+01	6.70E+00	0.921	5.46E−04	3.44E−05	0.937	7.39E−04	5.65E−05	0.924
35	8.51	1.54E−04	1.16E−04	0.247	8.55E+01	6.40E+00	0.925	5.49E−04	3.28E−05	0.940	6.52E−04	4.78E−05	0.927
36	–	–	–	–	–	–	–	–	–	–	–	–	–
37	9.02	1.69E−04	1.04E−04	0.385	8.03E+01	1.29E+01	0.839	5.13E−04	7.02E−05	0.863	6.52E−04	8.26E−05	0.873
38	9.02	1.73E−04	1.01E−04	0.417	8.03E+01	1.29E+01	0.839	5.13E−04	7.02E−05	0.863	6.52E−04	8.26E−05	0.873
39	9.04	1.70E−04	1.05E−04	0.386	8.32E+01	1.49E+01	0.821	5.33E−04	8.21E−05	0.846	6.52E−04	9.57E−05	0.853
40	–	–	–	–	–	–	–	–	–	–	–	–	–
41	9.53	1.71E−04	7.13E−05	0.584	9.40E+01	2.35E+01	0.750	6.09E−04	1.35E−04	0.779	7.39E−04	1.30E−04	0.824
42	9.53	1.72E−04	7.04E−05	0.591	9.40E+01	2.35E+01	0.750	6.09E−04	1.35E−04	0.779	6.96E−04	1.22E−04	0.825
43	9.51	1.71E−04	7.69E−05	0.551	9.38E+01	2.53E+01	0.730	6.08E−04	1.46E−04	0.760	7.39E−04	1.35E−04	0.818
44	–	–	–	–	–	–	–	–	–	–	–	–	–
45	10.02	1.35E−04	2.50E−05	0.815	1.64E+02	4.56E+01	0.723	1.12E−03	2.77E−04	0.752	1.13E−03	1.83E−04	0.838
46	10.02	1.35E−04	2.50E−05	0.815	1.64E+02	4.56E+01	0.723	1.12E−03	2.77E−04	0.752	1.17E−03	1.83E−04	0.844
47	10.04	1.33E−04	2.59E−05	0.806	1.53E+02	4.72E+01	0.692	1.04E−03	2.88E−04	0.722	1.04E−03	1.91E−04	0.817
48	–	–	–	–	–	–	–	–	–	–	–	–	–
49	10.05	1.49E−04	3.24E−05	0.783	1.48E+02	4.36E+01	0.705	9.98E−04	2.64E−04	0.735	1.04E−03	2.00E−04	0.808
50	10.05	1.48E−04	3.24E−05	0.781	1.48E+02	4.36E+01	0.705	9.98E−04	2.64E−04	0.735	1.04E−03	2.00E−04	0.808
51	10.02	1.47E−04	3.70E−05	0.748	1.45E+02	4.55E+01	0.686	9.76E−04	2.77E−04	0.717	1.04E−03	2.13E−04	0.796
52	–	–	–	–	–	–	–	–	–	–	–	–	–
53	9.55	1.48E−04	6.76E−05	0.544	9.95E+01	2.57E+01	0.742	6.48E−04	1.49E−04	0.771	8.26E−04	1.39E−04	0.832
54	9.55	1.49E−04	6.85E−05	0.540	9.95E+01	2.57E+01	0.742	6.48E−04	1.49E−04	0.771	8.26E−04	1.39E−04	0.832
55	9.54	1.50E−04	7.59E−05	0.494	9.79E+01	2.78E+01	0.716	6.37E−04	1.62E−04	0.746	7.83E−04	1.52E−04	0.806
56	–	–	–	–	–	–	–	–	–	–	–	–	–
57	9.03	1.50E−04	8.98E−05	0.401	9.27E+01	1.25E+01	0.865	6.00E−04	6.78E−05	0.887	7.39E−04	7.39E−05	0.900
58	9.03	1.52E−04	8.70E−05	0.427	9.27E+01	1.25E+01	0.865	6.00E−04	6.78E−05	0.887	7.39E−04	8.26E−05	0.888
59	9.03	1.46E−04	8.98E−05	0.386	7.73E+01	1.34E+01	0.827	4.92E−04	7.32E−05	0.851	6.09E−04	8.26E−05	0.864
60	–	–	–	–	–	–	–	–	–	–	–	–	–
61	8.56	1.69E−04	1.16E−04	0.317	7.34E+01	6.23E+00	0.915	4.65E−04	3.18E−05	0.932	6.09E−04	3.96E−05	0.935
62	8.56	1.69E−04	1.17E−04	0.311	7.34E+01	6.23E+00	0.915	4.65E−04	3.18E−05	0.932	6.09E−04	3.70E−05	0.939
63	8.52	1.69E−04	1.17E−04	0.308	6.75E+01	6.00E+00	0.911	4.25E−04	3.05E−05	0.928	5.65E−04	3.87E−05	0.932

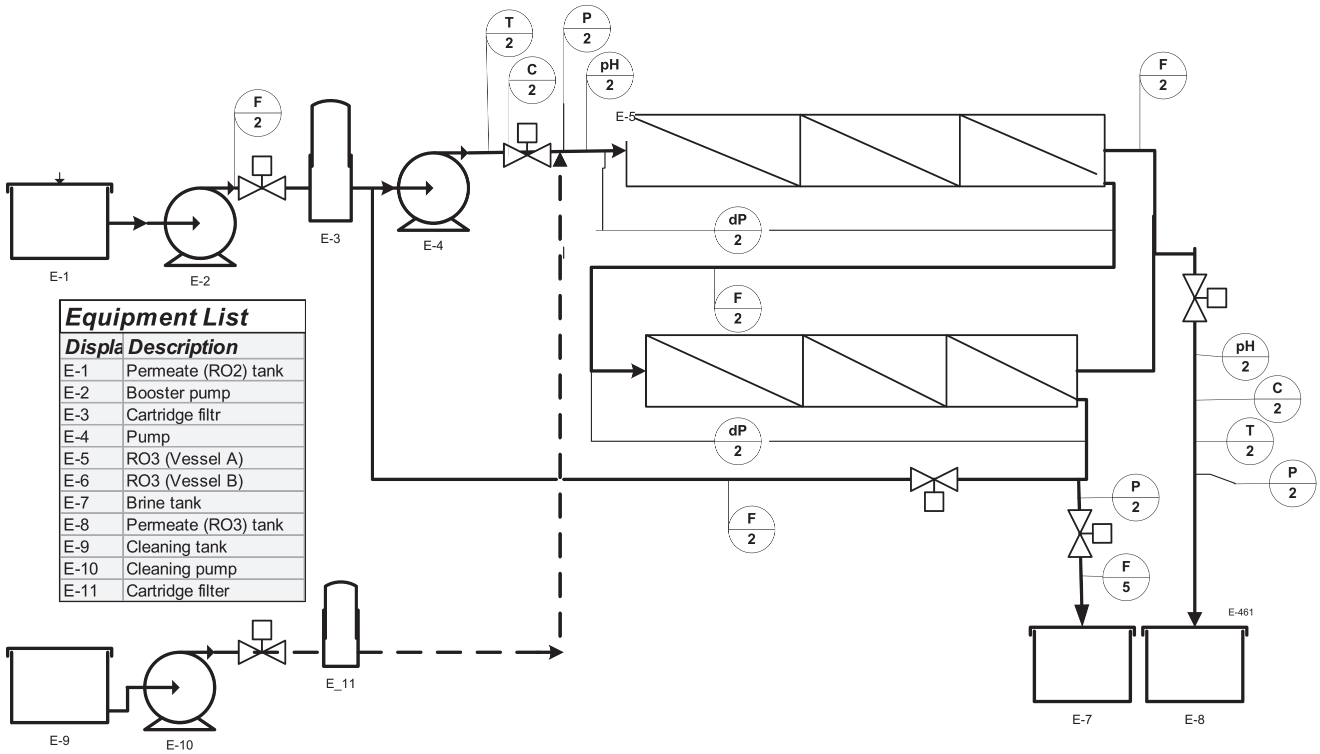

Fig. 1. The low-pressure section of the pilot plant. The system was equipped by spiral element based on film composite polyamide with total membrane area–47.4 m^2. Manufacturer: *Woongjin Chemical RE4040-BE* [19].

$$B(OH)_3 + H_2O = B(OH)_4^- + H^+ \quad (1)$$

The main components in feed solution are boric acid [H_3BO_3], deprotonated borate ion [H_2BO_3]$^{-1}$, and monovalent ions characterizing the level of ionic strength. As soon as the concentration of boron in seawater is around 4.8 mg/L [18], it was accepted that only mononuclear species $B(OH)_3$ and [$B(OH)_4$]$^{-1}$ are present in seawater. The distribution between two components, boric acid and borate anion, depends on the dissociation constant of boric acid (pKa), that in turn depends upon temperature, pressure, pH, and ionic strength. Boric acid is a weak acid with pKa1′ of 8.68 at 25℃ in high ionic strength solutions such as seawater with the first acid dissociation constant (p*K*a1) of 9.14 at 25℃ in a low ionic strength solution. Apparent first acid constant of boric acid *Ka1′* is defined as

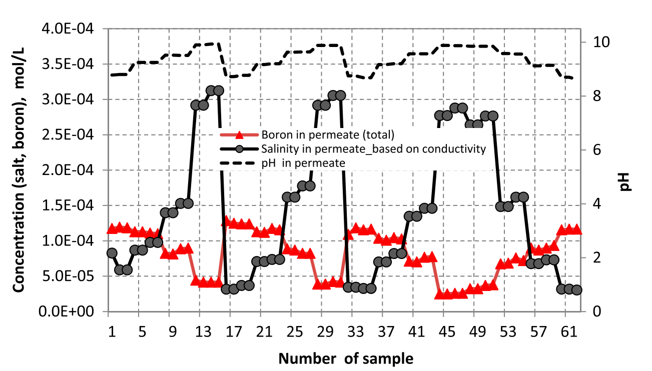

Fig. 2. Boron, salinity, and pH in the permeate of RO3 (Experiment).

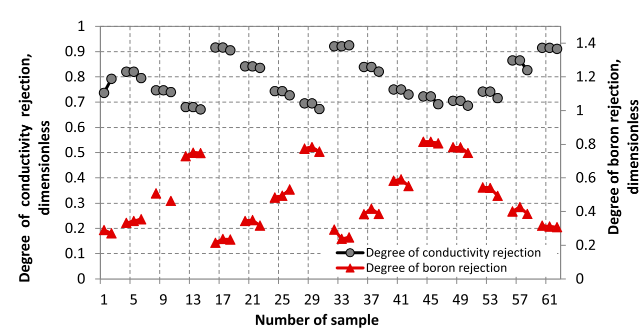

Fig. 3. The degree of observed rejection in terms of total boron concentration and conductivity.

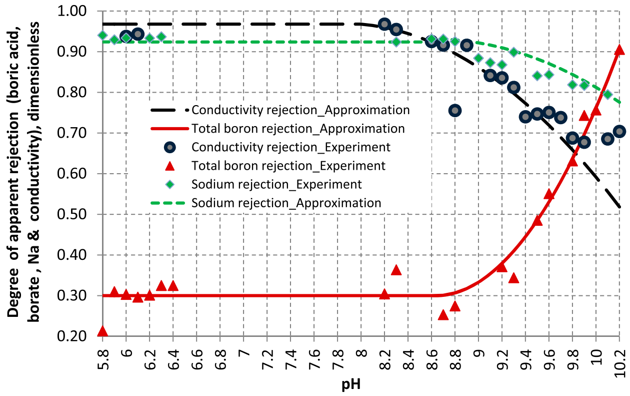

Fig. 4. The degree of observed rejection of boron and conductivity vs. pH.

$$K_{a1} = \frac{[H_2BO_3^{-1}]\{H^+\}}{[H_3BO_3]} \tag{2}$$

The equilibrium constant is defined using the concentration of boron species and the activity of proton. Since boric acid is weak, it mainly exists in uncharged form $[H_3BO_3]$ in the natural pH range. However, as pH increases, the fraction of negatively charged deprotonated borates $[H_2BO_3]^{-1}$ also increases and becomes dominant. The concentration of boric acid and deprotonated borate ion can be expressed as:

$$[H_3BO_3] = [B_\Sigma]\frac{\{H^+\}}{\{H^+\} + K_{a1}} \tag{3}$$

$$[H_2BO_3^{-1}] = [B_\Sigma]\frac{K_{a1}}{\{H^+\} + K_{a1}} \tag{4}$$

Since the second stage of dissociation was ignored, the total amount $[B_\Sigma]$ is equal the sum of boric acid and deprotonated borate.

Further processing of the experimental data (Figs. 2 and 3) gives us the relation between the degree of observed rejection of boron and salinity vs. pH (Fig. 4). 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 (expressed though conductivity and sodium concentration) remains pH independent in acidic and neutral range of pH (at $pH < 8.6$), while it changes sharply within alkaline domain. In particular, boron rejection (being at the level ~30–33% at $pH < 8.6$) goes up to 90%, while conductivity rejection reveals opposite behavior, namely it decreases from 95 to 70% (see Fig. 4).

An impact of pH on boron rejection has been scrutinized by many authors.

Experimental data by Hung et al. [2] show that boron rejection increases as feed pH increases due to increase of borate fraction. It increases from 70–85% at pH 7.5 to 90–98% at pH 10. Experimental data [15] demonstrated the growth of boron rejection from 78% (at pH 8) to 88% (at pH 8.55). According to the data submitted by Glueckstern and Priel [20], boron rejection goes from 75% (pH 7) to 97% (pH 11) and from 87% (pH 7) to 94% (pH 9) for low-pressure brackish water RO and seawater RO, respectively. Therefore, these published data demonstrate the similar shape of boron rejection, where the digital values are dependent on 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. Examination of existing published data on this topic, however, has revealed some disputable statements such as lack of meaningful relationship between transport of boric acid, borate, and monovalent ions [1,16,17].

4. Conclusions

Unlike the existing published data, this study indicates an obvious relationship between the rejection characteristics of monovalent ions [Na], boric acid $[H_3BO_3]$, and deprotonated form of borate $[H_2BO_3]^{-1}$ Having analyzed the pilot data and profile behavior (see Fig. 4), the following conclusions have be drawn:

(1) The degree of rejection of boron and salinity (expressed though conductivity and sodium concentration) remains pH invariant in acidic and neutral range of pH (at $pH < 8.6$), while it changes sharply within the alkaline domain. In particular, boron rejection (being at the level ~30–33% at $pH < 8.6$) goes up to 90%, whereas conductivity rejection reveals opposite behavior, namely it goes down from 95 to 70%.

(2) It can be concluded that a low-pressure RO process based on membrane *Woongjin Chemical RE4040-BE* can be recommended for boron elimination on the stage of post-treatment.

Since the considered case is characterized by 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), it should be noted that existing methods suitable for analysis of monocomponent concentration polarization cannot be applied in this case. The data received in this pilot study can be used for further quantitative analysis of concentration polarization conjugated for monovalent ions, boric acid, and deprotonated form of borate.

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