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Rejection of small solutes by reverse osmosis membranes for water reuse applications: a pilot-scale study

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Rejection of small solutes by reverse osmosis membranes for water reuse applications: a pilot-scale study

Abstract

N-nitrosamines and boron are small solutes of particular concern during water recycling applications. Here, we evaluated the rejection of seven N-nitrosamines and boron under a range of operating conditions and feed solution characteristics. The evaluationwas conducted using a pilot-scale reverse osmosis (RO) system to appropriately simulate hydrodynamic conditions of full-scale RO installations. The rejection of seven N-nitrosamines by the pilot RO system varied significantly in the range from 31 to 94%, and rejection increased in the increasing order of their molecular weight. Rejection values obtained from this pilot-scale study were lower than those previously reported in laboratory-scale studies. These discrepancieswere attributed to a difference in RO system operating condition (i.e. recovery) between the pilot-scale study (25%) and laboratory-scale study (b0.1%). Nevertheless, rejection data reported here validate the recent findings fromlaboratory-scale studies with respect to the impact of permeate flux, feed temperature and feed pH on separation efficiencies of N-nitrosamines. Data obtained from this pilot-scale study also validate the strong correlation between boron and NDMA rejection at or below pH 8 regardless of operating conditions and feed solution characteristics. The results suggest that boron rejection can be used as a surrogate for NDMA rejection in full-scale RO installations.

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Abstract

N-nitrosamines and boron are small solutes of particular concern during water recycling applications. Here, we evaluated the rejection of seven N-nitrosamines and boron under a range of operating conditions and feed solution characteristics. The evaluation was conducted using a pilot-scale reverse osmosis (RO) system to appropriately simulate hydrodynamic conditions of full-scale RO installations. The rejection of seven N-nitrosamines by the pilot RO system varied significantly in the range from 31 to 94%, and rejection increased in the increasing order of their molecular weight. Rejection values obtained from this pilot-scale study were lower than those previously reported in laboratory-scale studies. These discrepancies were attributed to a difference in RO system operating condition (i.e. recovery) between the pilot-scale study (25%) and laboratory-scale study (<0.1%). Nevertheless, rejection data reported here validate recent findings from laboratory-scale studies with respect to the impact of permeate flux, feed temperature and feed pH on separation efficiencies of N-nitrosamines. Data obtained from this pilot-scale study also validate the strong correlation between boron and NDMA rejection at or below pH 8 regardless of operating conditions and feed solution characteristics. The results suggest that boron rejection can be used as a surrogate for NDMA rejection in full-scale RO installations.

Keywords: Boron; N-nitrosodimethylamine (NDMA); N-nitrosamines; spiral wound membrane elements; reverse osmosis (RO); water reuse.

1. Introduction

Water reuse is an increasingly important strategy in many parts of the world to supplement drinking water resources [1]. Municipal wastewater is abundantly available in the urban environment where water demand is high. In a planned indirect potable water reuse scheme, secondary or tertiary treated wastewater is reclaimed using advanced treatment technologies to produce high quality water. Reclaimed water can potentially contain pathogenic agents and traces of organic contaminants which may cause adverse impact on human health. Thus, water reuse is subjected to a stringent set of regulations for public health protection. Among a series of water treatment processes in a water reclamation plant, reverse osmosis (RO) filtration serves as an efficient barrier for removing most trace organic chemicals [2]. Nevertheless, a small compound, N-nitrosodimethylamine (NDMA) has been detected in RO permeate at a higher concentration than its regulatory value (i.e. 10 ng/L) [3, 4]. Moreover, NDMA rejection evaluated in full-scale plants has been reported to be variable and therefore difficult to predict (10-55%) [3-6].

NDMA is frequently detected in secondary treated wastewater [7]. In addition, NDMA concentration can increase during chloramination, which is commonly employed prior to RO filtration. The other N-nitrosamines that can occur in treated wastewater include N-nitrosomethylethylamine (NMEA), N-nitrosopyrrolidine (NPYR), N-nitrosodiethylamine (NDEA), N-nitrosopiperidine (NPIP), N-nitrosomorpholine (NMOR), N-nitrosodipropylamine (NDPA), N-nitrosodi-n-butylamine (NDBA) [8-10]. NDMA and some other N-nitrosamines have been classified as probable human carcinogens by the US EPA [11]. The permeation of NDMA and other N-nitrosamines through RO membranes has been investigated in many laboratory-scale studies using small membrane coupons [12-16]. These laboratory-scale studies suggest that NDMA rejection by RO membranes is affected by changes in operating conditions (e.g. feed temperature and permeate flux). Nevertheless, findings obtained from these laboratory-scale studies employing flat-sheet membrane specimen cannot be directly used to accurately simulate hydrodynamic conditions of full-scale RO system. Despite the differences between small membrane coupons and spiral wound membrane elements, investigations evaluating the rejection of N-nitrosamines using pilot-scale RO systems remain scarce [5] and no previous pilot-scale RO studies have been conducted to mechanistically elucidate the impact of various operating conditions on N-nitrosamine rejection.

Regularly monitoring N-nitrosamines in aqueous solution at parts per trillion levels is another challenge for most water authorities. Analysis of N-nitrosamine concentrations is generally carried out using an analytical method which comprises solid phase extraction followed by gas chromatography or high pressure liquid chromatography with tandem mass spectrometry detector [17]. The analysis of N-nitrosamines at parts per trillion levels is expensive and not always readily available. Thus, any alternative indicators that simulate the rejection of N-nitrosamines, in particular NDMA, by reverse osmosis could be very useful for monitoring purposes. A recent laboratory-scale study by Tu et al. [18] reported that boron rejections by RO membranes under a range of a specific operating condition (i.e. permeate flux or feed temperature) correlate with NDMA rejection. The similarity in the rejection behaviour between boron and NDMA is very likely due to their similar physicochemical characteristics. In fact, both boron (as a boric acid) and NDMA molecules are small (62 g/mol and 74 g/mol, respectively) and uncharged under operating conditions representative of RO installations (i.e. pH 6-8) [19, 20]. It is noteworthy that boron in reclaimed water is also of great concern in places where reclaimed water is used for irrigation. Unlike NDMA, boron is abundantly detected in treated wastewater at 0.3 - 4 mg/L [21] and several commercially available online boron analysers have parts per billion to trillion sensitivity. Thus, boron rejection can potentially be used as a surrogate to estimate NDMA rejection. The use of boron as an NDMA surrogate will not replace compliance monitoring of NDMA in the final product water. However, it will facilitate better system design and reduce the number of analyses, and hence the cost associated with ongoing NDMA monitoring.

To date, the potential use of boron rejection as a surrogate of NDMA rejection has only been demonstrated by laboratory scale experiments using a small membrane sample. The hydraulic and hydrodynamic conditions as well as operating conditions (i.e. water recovery) between a lab-scale RO set-up using small flat sheet membrane coupons and pilot- and full-scale installations using spiral wound membrane elements can differ significantly. Thus, it is warranted to validate whether the observed correlation between boron and NDMA rejections also applies to RO systems employing spiral-wound elements. The aim of this study was to demonstrate the rejection of N-nitrosamines and boron using a pilot-scale RO system simulating hydrodynamic conditions of full-scale RO installations under a range of different operating conditions (i.e. permeate flux, feed pH and feed temperature).

2. Materials and methods

2.1. Chemicals

Analytical grade N-nitrosamines and boric acid (Table 1) were purchased from Sigma-Aldrich (St Louis, MO, USA). These N-nitrosamines include NDMA, NMEA, NPYR, NDEA, NPIP, NMOR and NDPA. A stock solution was prepared in pure methanol at 10 mg/L of each N-nitrosamine. Deuterated N-nitrosamines used for analysis were supplied by CDN isotopes (Pointe-Claire, Quebec, Canada) and these chemicals include N-nitrosodimethylamine-D6, N-nitrosomethylethylamine-D3, N-nitrosopyrrolidine-D8, N-nitrosodiethylamine-D10, N-nitrosopiperidine-D10, N-nitrosomorpholine-D8 and N-nitrosodipropylamine-D14. A surrogate stock solution of 100 μg/L of each deuterated N-nitrosamine was prepared in pure methanol. These stock solutions were kept at -18 °C in the dark and were used within 1 month of preparation. Analytical grade NaCl, $CaCl_2$ and $NaHCO_3$ were obtained from Ajax Finechem (Taren Point, NSW, Australia) and used as the background electrolytes during the filtration experiments.

[Table1]

2.2. Pilot-scale filtration system and experimental protocols

The pilot-scale RO filtration system used for this investigation has a permeate design flow of 0.72 m^3/h (Figure 1). The system consists of three 4 inch glass-fibre pressure vessels, a feed reservoir, a centrifugal feed pump (CRN 3-25, Grundfos, Bjerringbro, Denmark), and variable flow meters. Pipe lines in the feed stream were made of stainless steel and those in the permeate stream were made of PVC. Each pressure vessel could hold one RO membrane element (4 inch × 40 inch). Feed solution temperature was controlled in the feed reservoir using stainless steel heat exchanging pipes connected to a chillier unit (Aqua Cooler S360PD-CT, Chester Hill, NSW, Australia). The TFC-HR membrane (TFC 4040HR, Koch Membrane Systems, San Diego, CA, USA) used in the study is a composite polyamide RO membrane designed for brackish water treatment with a membrane area of 7.9 m^2/element.

Prior to the pilot-scale filtration experiment, 200 L of RO feed solution was prepared in the feed tank. RO feed solutions used for this investigation were conditioned to obtain 20 mM NaCl, 1 mM $CaCl_2$ and 1 mM $NaHCO_3$ in deionised water or actual microfiltration (MF) permeate which was collected from a local full-scale water recycling plant in Australia. The TOC, conductivity, and pH of the MF permeate were 9.6 mg/L, 0.91 mS/cm, and 6.9, respectively. In addition, N-nitrosamines and boric acid were dosed into the feed solution at approximately 250 ng/L of each N-nitrosamine and 5 mg/L of boron. Subsequently, the pilot RO system was continuously operated under standard conditions (permeate flux 20 L/m^2h, feed temperature 20 °C, feed pH 8.0) for 15 hours prior to any sampling events. The concentrate and permeate solutions were recirculated into the feed tank. The overall system recovery was maintained at 25% throughout the experiments. For N-nitrosamine analysis, 200 mL of RO feed and permeate samples were collected in amber glass bottles and the N-nitrosamine surrogate stock solution was dosed into each sample to obtain 50 ng/L of each isotope labelled N-nitrosamine. From the same sampling point, 20 mL sample was also collected in plastic bottles for the analysis of boron and sodium ions. Experiments with variable operating conditions started with low permeate flux of 10 L/m^2h, which was increased stepwise to 30 L/m^2h. When operating conditions were changed, sampling was conducted after at least 1 hour of system operation. Experiments with variable feed temperature were first evaluated under low feed temperature (10 °C) and the feed temperature was incrementally increased to 35 °C. Experiments with variable feed pH were conducted by adjusting feed pH (5 - 9.5) with a dose of either HCl or NaOH.

[Figure 1]

2.3. Analytical technique

N-nitrosamine concentrations in the RO feed and permeate were determined using an analytical method reported by McDonald et al. [17] and a brief summary is provided here. N-nitrosamines in each sample were first extracted by solid phase extraction method using Supelclean^{TM} Coconut Charcoal SPE cartridges (Supelco, St Louis, MO, USA). The determination of N-nitrosamine concentrations were carried out using an Agilent 7890A gas chromatograph coupled with an Agilent 7000B triple quadrupole mass spectrometer. In brief, boron concentration was determined using an inductively-coupled plasma-mass spectrometer (7500Cs, Agilent Technologies, Wilmington, DE, USA). The details of boron and sodium ion analysis have been described by a previous study [22]. Calibration of the analysis was conducted using a Merck ICP multi-element standard solution.

3. Results and discussion

3.1. *N-nitrosamine and boron rejection*

Transport of neutral solutes across RO membranes involves the partitioning of the solute into the membrane active skin layer followed by diffusion to the permeate side. In principal, solutes larger than free-volume holes of the membrane active skin layer cannot enter the membrane matrix. Once smaller solutes have entered the membrane polymeric matrix, their transport across the active skin layer is dependent on a chemical potential but is essentially independent of the feed pressure [23]. The diffusivity within the membrane matrix can decrease with increasing the solute size (i.e. Stokes radius) as suggested by Kiso et al. [24]. This indicates that the diffusivity of large solutes within the membrane is more likely to be slower than that of small solutes. In fact, N-nitrosamine rejection by RO membranes is mostly governed by size exclusion as reported in previous laboratory-scale studies [12, 25], suggesting that smaller N-nitrosamines permeate through RO membranes more easily than larger N-nitrosamines.

As expected, the rejection of N-nitrosamines by the pilot RO system varied significantly in the range from 31 to 94% and rejection generally increased in the increasing order of the molecular weight of N-nitrosamines with the exception of NMOR (Figure 2). The rejection values of N-nitrosamines and boron observed in this pilot-scale study differed from those reported in previous laboratory-scale studies [12, 18]. For example, the rejections of NDMA and boron obtained in this pilot-scale study (31 and 27%, respectively) were lower than those reported in the laboratory-scale studies (46% and 57%, respectively) (Figure 2). Likewise, the rejections of the other N-nitrosamines by spiral wound membranes (pilot-scale study) were 2-19% in rejection value lower than those by flat sheet membranes. The variation in rejection between the laboratory- and pilot-scale systems was negligible for higher molecular weight N-nitrosamines (i.e. NPIP and NDPA) with the exception of NMOR (Figure 2). Results reported here suggest that there can be a large variation between the rejections of these low molecular weight compounds obtained from laboratory- and pilot-scale experiments.

It is noteworthy that the pilot-scale RO system was operated under a similar condition (i.e. 20 L/m^{2}h average permeate flux and 20 °C feed temperature) and with the same feed water (i.e. 20 mM NaCl, 1 mM $NaHCO_3$, 1 mM $CaCl_2$, and feed pH 8.0) compared to the previous laboratory-scale studies [12, 18]. A notable difference in operating conditions between the pilot- and laboratory-scale studies was RO system recovery (25 and <0.1%, respectively). In general, the rejection of trace organic chemicals by nanofiltration/RO membranes decreases with increasing system recovery [26, 27]. When increasing system recovery solute concentrations increase toward the exit of the feed stream due to the accumulation of rejected solutes [27]. Moreover, with increasing system recovery a reduction in cross flow velocity and subsequent increase in concentration polarisation occur toward the tail-end elements of the latest stage of a membrane skid. The difference in membrane geometry between the laboratory- and pilot-scale systems can also be another factor. Although clarifying the contribution of these three factors described above is beyond the scope of this study, one or several of these factors are likely to have resulted in different solute rejections between these two studies.

[Figure 2]

3.2. Effects of RO feed solution chemistry and operating conditions

3.2.1. Permeate flux

Increasing permeate flux resulted in an increase in the rejection of N-nitrosamines, sodium ion and boron (Figure 3a). The observed solution rejection increase can be attributed to the larger increase in water compared to solute transport through RO membranes as can be described by the irreversible thermodynamic model [23]. Of the seven N-nitrosamines used in this investigation, three N-nitrosamines with the lowest molecular weight (i.e. NDMA and NMEA, NPYR) exhibited a significant increase in rejection (approximately 20%) when permeate flux increased from 10 to 30 L/m^{2}h (Figure 3a). Similarly, boron rejection also increased by approximately 24% for the same change in permeate flux (Figure 3b). Changes in NDMA and boron rejection obtained from this pilot-scale study were similar to those reported in previous laboratory-scale studies [12, 18] where the rejection of NDMA and boron increased by approximately 21 and 28%, respectively. The increase in the rejection of the other N-nitrosamines obtained from this pilot-scale study (2-11% in rejection) was smaller than that of the two smallest compounds (i.e. boron and NDMA). The rejection of sodium ion was high (96-98%) despite its small molecule size (molecular weight 23 g/mol) (Figure 3b). This is because sodium ions are present in hydrate form (Na^+-$6H_2O$) in a pH 8 aqueous solution, and electrostatic interactions occurring between sodium ion and membranes enhance the rejection of sodium ions.

Full-scale RO systems are usually operated at an overall permeate flux of approximately 18-20 L/m^{2}h [28]. Nevertheless, the permeate flux range of 10 to 30 L/m^{2}h was selected to account for the distribution of local permeate flux within the system. It is noteworthy that the local permeate flux of each of the three elements differed slightly from average (system) flux (Table 2). In a full-scale RO plant, each pressure vessel usually holds 6 or 7 membrane elements. Consequently, distribution of the local permeate flux within a pressure vessel can be larger than that shown Table 2. This is demonstrated by computer simulation using the IMSDesign software from Hydranautics (Table 3).

[Figure 3]

[Table 2]

[Table 3]

3.2.2. Feed temperature

Changes in feed temperature considerably affected the rejection of all compounds selected in this investigation. For example, when feed temperature increased from 10 to 35 °C, the two smallest compounds (i.e. NDMA and boron) exhibited a considerable drop in rejection from approximately 40% to negligible level (< 10%) (Figure 4a,b). Other N-nitrosamines also exhibited a moderate to large decrease in rejection with increasing temperature, but the impact was less pronounced with increasing molecular weight. The impact of temperature on rejection observed from this pilot-scale study was generally similar to those reported in previous laboratory-scale studies [12, 18].

The effects of feed temperature on the rejection of N-nitrosamines and boron were also investigated using the MF permeate. The rejection of NDMA and boron in the MF permeate were found to be approximately 20% higher than those in the clean solution for all feed temperatures evaluated (Figures 4 and 5). It is noted that the difference in conductivity (or ionic strength) between the clean (synthetic) solution and MF permeate (approximately 0.9 and 2.4 mS/cm, respectively) is not expected to assert any considerable influence on NDMA and boron rejection as reported in previous studies [12, 22]. On the other hand, the TOC concentration of the MF permeate was 9.6 mg/L. The presence of bulk organic matter in the feed or on membrane surface might explain for the better NDMA and boron rejection obtained from the MF permeate compared to those from a clean (synthetic) solution. In fact, a previous laboratory-scale study [13] has reported a considerable increase in NDMA rejection (from 34% to 73%) due to fouling which was simulated using sand-filtered tertiary wastewater.

The decreased rejection can occur because the active skin layer of RO membranes acts as a separation layer and its physicochemical characteristics can change by modifying feed solution characteristics including feed temperature [29]. In addition, permeability coefficient of solutes increases when feed temperature increases which results in a decrease in solute rejection [30]. The results obtained here suggest for wastewater in cold climates (e.g. 10 ºC), the rejection of NDMA and boron can be expected to be moderate (approximately 40%) and the rejection of the other N-nitrosamines can be very high (>80%). On the other hand, in warm locations where wastewater temperature might reach 30-35 ºC, N-nitrosamines may exhibit a large variation in rejection and NDMA and boron possibly resulting in negligible rejection.

[Figure 4]

[Figure 5]

3.2.3. Feed pH

Changes in feed pH did not significantly affect the rejection of N-nitrosamines. For example, a large shift in feed pH (from 5 to 9.5) resulted in a small increase in NDMA rejection (from 29 to 38%) (Figure 6a). The impact of the changes in feed pH on the rejection of the other N-nitrosamines was negligible which is consistent with that obtained from a previous laboratory-scale study [12]. Nevertheless, a different rejection trend was observed for boron at high pH. Boron rejection in feed solution of pH 9.5 reached as high as 72% while boron rejection at the other pH (5-8) was as low as 27-32% (Figure 6b). As the solution pH increases beyond the pK_a value (9.2) of boric acid, boric acid (which is neutral and is the primary form of boron in the aqueous phase) is deprotonated to form tetrahydroxyborate ($B(OH_4)^-$) which is a negatively charged species [18]. Rejection of the negatively charged species is governed by both size exclusion and electrostatic interaction mechanisms [31], and thus boron rejection at above pH 9.2 can be significantly higher than that at pH 8 and below (Figure 6b). It is noteworthy that full-scale RO system does not undergo such a high feed pH (9.5) under normal operation conditions, because full-scale systems are commonly operated at feed pH of 6-8 to minimise scale formation on membrane surfaces [28].

[Figure 6]

3.3. *Correlation between boron and NDMA rejection*

The reported results show that the rejection behaviour of NDMA is similar to that of boron. In fact, a strong correlation ($R^2 = 0.90$) between boron and NDMA rejections at various operating conditions was observed (Figure 7). Although boron rejection value was generally similar to NDMA rejection, a large variation in the rejection of these solutes was observed in the clean solution within the range of low rejection values (<30%) obtained under different operating conditions. For example, when permeate flux was 10 L/m^2h boron rejection (8%) was considerably lower than NDMA rejection (21%) (Figure 7). In contrast, boron rejection (11%) was higher than NDMA rejection (4%) when feed temperature was 35 °C. Separation performance of neutral and small compounds are dominated partly by the size of passage within the active skin layer of RO membranes (also referred as free-volume hole-radius) [32], and the conformation of polymer matrix within the active skin layer can vary depending on operating conditions (i.e. feed temperature and pH) [29, 33]. Because there is discernible difference in molecular size (e.g. maximum molecular length and molecular width) between boron and NDMA (Table 1), the variation in these rejections can be attributed to the difference in their subjectivity to changes in the conformation of polymer matrix.

[Figure 7]

4. Conclusions

We examined the rejection of N-nitrosamines and boron using a pilot-scale reverse osmosis system. There were discernible differences between the rejection values obtained from this pilot-scale study and those previously reported in laboratory-scale studies. These discrepancies can be attributed to the high recovery under which the pilot-scale RO system was operated. The rejection of N-nitrosamines and boron by a spiral wound RO membrane system could vary considerably as a function of permeate flux and feed solution temperature. The variation was more significant for small solutes (i.e. boron and low molecular weight N-nitrosamines such as NDMA, NMEA and NPYR). When permeate flux increased from 10 to 30 L/m^{2}h, three lowest molecular weight N-nitrosamines (i.e. NDMA and NMEA, NPYR) in the clean solution exhibited a significant increase in rejection (approximately 20% in rejection value). The two smallest compounds (i.e. NDMA and boron) in the clean solution exhibited a considerable drop in rejection from approximately 40% to negligible level when feed temperature increased from 10 to 35 ºC. The rejection of NDMA and boron in the MF permeate were found to be approximately 20% higher than those in the clean solution under any feed temperature tested here (10-35 ºC). Changes in feed pH (5-9.5) revealed a negligible impact on the rejection of N-nitrosamines and sodium ions in the clean solution. Nevertheless, boron exhibited a very high rejection (72%) at pH 9.5 while boron rejection at the other pH (5-8) were low (27-32%). The changes in small solute rejection in response to variation in operating parameters (i.e. permeate flux, temperature, and pH) obtained from this pilot-scale study were similar to those from previous laboratory-scale studies. As a result, a strong correlation between the rejections of NDMA and boron was observed. These results confirm that boron rejection can be used as a surrogate for NDMA rejection using spiral wound RO membrane elements. Additional work is recommended to confirm the correlation between NDMA and boron rejections at full-scale plants.

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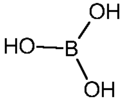
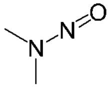
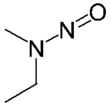
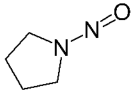
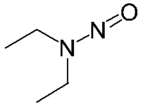
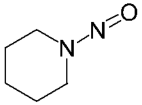
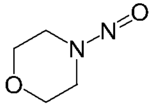
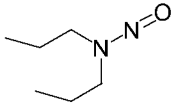
Table 1: Physicochemical characteristics of the selected N-nitrosamines and boric acid.

Name	B (Boric acid)	NDMA	NMEA	NPYR	NDEA	NPIP	NMOR	NDPA
Structure								
Molecular Formula	$B(OH)_3$	$C_2H_6N_2O$	$C_3H_8N_2O$	$C_4H_8N_2O$	$C_4H_{10}N_2O$	$C_5H_{10}N_2O$	$C_4H_8N_2O_2$	$C_6H_{14}N_2O$
Molecular weight [g/mol]	61.83	74.05	88.06	100.06	102.08	114.08	116.06	130.11
Maximum molecular length [a] [nm]	0.621	0.705	0.832	0.761	0.851	0.773	0.765	1.035
Molecular width [a] [nm]	0.257	0.270	0.306	0.318	0.322	0.325	0.317	0.365
Log K_{ow} [b]	-0.62	-0.50	0.01	-0.09	0.52	0.44	-0.81	1.54

[a] Calculated using Molecular Modeling Pro (ChemSW Inc., Fairfield, CA, USA). The maximum molecular length represents the distance between the two most distant atoms in the molecule. The molecular width represents a half of the square root of the area of the rectangle which encloses the projection of the molecule [22].

[b] ACD/PhysChem Suite software (Advanced Chemistry Development, Inc., Ontario, Canada).

Table 2: Measured local flux of each RO element.

Average system flux [L/m^{2}h]	Local flux [a] [L/m^{2}h]		
	Lead-element	2nd element	Tail-element
10	11	10	9
20	23	19	18
30	35	29	27

[a] Experimental conditions are as described in Figure 3.

Table 3: Simulated local flux of lead- and tail-element among seven RO elements.

Average system flux [L/m^{2}h]	Local flux [a] [L/m^{2}h]	
	Lead-element	Tail-element
10	13	7
20	25	16
30	37	24

[a] Calculated using a software (IMSDesign) provided by Hydranautics (20 mM NaCl, 1 mM $NaHCO_3$, 1 mM $CaCl_2$, feed pH 8.0, feed temperature 20 ºC, system recovery 50%).

LIST OF FIGURES

Figure 1: (a) A picture of the pilot system; and (b) schematic diagram of the pilot system.

Figure 2: Rejection of N-nitrosamines and boron in the clean solution by the TFC-HR membrane using the pilot RO system as a function of their molecular weight (20 mM NaCl, 1 mM $NaHCO_3$, 1 mM $CaCl_2$, average permeate flux 20 L/m^{2}h, feed pH 8.0 ± 0.1, feed temperature 20.0 ± 0.1 ºC). Values reported here are the average and ranges of duplicates results. Open symbols (○ and □) represent the rejections of N-nitrosamines and boron by the TFC-HR membrane using a laboratory-scale RO filtration setup [12, 18] (20 mM NaCl, 1 mM $NaHCO_3$, 1 mM $CaCl_2$, permeate flux 20 L/m^{2}h, feed pH 8.0 ± 0.1, feed temperature 20.0 ± 0.1 ºC).

Figure 3: (a) N-nitrosamine rejection and (b) inorganic solute rejection in the clean solution by the pilot RO system using the TFC-HR membranes as a function of permeate flux (20 mM NaCl, 1 mM $NaHCO_3$, 1 mM $CaCl_2$, feed pH 8.0 ± 0.1, feed temperature 20.0 ± 0.1 ºC). N-nitrosamine rejection values reported here are the average and ranges of two replicate experiments.

Figure 4: (a) N-nitrosamine rejection and (b) inorganic solute rejection in the clean solution by the pilot RO system using the TFC-HR membranes as a function of feed temperature (20 mM NaCl, 1 mM $NaHCO_3$, 1 mM $CaCl_2$, permeate flux 20 L/m^{2}h, feed pH 8.0 ± 0.1). N-nitrosamine rejection values reported here are the average and ranges of two replicate experiments.

Figure 5: (a) N-nitrosamine rejection and (b) inorganic solute rejection in the MF permeate by the pilot RO system using the TFC-HR membranes as a function of feed temperature (permeate flux 20 L/m^{2}h, feed pH 6.9 ± 0.1). N-nitrosamine rejection values reported here are the average and ranges of two replicate experiments.

Figure 6: (a) N-nitrosamine rejection and (b) inorganic solute rejection in the clean solution by the pilot RO system using the TFC-HR membranes as a function of feed pH (20 mM NaCl, 1 mM $NaHCO_3$, 1 mM $CaCl_2$, permeate flux 20 L/m^{2}h, feed temperature 20.0 ± 0.1 ºC). N-nitrosamine rejection values reported here are the average and ranges of two replicate experiments.

Figure 7: Correlation between rejection of boron and NDMA in the (a) clean solution and (b) MF permeate by the pilot RO system using the TFC-HR membranes. Results using the TFC-HR membrane were obtained from Figure 3-6 with the exclusion of data of feed pH 9.5.

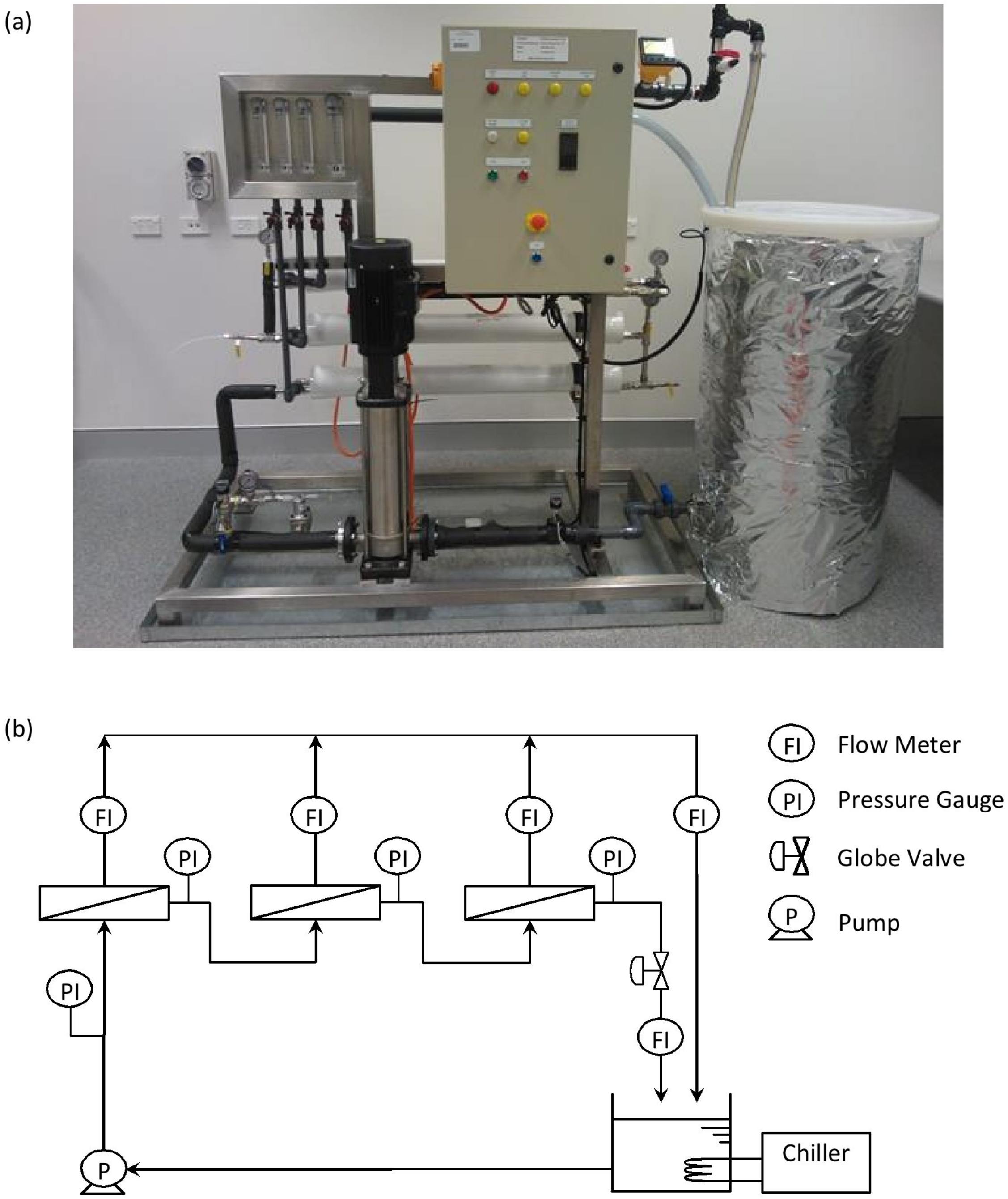

(a)
(b)
FI
PI
FI
PI
FI
PI
FI
PI
FI
P
Chiller
FI Flow Meter
PI Pressure Gauge
Globe Valve
P Pump

Figure 1

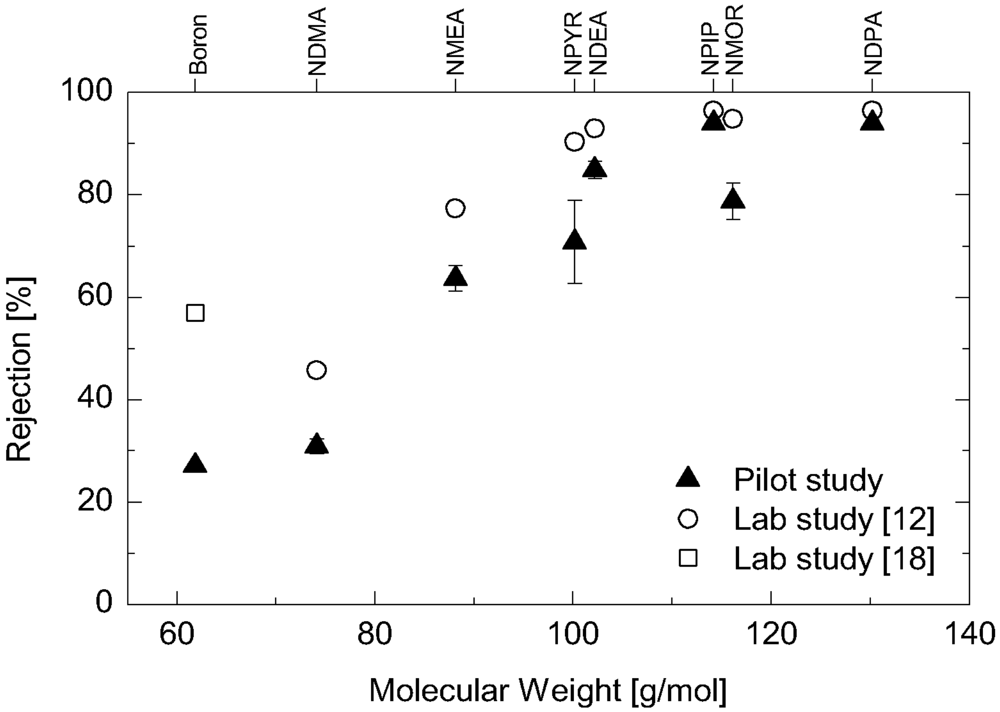

Boron
NDMA
NMEA
NPYR
NDEA
NPIP
NMOR
NDPA
100
80
60
40
20
0
Rejection [%]
60
80
100
120
140
Molecular Weight [g/mol]
▲ Pilot study
○ Lab study [12]
□ Lab study [18]

Figure 2

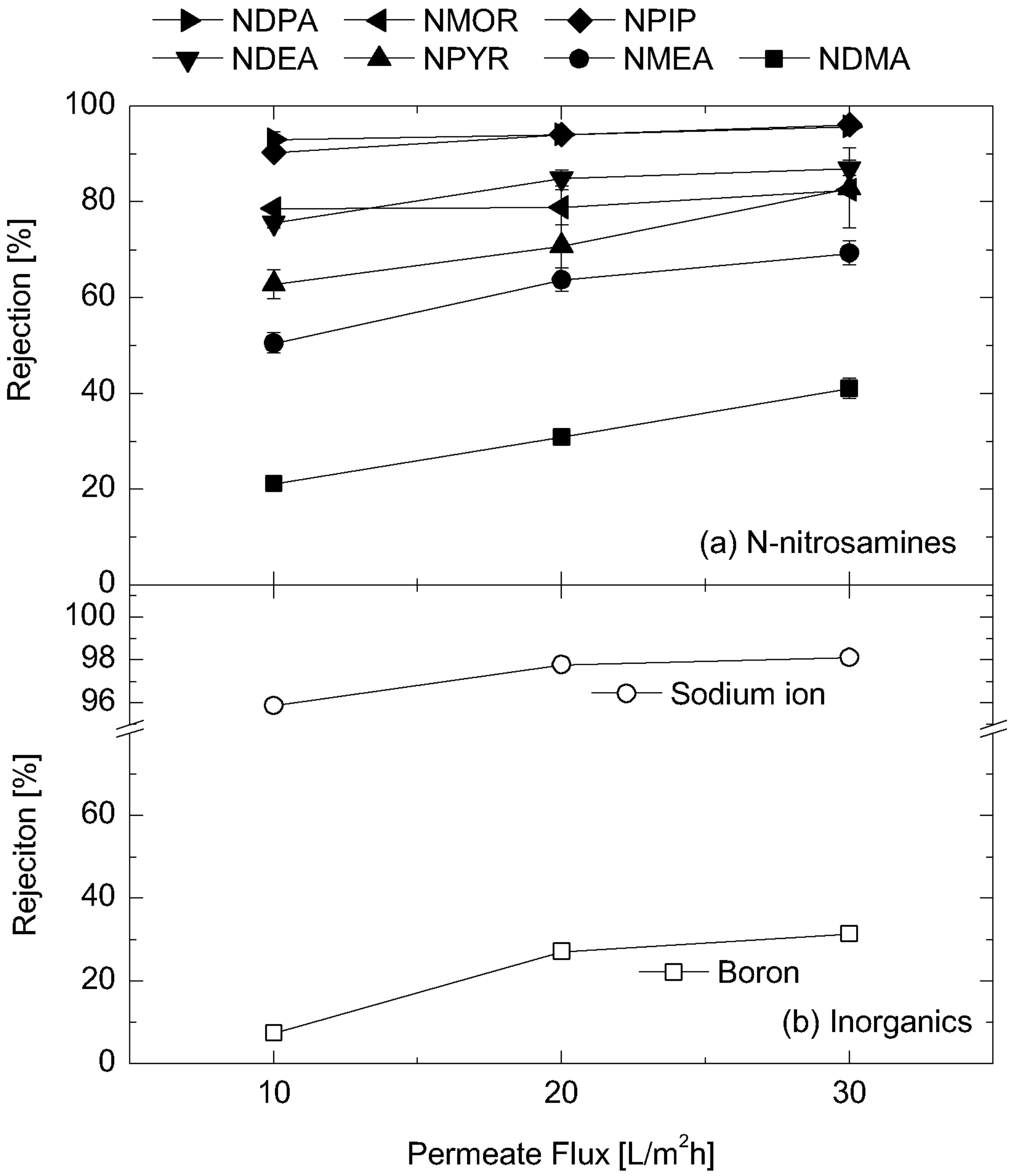

NDPA
NMOR
NPIP
NDEA
NPYR
NMEA
NDMA
Rejection [%]
100
80
60
40
20
0
(a) N-nitrosamines
100
98
96
Sodium ion
Rejeciton [%]
60
40
20
0
Boron
(b) Inorganics
10
20
30
Permeate Flux [L/m2h]

Figure 3

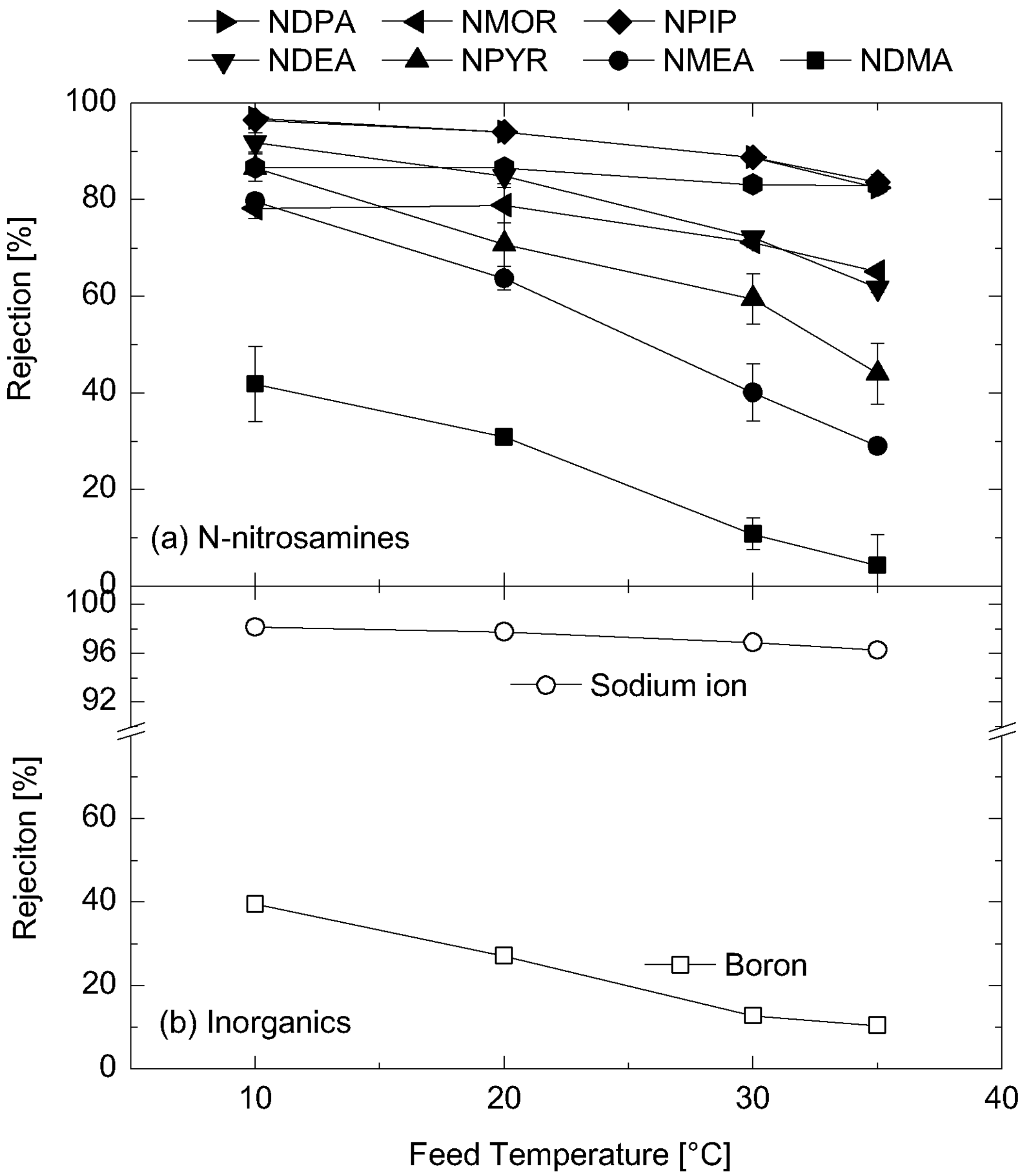
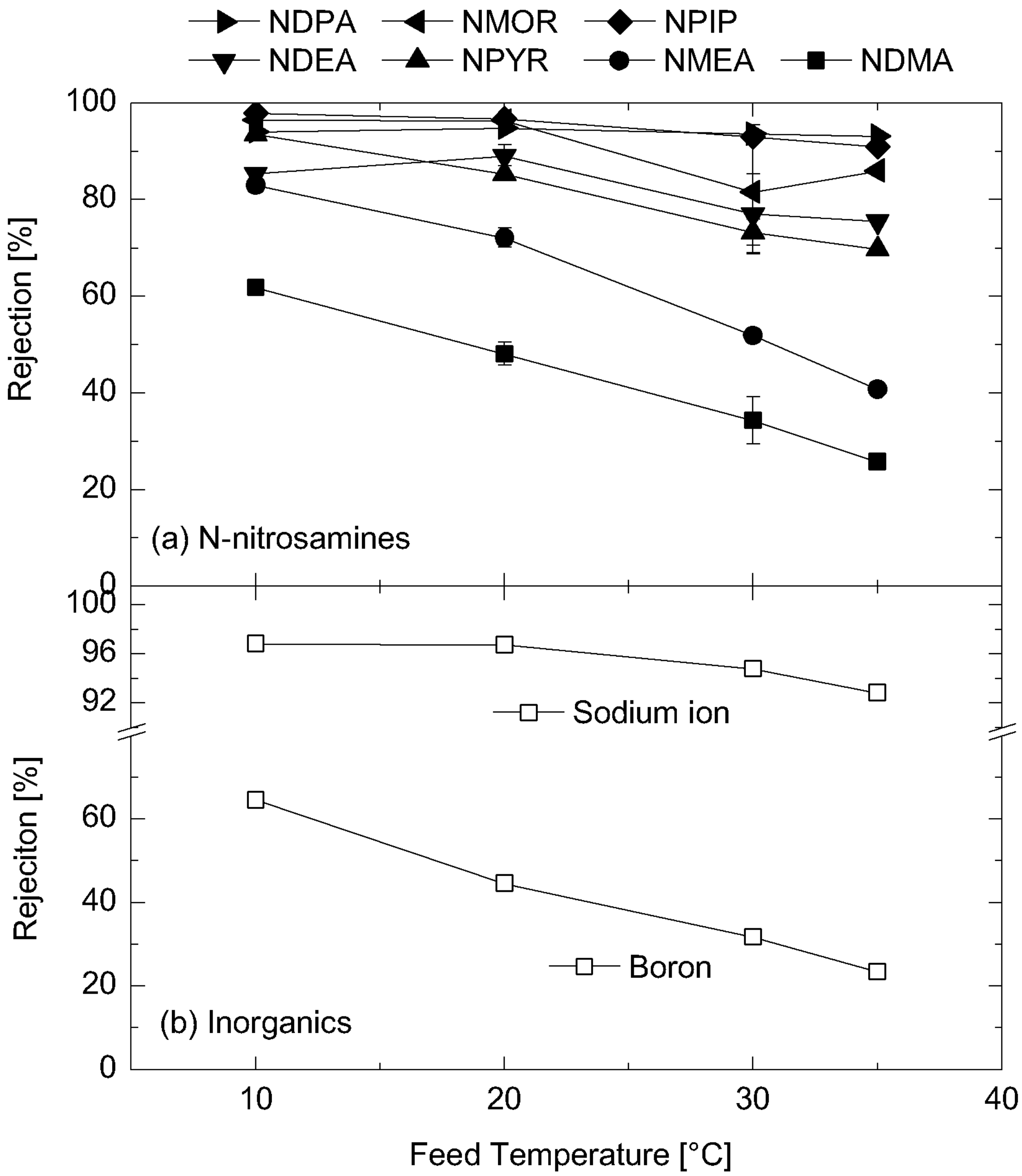

NDPA
NMOR
NPIP
NDEA
NPYR
NMEA
NDMA
Rejection [%]
100
80
60
40
20
0
(a) N-nitrosamines
100
96
92
Sodium ion
Rejeciton [%]
60
40
20
0
Boron
(b) Inorganics
10
20
30
40
Feed Temperature [°C]

Figure 4



NDPA
NMOR
NPIP
NDEA
NPYR
NMEA
NDMA
Rejection [%]
(a) N-nitrosamines
0
20
40
60
80
100
92
96
100
Sodium ion
Rejeciton [%]
Boron
(b) Inorganics
10
20
30
40
Feed Temperature [°C]

Figure 5

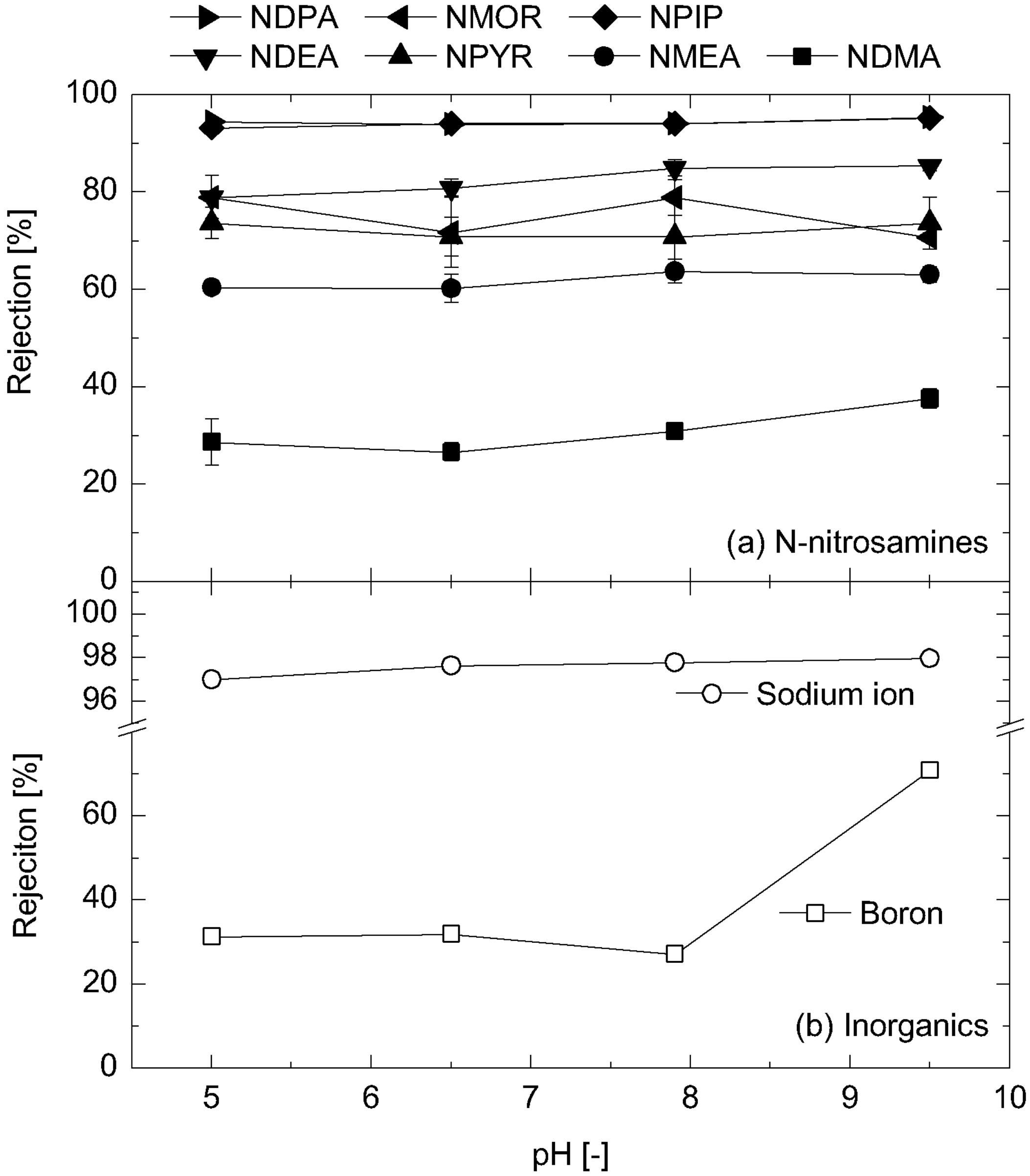
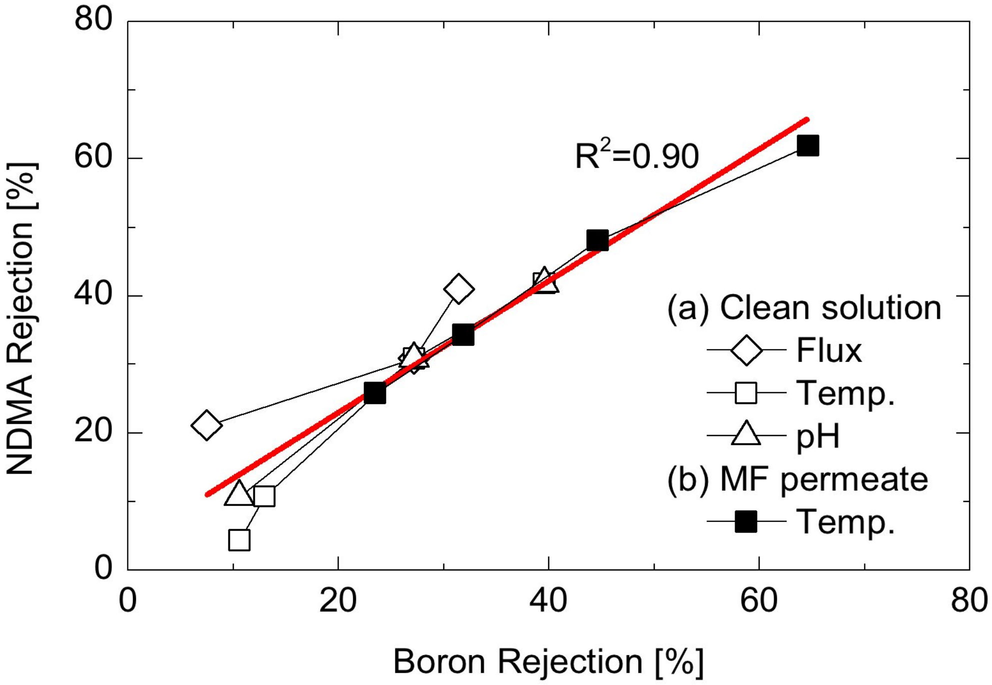

NDPA
NMOR
NPIP
NDEA
NPYR
NMEA
NDMA
Rejection [%]
100
80
60
40
20
0
(a) N-nitrosamines
100
98
96
Sodium ion
Rejeciton [%]
60
40
20
0
Boron
(b) Inorganics
5
6
7
8
9
10
pH [-]

Figure 6



80
60
40
20
0
0
20
40
60
80
NDMA Rejection [%]
Boron Rejection [%]
R^2=0.90
(a) Clean solution
Flux
Temp.
pH
(b) MF permeate
Temp.

Figure 7