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# Boron removal from geothermal water using DOW chemical high separation **BWRO** membrane

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# ABSTRACT

Boron presence in groundwater and surface water is caused by both natural and anthropogenic factors. Owing to their molecular nature, the removal of excessive boron concentrations from water is a serious problem. Among the water treatment systems used on an industrial scale, the most important ones are membrane systems, and among these a special role is played by reverse osmosis (RO). The study assessed the potential of RO with lowpressure spiral wound DOW FILMTEC BW30HR-440i polyamide thin-film composite membranes to enhance removal of boron. The tests carried out by the authors for the selected membrane have demonstrated that the retention level is dependent on the concentration of boron in the feed water and on the forms in which it is present. At an operating pressure of 11 bar and a feed pH of 8, the retention ranged from ca. 50-87%, with better results being obtained for lower boron concentrations in the feed water.

Keywords: Boron; Desalination; Reverse osmosis; Geothermal water

# 1. Introduction

Boron compounds are quite common under natural conditions, in lower or higher concentrations. Borates such as borax, boric acid or sodium perborate, and also minerals such as ulexite or colemanite are of commercial importance and are used in industry. Boron is one of the most important microelements that play a significant role in plant cultivation and breeding animals. The problem, however, is the small difference

between the dose required for the proper functioning of these organisms and an excessive dose. The daily dose tolerated by adult humans should not exceed 0.16 mg per kg body weight. Where this dose is exceeded, boron may become teratogenic [1,2].

Studies of boron ion content in drinking water and measures aimed at controlling it started to intensify in the early 1990s. National standards for drinking water quality are usually based on World Health Organization (WHO) guidelines and in European Union member countries also on the requirements of Directive 98/83/ EC [3]. The first 1958 "International Standards for

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Drinking-Water" and their subsequent 1963-1971 editions published by the WHO did not address boron concentrations in drinking water. In the first edition of Guidelines for Drinking-water Quality (GDWQ) published in 1984, it was stated that "no action was required" for boron [4]. In 1993, the WHO made an attempt to establish the upper limit for boron content in drinking water. The boron concentration limit was set at 0.3 mg/L [5]. In the annex to the guidelines published in 1998, permissible boron content in drinking water was raised to 0.5 mg/L [6]. This level was designated as temporary at the time. However, in 2003, WHO specialists, using new data and assessments conducted in the US, decided to re-examine the permissible content of boron in drinking water. At the same time, a group of experts dealing with water desalination pointed out that the permissible boron content in drinking water should take into account both the new data on its toxicity and the complex methodology for removing it from sea water, which exhibits high boron concentrations. In connection with this, in 2009, the WHO Drinking-water Quality Committee recommended that the permissible concentration of boron in drinking water be adjusted to 2.4 mg/L. The revised Guideline Value and Summary Statement were published in 2011, in the fourth edition of GDWQ [6].

The legal norms applicable in the European Union and Poland stipulate that the permissible concentration of boron in drinking water and in wastewater disposed to sewage and soil should be lower than 1.0 mg/L [3,7,8].

Boron presence in groundwater and surface water is caused by both natural and anthropogenic factors. Natural boron concentrations in freshwater result from the geochemical nature of geological structures, and in particular, from the borate content of soils and rocks, the mixing of waters from different aquifers and the effects of sea water intrusion. Rock weathering facilitates the passage of boron to the solution. Thereupon, a series of anions form:  $BO_2^-$ ,  $B_4O_7^{2-}$ ,  $BO_3^{3-}$ ,  $H_2BO_3^$ and  $H_4BO_4^-$  [1,9].

Owing to their molecular nature, the removal of excessive boron concentrations from water is a serious problem related to the supply of drinking water, particularly in areas that have insufficient freshwater resources and where treated seawater forms the basis for supplying water for human consumption.

Methods for removing boron from water can be divided into several categories. The first group of methods involves coagulation and electrocoagulation, and the other two are adsorption and ion exchange. The second includes membrane processes such as reverse osmosis (RO), nanofiltration (NF), electrodialysis and electrodeionisation as well as ultrafiltration combined with polymer complexation [9,10]. Table 1 shows the efficiency of individual boron removal methods for different types of water [1,10]. The most efficient methods are adsorption and complexing on ion-exchange resins, two-stage RO and adsorption on activated carbon.

In the treatment of water with high boron content, two methods are primarily used on an industrial scale, i.e. RO and ion exchange [10,11]. Research by a number of researchers [12–14] has demonstrated that the boron retention coefficient in the RO process is significantly affected by the pH of "raw" water and its temperature, ionic strength and the operating pressure of the process. It has been observed that the degree of removal decreases as temperature increases.

The retention of boron by RO membranes at low or neutral pH values that are typical of most ground water usually does not exceed 60%, which is in many cases insufficient to achieve the standards required for drinking water. This low removal ratio is due to the presence of boron in water and the dissociation process of boric acid, which is only hydrated at high water pH values [15–18].

The hydrated form of borate has a greater diameter and is negatively charged, which allows for better retention. However, raising the pH of the water that feeds the RO system exposes the membrane surface to scaling and fouling. Therefore, in systems for treating water with high boron content, water pre-treatment processes are important, which primarily serve the removal of divalent ions (Ca<sup>2+</sup>, Mg<sup>2+</sup>), e.g. in the NF process. Another significant factor is multistage RO systems in which boron is removed from the permeate (after pH adjustments) obtained after the first or subsequent RO stage [10,11].

Manufacturers of RO membranes have suggested solutions dedicated specifically for the treatment of water with high boron ion concentrations. Their design is aimed primarily at reducing the affinity for boron and increasing the affinity for water, and the membranes have tight molecular structures (with smaller pore sizes that enable the removal of boric acid particles). Such membranes are offered e.g. by Toray, Dow Chemical and Hydranautics [19–21]. Boron retention values guaranteed by manufacturers for selected membranes are presented in Table 2. For the Toray [19] or Hydranautics [21] membranes, the retention coefficient under consideration is determined for a standard concentration of 5 mg B/L in the water tested.

According to the specifications and tests conducted by membrane manufacturers, better boron removal effects are obtained using SWRO (seawater RO) membranes: 91–96% at a pH of 8. Hydranautics declares boron removal levels of 95% for the SWC4B and

Table 1

Processes efficiency in boron removal from water (based on [1,10])

Technology	Water	Retention coefficient		
Softening	Sea	Small		
Coagulation	Drinking	<28%		
Reverse osmosis	Sea	Up to 90%		
Adsorption on activated carbon	Treated feed water	43-78%		
Adsorption/complexation onto ion	RO permeate	>99%		
Exchange resins	RO permeate	40-100%		
Two-step RO with pH adjustment	Treated feed water	>80%		
Boron complexation	Permeate RO	>98%		

Table 2

Examples of RO membranes dedicated to water containing boron (based on [19-21])

Manufacturer	Membrane type	Boron retention coefficient (manufacturer's declaration) (%)	Operational pH
SWRO (sea wa	ater reverse osmosis)		
DOW	FILMTEC SW30HR LE-400i, FILMTEC	91 <sup>a</sup>	8
chemical	SW30HR-320		
	FILMTEC SW30XLE-400i, SW30XLE-440i	91.5 <sup>a</sup>	8
	SW30HRLE-370/34i	92 <sup>a</sup>	8
Toray	TM820E-400	91 <sup>a</sup>	8
2	TM840V-1760	92 <sup>a</sup>	8
	TM810C, TM820-400, TM820A-370,	93 <sup>a</sup>	8
	TM820A-400		
	TM820M-400, TM820M-440, TM840M-1760	95 <sup>a</sup>	8
	TM820K-400, TM820K-440	96 <sup>a</sup>	8
Hydranautics	SWC4 MAX, SWC4+, SWC4-LD	93 <sup>a</sup>	8
	SWC4B, SWC4B MAX	95°	6.5–7
BWRO (bracki	sh water reverse osmosis)		
DOW	BW30XFR-400/34i	80 <sup>a</sup>	8
chemical	BW30HR-440i	83 <sup>a</sup>	8
Toray	TM720C-440	95 <sup>b</sup>	10

<sup>a</sup>pH 8.

<sup>ь</sup>рН 10.

<sup>с</sup>рН 6.5–7.

SWC4B MAX membranes, even with natural or slightly acidic water reaction. For membranes used in the brackish water treatment process, the effectiveness of membranes is estimated at ca. 80% at a pH of 8 to 95% at a pH of 10 [19–21].

The paper presents the results of investigation of boron removal from geothermal water to compare the effectiveness to those which are declared by the manufacturer of membrane, especially for the pH of feed water. The RO system was equipped with spiral wound DOW FILMTEC BW30HR-440i polyamide thin-film composite membranes. The membrane type was selected by the specialist of the Veolia Water Technologies as the most effective to reduce high boron concentration from geothermal water. Membrane type was selected in accordance to feed water quality (brackish water) and increasing boron content.

# 2. Methods

#### 2.1. Apparatus

A dual hybrid process combining ultrafiltration and two independent stages (RO-1 and RO-2) connected in series was applied. The professional RO pilot facility was fitted with typical industrial plant components. Its continuous-cycle (i.e. 24 h/d) capacity was expected to be  $1 \text{ m}^3/\text{h}$ . The RO station was equipped with  $8^{\prime\prime}$  DOW FILMTEC BW30HR-440i membranes: polyamide thin-film composite membranes. The DOW FILMTEC<sup>TM</sup> BW30HR-440i RO

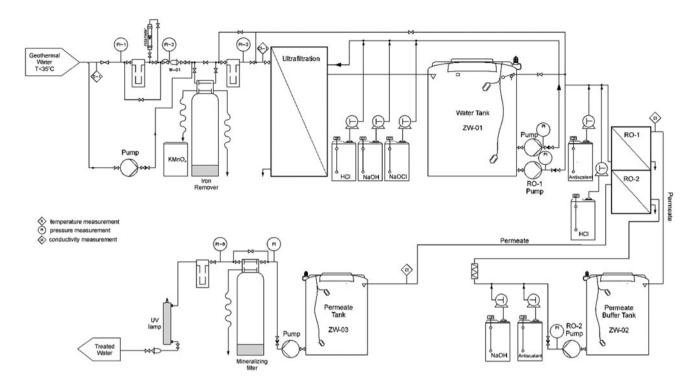


Fig. 1. Process diagram of the geothermal water desalination facility (after [22]).

Element is a high-performing, high-productivity element combining the highest active membrane area available in the industry today with the maximum retention coefficient brackish water RO membrane. It incorporates Dow's innovative BW30HR membrane sheet technology, designed to deliver the highest quality RO permeate. This is combined with the cleanability of a 28-mil feed spacer to minimise capital expenses in high-purity technological water applications without increasing operating flux [20].

The producer's advertisement states that the maximum temperature of the feed in the desalination process is below 45 °C, with a maximum pressure of 4.1 MPa, pH ranging from 2–11 and a maximum SDI feed water of 5. Minimum removal of salt is 99.4%, removal of silica (SiO<sub>2</sub>) is ca. 99.9%, boron—83% at a pressure of 1.55 MPa, a water temperature of 25 °C and a pH of 8 (Table 2). These membranes may be used for drinking water production.

To bring the boron concentration below its maximum level for drinking water (1 mg/L), the desalination was carried out in a two-step process (Fig. 1). The first step of reverse osmosis (RO-1) was equipped with two filtration modules and the second step (RO-2) had one module. Each module had one BWRO membrane, with an active area of 41 m<sup>2</sup> [22].

After water pre-treatment (iron-removal, ultrafiltration), the membrane separation performance was realised using the water from the two wells involved boron removal at the first RO stage (RO-1) at a constant pressure of 11 bar, acidic feed water at RO-1 (pH 5) and alkaline (pH 8) feed water at the second RO stage (RO-2). Both of the two RO stages worked independently and were connected in series.

The pH of the permeate after RO-1 was corrected to 8.0 and put to further filtration at a transmembrane pressure of 11 bar. The process yielded a water recovery level of about 80% on the second RO stage. Membranes were backflushed by a periodic pulsed permeate backflow at regular intervals after each short-term stoppage. Water temperature during testing was ca. 30°C. In both cases of water treatment, the samples of permeate were collected after 11 h of RO unit running under mentioned condition.

The studies were conducted at the Geothermal Laboratory of the Mineral and Energy Economy Research Institute of the Polish Academy of Sciences.

# 2.2. Water quality

All chemical analyses were performed at the certified Hydrogeochemical Laboratory of the

Hydrogeology and Engineering Geology Department of the University of Science and Technology in Cracow (PCA certificate No. AB 1050), using the inductively coupled plasma mass spectrometry (ICP-MS) and inductively plasma optical emission spectrometry (ICP-OES) methods. Chloride ion content and water alkalinity were determined by titration in accordance with accredited testing procedures.

The total dissolved solids (TDS) of geothermal waters tested were 6.5 g/L (water No. 1) and 2.5 g/L (water No. 1) and they have high concentrations of boron, respectively, 2.5–8.9 mg/L. The permeate after RO-1 exhibited different TDS values: (1) 573.0 mg/L and (2) 248.6 mg/L, and still elevated and high boron contents: 2.5 mg B/L (No. 1) and 6.0 mg B/L (No. 2) (Table 4).

#### 2.3. Methodology of boron species analysis

The concentration of boron in the solution was calculated as a function of pH using the Phreeqc Interactive 2.17.4799 program (PHREEQCI) [23]. Speciation modelling uses a chemical analysis of the water to calculate the distribution of aqueous ion species using an aqueous ion-association model.

# 3. Results

The modelling of the chemical species of boron present in the geothermal waters analysed has shown that 99.9% of the boron present in the water of slightly acidic pH (pH 5 of the feed water before RO-1) has the form of undissociated boric acid (H<sub>3</sub>BO<sub>3</sub>) and the rejection of that species varied between 48% in water No. 1 and 56% in water No. 2 [22]. For permeate after RO-1, at pH of 8, undissociated boric acid (H<sub>3</sub>BO<sub>3</sub>) is the dominant form of boron present in water (93.5%—No. 1 and 94.2%—No. 2). The remaining 6.5 and 5.8%, respectively, is the H<sub>2</sub>BO<sub>3</sub><sup>-</sup> metaboric ion (Table 3). It is meant that approximately only 6% of boron has the dissociated form and this results in a bigger size and larger retention of the passage of the molecule by the membrane.

Table 3

Analysis of boron speciation at pH 8 in the geothermal waters studied (feed water after RO-1)

Speciation	Water No. 1 (%)	Water No. 2 (%)
H <sub>3</sub> BO <sub>3</sub>	93.50	94.20
$H_2BO_3^-$	6.50	5.8

The assessment of the degree of dissociation of boron is important for predicting boron removal efficiency using RO membranes. The overall boron removal by RO is dependent on the boric acid/borate ion ratio and using an RO membrane for feed water with a high proportion of boric acid will lead to unsatisfactory levels of boron in the permeate [24]. These ratios for the tested (No. 1 and No. 2) waters were 14.38–16.22, respectively.

Earlier work by the authors [22] conducted for geothermal waters revealed the existence of a relationship between boron retention and its concentration in the feed. This efficiency decreased as boron content in the water increased, particularly for acidic or slightly basic water. On the other hand, favourable retention ratios of 96–97% were always obtained for highly basic feed (pH values exceeding 10). Conversely, for water pH of 10, the  $H_2BO_3^-$  metaboric anion was, at 95.5%, the main form of boron present at water analysed, and at pH 11, it formed 98.5% of total boron. The results of the analyses carried out confirm a direct relationship between boron removal and the form in which boron is present in the water solution [22].

The tests carried out using DOW FILMTEC BW30HR-440i membranes and two types of geothermal water with lower (2.53 mg/L) and higher (6.01 mg/L) boron concentrations showed—for a water pH of 8—values at the level of 87.4 and 50.25%, respectively. Analysis results are summarised in Table 4.

For the first type of water tested after RO-1, which had a slightly higher TDS (573.0 mg/L), but also lower boron content in the feed (2.53 mg/L), the required standards for drinking water in European Union countries were achieved. The efficiency of boron removal was also higher than for the second type of water. The retention ratio for water with lower mineral content but higher boron content was 50.25%. Boron concentration was reduced from 6.01 to 2.99 mg/L (Table 4), which precludes the use of this water as drinking water. The results of this test demonstrate clearly that as boron content in the feed water increases, the degree of its retention coefficient on RO membranes decreases. At the same time, at a feed pH of 8, as boron content in the water increases, the share of undissociated boric acid increases as well and its removal from water is ineffective.

As concerns the NaCl and SiO<sub>2</sub> reduction ratios, the results of the tests conducted were somewhat worse than those declared by the manufacturer, at, respectively, 95% for NaCl, and 95.3% for SiO<sub>2</sub> for No. 1 water (higher TDS) and 89.4% for NaCl and 88.8% for SiO<sub>2</sub> for No. 2 water (lower TDS) (Table 5).

Table 4

	Water No. 1			Water No. 2			
Parameter	Permeate after RO-1 (mg/L)	Permeate after RO-2 (mg/L)	Retention coefficient (%)	Permeate after RO-1 (mg/L)	Permeate after RO-2 (mg/L)	Retention coefficient (%)	
TDS	573.0	158.1	72.4	248.6	57.1	77.03	
Na	165.0	18.8	88.6	52.02	5.50	89.42	
Ca	18.00	0.241	98.7	10.0	0.3	97	
Mg	3.202	< 0.10	96.9	0.395	< 0.10	74.7	
Cl	220.0	11.2	94.9	115.1	8.6	92.5	
$SO_4$	37.7	2.5	93.4	10.25	1.3	87.3	
В	2.53	0,317	87.4	6.01	2.99	50.25	
SiO <sub>2</sub>	6.69	0.31	95.3	7.13	0.80	88.8	

Chemical	narameters	of treated	geothermal	water	(nermeate	after RO	2) and	the result	of retention	coefficient
Chemicai	parameters	of freateu	geomerman	water	(permeate)	aner KO	-2) anu	the result	of retention	coefficient

In their declarations, membrane manufacturers often provide basic laboratory testing parameters, including temperature, pressure, recovery percentage and standard salinity of test water, which was 2.0 g/L NaCl for the membrane under consideration. The concentration of boron in the feed was not declared, on the other hand, which could be of considerable significance when predicting the effects of reducing pollutant contents of water. Given the test results obtained, such data should be included in the specification of the membrane as it provides an important piece of information for engineers and technologists who design individual plants.

Possible reasons for the difference between the results observed and the manufacturer's declaration may also include slightly different process conditions. Laboratory tests carried out by membrane manufacturers assume a permeate recovery level of 15%, which

almost never occurs in industrial practice. A low permeate recovery level may protect the membrane from fouling or scaling, but such poor system efficiency is unacceptable in commercial use. For the tests carried out by the authors, the permeate recovery level adopted was ca. 80%, which significantly reduced the amount of the concentrate discharged.

Another important factor that affected the observed discrepancies between our own research and the laboratory tests carried out by the manufacturer could be water temperature during testing and the operating pressure used in the desalination process. The tests were run at a lower operating pressure of 11 bar, while the boron removal level of 83% declared by the manufacturer at a water pH of 8 was obtained at a pressure of 15.5 bar.

There have been several studies to discover the correlations between the dissociation constant of boric

Table 5

The comparison between manufacturer's specifications of the DOW FILMTEC BW30HR-440i membrane (after [24] and the result of investigation)

Parameter	Manufacturer's specifications	Water No. 1	Water No. 2
TDS (mg/L)	_	573.0	248.6
NaCl (mg/L)	2,000	362	132
$SiO_2 (mg/L)$	_	6.69	7.13
Boron (mg/L)	_	2.53	6.01
Permeate recovery (%)	15	80	80
Operating pressure (bar)	15.5	11	11
Temperature (°C)	25	30	30
pH	8	8	8
TDS retention coefficient (%)	_	72.4	77.03
Salt retention coefficient (NaCl) (%)	99.4	95.0	89.4
$SiO_2$ retention coefficient (%)	99.9	95.3	88.8
Boron retention coefficient (%)	83.0	87.4	50.25

acid and each governing factor such as temperature, pressure, pH and salinity. pH is the most influential parameter, while the effect of pressure is negligible at normal pressure ranges [25]. In regard to temperature and salinity, boric acid is easily dissociated as temperature and salinity increase. Increased feed salinity and/or temperature lead to increased amounts of boron in the permeate [25]. In theory, a higher salinity and temperature cause the  $pK_a$  value to decrease, which increases the fraction of the borate ion at a given pH. This suggests the possibility of removing boron at lower pH values by increasing salinity or temperature. However, the results of our investigations demonstrated that boron removal declines as salinity increases and this may be explained as the effect of charge neutralisation or of the fact that membrane surface potential is hindered at high salinity [25,26].

Thus, it is critical to determine the relationship between the boron removal ratio and the parameters that affect it, and to develop reliable designs in order to optimise the boron removal process.

#### 4. Conclusions

The efficient removal of boron from surface water and groundwater, including geothermal water, is a serious issue in operating water supply systems.

The efficiency of removal of boron from water declared by membrane manufacturers in SWRO water systems for a feed water pH of 8 ranges from 91 to 96%, and for BWRO systems it ranges from ca. 80% at a pH of 8 to 95% at a pH of 10.

The tests carried out by the authors for the selected DOW FILMTEC BW30HR-440i membrane have demonstrated that the retention level is also dependent on the concentration of boron in the feed. At an operating pressure of 11 bar and a feed pH of 8, the retention ranged from ca. 50–87%, with better results being obtained for lower boron concentrations in the feed.

The results of the analyses carried out confirm a direct relationship between boron removal and the form in which boron is present in the water solution. The modelling of the chemical species of boron present in the waters tested has shown that 93-94% of the boron present in a water of pH 8 has the form of undissociated boric acid (H<sub>3</sub>BO<sub>3</sub>). It is mean that approximately only 6% of boron has the dissociated form and this results in a bigger size and larger retention of the passage of the molecule by the membrane.

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