

Electrodialysis of coal mine water

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ABSTRACT

The treatment of model solution resembling the coal mine water from the “Bolesław Śmiały” coal mine in Łaziska Górne, Poland, was investigated. Two cases were investigated: 1) electrodialysis is directly applied in concentration of coal mine water; 2) coal mine water is pre-concentrated in the NF-RO system and then fed to the electrodialysis. Based on the bench-scale experiments, a model describing ions and water transport across the ion-exchange membranes was established. The model predicted that it is possible to obtain saturated brine, but the coal mine water must be pre-concentrated in the NF-RO system before the electrodialytic treatment.

1. Introduction

Hard coal mining is an important branch of European economy. Eleven of the EU member states mine coal, which is important source of energy for almost half of EU countries. Coking coal has been designated as a critical raw material by the European Commission [1] because of its importance for the metallurgical sector. Vast amounts of waste waters are generated by the Poland's coal mining industry. The coal mining waste waters have high salinity, sometimes even exceeding the sea water salinity, due to the seams of the coalfield lying in predominantly sandstone strata in which waters within this aquifer gradually increase in salinity with increasing depth [2]. The high salinity of coal mine waste waters is especially problematic in Upper Silesia region of Poland, as the nearest sea shore lies hundreds of kilometers from the mines, so only inland discharge to the rivers is possible. About 2–3 mln tonnes of salt is discharged annually to the rivers [3]; the Poland's main river, Vistula, has been experiencing excessive salinity for years, with 94% of chlorides originating from the coal mining. This creates massive ecological problems and creates the necessity for waste water utilization. Various methods have been proposed for the coal mine water treatment: application nanofiltration as a thermal method pretreatment [4], integrated reverse osmosis-forward osmosis system for treatment of low salinity coal mine water [5], capacitive deionization [6], and electrodialysis [7,8]. One of the feasible methods for coal mine waters utilization can be their concentration either by thermal or membrane methods and subsequent use in the production of evaporated salt.

Electrodialysis has been studied as the method of saline waste water concentration [9,10]. Turek et al. examined ED for desalination of brackish water, seawater, and coal-mine brines with simultaneous obtaining of concentrated brines. Electrodialytic desalination/concentration of brackish water (TDS ca. 10 g dm⁻³) in an integrated ED – thermal desalination system was examined. The ED unit stacks geometry, and the ED system arrangement and operating conditions were optimized to identify minimum desalination/concentration costs. The ED stack with relatively low 0.19 mm membrane to membrane distance, and three stages ED system arrangement was investigated to identify optimum process parameters. The minimum total desalination/concentration cost found at 255 g dm⁻³ NaCl concentrate content. When electrodialysis alone was applied for desalination - concentration up to TDS of 300 g dm⁻³ (close to NaCl saturation level) relatively low value of energy consumption, equal to 275 kWh/t of salt, was observed at optimum

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Table 1

The ionic composition of feed model solutions.

Water	“Bolesław Śmiały” coal mine water				RO retentate			
Ion	Cl ⁻	SO ₄ ²⁻	Mg ²⁺	Ca ²⁺	Cl ⁻	SO ₄ ²⁻	Mg ²⁺	Ca ²⁺
Concentration [mg·dm ⁻³]	13 300	790	224	315	32 800	25	18	153

conditions [11]. When ED-EDR concentration of coal-mine brine of salinity ca 58 g dm⁻³ up to 145 g dm⁻³ was considered, the energy consumption found, at 0.19 mm membrane to membrane distance, was equal to 162 kWh/t of salt produced while up to saturation electro-dialytic concentration consumed ca 307 kWh/t [12].

In this paper, an electro-dialytic treatment of model solution resembling the coal mine water from the “Bolesław Śmiały” coal mine in Łaziska Górne, Poland, was investigated. Two cases have been investigated: direct treatment of coal mine waste water with electro-dialysis and the pre-concentration of coal mine waste water in the nanofiltration (NF)-reverse osmosis (RO) system with electro-dialysis working on the RO retentate.

2. Experimental

Plate-and-frame electro-dialyzer of effective electrode area of 84 cm² and effective membrane length of 42 cm, equipped with four pairs of Neosepta CMX/AMX membranes and 0.36 mm thin spacers, was used for the experiments. The spacer used was based custom-designed hexagonal net [13–15], which exhibits enhanced mass transfer when compared to a commercial spacers, as well as narrow residence time distribution. The flow rate was set using the Heidolph 5101 peristaltic pump. Two feed model solutions were used for the tests: the first resembling “Bolesław Śmiały” coal mine water and the second resembling the retentate from the nanofiltration-reverse osmosis unit treating the “Bolesław Śmiały” coal mine water – see Table 1.

Membranes were stored in 3% sodium chloride, then mounted inside the module and washed with deionized water for 30 min. Before each experimental series, the feed solution was circulated through the electro-dialyzer stack for 15 min. The electro-dialysis was operated in a counter-current, constant voltage mode, the current distribution along the segmented electrode was measured for 15 min (1 being the diluate outlet/concentrate inlet, 5 being the diluate inlet/concentrate outlet). The diluate and concentrate were recycled back to a single tank to simulate a pseudo-steady state achieved during the single-pass operation. After 15 min, the linear flow velocity of the diluate and concentrate was measured, the diluate and concentrate samples were collected. Concentration of major ions in the collected samples was analyzed with ThermoDionex ICS-500 ion chromatograph equipped with a CS-16 column for cations or an AS-19 column for anions.

3. Results and discussion

Tables 2 and 3 present the results of batch-scale electro-dialysis experiments. The raw data obtaining during electro-dialytic desalination of model solutions was used for the estimation of empirical correlations describing the transport of water, chlorides, sulphates, magnesium and calcium across the Neosepta CMX/AMX membranes.

3.1. Mass transfer modelling

The molar flux of *i*-th ion, J_i , was calculated as:

$$J_i = (C_i^c u_c - C_i^f u_{c,f}) \cdot \frac{A}{M_i A_m} \quad (1)$$

Where C_i^c is the concentration of *i*-th ion in the concentrate, C_i^f is the concentration of *i*-th ion the feed, u_c is the concentrate linear flow velocity, $u_{c,f}$ is the feed linear flow velocity on the concentrate side, A is the cross-section area of the flow (4 compartments x 0.36 mm spacer x 2 cm channel width), A_m is the single membrane area (84 cm²) and M_i is the molar mass of *i*-th ion. The water flux, J_w , was calculated as:

$$J_w = (u_c - u_{c,f}) \cdot \frac{A}{A_m} \quad (2)$$

The concentration gradient across the membrane was assumed as the difference between the average chloride concentration in both compartments:

$$\Delta C = \frac{C_{Cl^-}^f + C_{Cl^-}^d}{2} - \frac{C_{Cl^-}^c + C_{Cl^-}^f}{2} \quad (3)$$

The water flux in electro-dialysis is a combination of osmosis (concentration gradient-dependent, mostly negligible due to extremely low hydraulic permeability of ion-exchange membranes) and electroosmosis (current density-dependent). Following model was assumed to explain the water transport across the membrane:

$$J_w = a_1 \cdot i \quad (4)$$

Table 2

The results of the batch-scale electrodialysis experiments performed on the model solution resembling “Bolesław Śmiały” coal mine water.

Series	Linear flow velocity [cm·s ⁻¹]				Current density [A·m ⁻²]	Voltage drop [V]	Concentration [mg·dm ⁻³]							
	Inlet		Outlet				Diluate				Concentrate			
	Diluate	Concentrate	Diluate	Concentrate			Cl ⁻	SO ₄ ²⁻	Mg ²⁺	Ca ²⁺	Cl ⁻	SO ₄ ²⁻	Mg ²⁺	Ca ²⁺
1	0.294	2.671	0.294	2.692	21	3	10590	751	181	182	13660	828	276	348
2	5.032	0.511	4.89	0.518	116	5	11720	777	201	218	13660	930	458	691
3	5.032	0.511	4.34	0.579	317	9	9490	718	155	166	13660	1339	601	834
4	1.94	0.186	1.918	0.19	18	3	13030	811	197	260	13660	889	343	468
5	1.94	0.186	1.908	0.208	101	5	11520	815	202	231	13660	1084	643	996
6	1.94	0.186	1.877	0.218	171	7	9930	742	171	188	13660	1592	749	1045
7	1.94	0.186	1.763	0.246	248	9	8720	695	127	149	13660	1925	738	1037
8	2.69	0.28	2.61	0.269	14	3	12610	833	212	271	13660	908	297	396
9	2.69	0.28	2.591	0.286	82	5	11440	815	199	214	13660	1050	471	703
10	2.69	0.28	2.591	0.296	127	7	10760	791	185	195	13660	1198	527	782
11	2.69	0.28	2.553	0.301	148	9	9880	753	162	175	13660	1234	451	598
12	2.69	0.28	2.65	0.312	148	11	10170	764	150	150	13660	1352	483	644
13	1.36	1.172	1.351	1.181	20	3	12072	784	249	276	13660	811	289	357
14	1.36	1.172	1.31	1.169	96	5	10269	750	195	211	13660	903	324	411
15	1.36	1.172	1.281	1.161	96	7	9963	730	182	198	13660	926	312	392
16	1.9	1.172	1.837	1.646	20	3	12523	830	234	255	13660	841	258	307
17	1.9	1.172	1.847	1.645	103	5	10712	775	194	206	13660	849	295	371
18	1.9	1.172	1.837	1.669	167	7	9312	703	166	179	13660	927	306	389
19	1.9	1.172	1.745	1.719	249	9	7620	607	135	127	13660	987	288	373
20	1.9	1.172	1.693	1.669	361	11	5277	464	23	NA	13660	1020	262	355
21	2.4	2.141	2.257	2.284	27	3	12956	797	265	319	13660	798	284	367
22	2.4	2.141	2.33	2.315	132	5	11114	770	212	241	13660	826	313	426
23	2.4	2.141	2.143	2.315	143	7	10780	726	205	242	13660	881	309	412
24	2.11	2.141	2.018	2.315	97	9	11413	749	217	256	13660	860	288	371
25	2.11	2.141	1.887	2.299	64	11	11855	769	227	269	13660	834	264	331

Table 3

The results of the batch-scale electrodialysis experiments performed on the model solution resembling RO retentate from the NF-RO system working on “Bolesław Śmiały” coal mine water.

Series	Linear flow velocity [cm·s ⁻¹]				Current density [A·m ⁻²]	Voltage drop [V]	Concentration [mg·dm ⁻³]							
	Inlet		Outlet				Diluate				Concentrate			
	Diluate	Concentrate	Diluate	Concentrate			Cl ⁻	SO ₄ ²⁻	Mg ²⁺	Ca ²⁺	Cl ⁻	SO ₄ ²⁻	Mg ²⁺	Ca ²⁺
1	1.719	0.202	1.497	0.273	153	5	29358	38	16	121	57790	20	35	364
2	1.719	0.202	1.206	0.334	501	9	27690	45	24	113	83120	22	41	401
3	2.184	0.224	2.092	0.285	260	7	28700	34	18	114	61400	36	40	282
4	1.566	0.162	1.497	0.273	488	11	23009	49.6	19.6	84.1	86813	61.8	48.3	261.9
5	1.566	0.162	1.516	0.242	368	8	24121	58	22.3	80.9	75390	41.6	50.7	265.4
6	1.566	0.162	1.55	0.201	148	5	28089	62	31.2	113.2	55661	38.9	56.5	243.2
7	0.951	0.951	0.946	0.956	28	3	31430	67	35	167	32281	58	37	185
8	0.951	0.951	0.899	0.975	147	5	26518	83	27	117	36094	53	44	214
9	0.951	0.951	0.864	1.001	286	7	21084	93	19	82	40607	52	57	238
10	0.951	0.951	0.839	1.034	420	9	15734	80	10	53	45111	121	77	241
11	0.951	0.951	0.813	1.059	522	11	11896	61	NA	40	44880	75	74	216

Table 4

The estimated values of parameters a_1 – a_8 of equations (4)–(9), electrodialysis working on the model solution resembling “Bolesław Śmiały” coal mine water.

Parameter	Value	Unit	p-value
a_1	$(3.46 \pm 0.82) \cdot 10^{-8}$	$\text{m} \cdot \text{A}^{-1} \text{s}^{-1}$	<1%
a_2	$(8.86 \pm 3.88) \cdot 10^{-6}$	$\text{m} \cdot \text{s}^{-1}$	3.2%
a_3	$(5.27 \pm 0.51) \cdot 10^{-5}$	$\text{mol} \cdot \text{A}^{-1} \text{s}^{-1}$	<1%
a_4	$(-2.34 \pm 0.39) \cdot 10^{-3}$	–	<1%
a_5	$(-8.48 \pm 0.13) \cdot 10^{-3}$	–	<1%
a_6	$(-4.71 \pm 0.97) \cdot 10^{-3}$	–	<1%
a_7	0.117 ± 0.015	$\text{kg} \cdot \text{s}^{-1} \text{m}^{-3}$	<1%
a_8	0.079 ± 0.011	$\text{kg} \cdot \text{A}^{-1} \text{m}^{-1}$	<1%

Table 5

The estimated values of parameters a_1 – a_8 of equations (4)–(9), electrodialysis working on the model solution resembling RO retentate from the NF-RO system working on “Bolesław Śmiały” coal mine water.

Parameter	Value	Unit	p-value
a_1	$(7.74 \pm 0.46) \cdot 10^{-9}$	$\text{m} \cdot \text{A}^{-1} \text{s}^{-1}$	<1%
a_2	$(-6.60 \pm 2.68) \cdot 10^{-6}$	$\text{m} \cdot \text{s}^{-1}$	3.7%
a_3	$(2.61 \pm 0.41) \cdot 10^{-5}$	$\text{mol} \cdot \text{A}^{-1} \text{s}^{-1}$	<1%
a_4	$(7.61 \pm 0.92) \cdot 10^{-3}$	–	1.7%
a_5	$(1.35 \pm 0.23) \cdot 10^{-2}$	–	<1%
a_6	$(9.99 \pm 1.41) \cdot 10^{-3}$	–	<1%
a_7	0.264 ± 0.037	$\text{kg} \cdot \text{s}^{-1} \text{m}^{-3}$	<1%
a_8	0.087 ± 0.014	$\text{kg} \cdot \text{A}^{-1} \text{m}^{-1}$	<1%

The ionic flux in electrodialysis follows the Nernst-Planck equation, so following model was assumed to explain the chloride molar flux:

$$J_{\text{Cl}^-} = a_2 \Delta C + a_3 i \quad (5)$$

Where i is the applied current density. Next, for the sake of simplicity, instead of calculating fluxes of each remaining ions, the change in the remaining ions concentration was calculated as:

$$\frac{C_{\text{SO}_4^{2-}}^f - C_{\text{SO}_4^{2-}}^c}{C_{\text{SO}_4^{2-}}^f} = a_4 J_{\text{Cl}^-} \quad (6)$$

$$\frac{C_{\text{Mg}^{2+}}^f - C_{\text{Mg}^{2+}}^c}{C_{\text{Mg}^{2+}}^f} = a_5 J_{\text{Cl}^-} \quad (7)$$

$$\frac{C_{\text{Ca}^{2+}}^f - C_{\text{Ca}^{2+}}^c}{C_{\text{Ca}^{2+}}^f} = a_6 J_{\text{Cl}^-} \quad (8)$$

Next, to assess the maximum chloride concentration in the concentrate in a pilot-scale unit, an empirical model was developed, linking the inlet concentrate linear flow velocity and applied current density:

$$C_{\text{Cl}^-}^c = a_7 \tau + a_8 i \quad (9)$$

Where the space-time is given by:

$$\tau = \frac{L}{u_{c,f}} \quad (10)$$

Where L is the module length (in the case of experimental unit, 42 cm). Based on the obtained model, a maximum chloride concentration achievable in a pilot-scale electrodialyzer of 100 cm membrane length was estimated.

The values of a_1 – a_8 parameters were estimated using Levenberg-Marquardt non-linear least squares algorithm as implemented in “minpack.lm” module of GNU R statistical software and are presented in Tables 4 and 5.

The analysis shows that it is possible to predict the ionic fluxes and water flux across the Neosepta CMX/AMX membranes in the tested conditions (counter-current, single-pass operation, 0.36 mm intermembrane spacers) knowing the applied current density, feed concentration, and the required diluate and concentrate concentrations. Based on the empirical model linking the concentration of chlorides in the concentrate stream, space-time of the electrodialyzer, and the applied current density, chloride concentration achievable in a 100 cm long electrodialyzer unit was estimated at a linear flow velocity range of 0.15–0.42 cm/s and current density

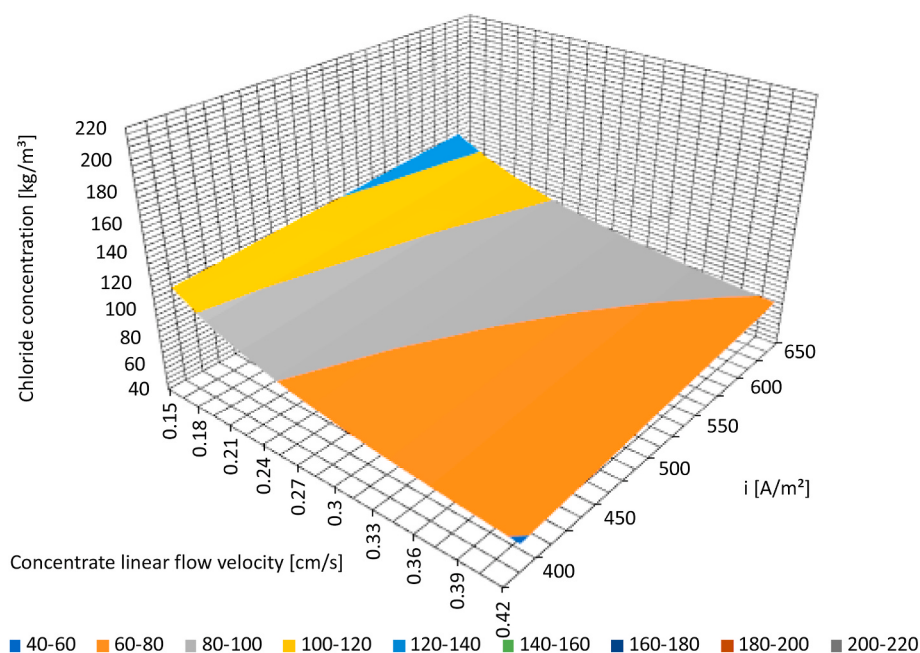


Fig. 1. Estimated effect of concentrate linear flow velocity and applied current density on the final chloride concentration in the produced concentrate (membrane length: 100 cm, feed water: the model solution resembling “Bolesław Śmiały” coal mine water).

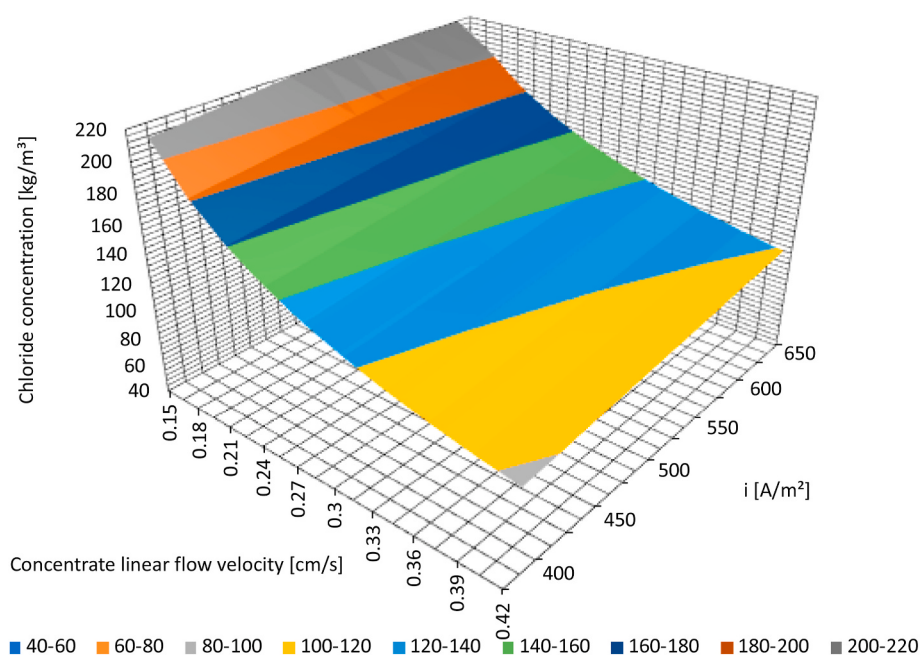


Fig. 2. Estimated effect of concentrate linear flow velocity and applied current density on the final chloride concentration in the produced concentrate (membrane length: 100 cm, feed water: the model solution resembling RO retentate from the NF-RO system working on “Bolesław Śmiały” coal mine water).

range of 400–650 A/m^2 . The results are presented in Figs. 1 and 2.

The results indicate that it should be possible to reach the desired level (180 g dm^{-3} of chlorides) in electrodialysis if the membrane length is 100 cm and the electrodialysis works on the RO retentate; however, it's not possible to produce saturated brine if the electrodialysis works directly on the coal mine water, at least in the investigated conditions (membrane length 100 cm, current density 400–650 A m^{-2} , concentrate linear flow velocity 0.15–0.42 cm s^{-1}). The results are in agreement with the results obtained by Casas et al.

[16,17], who modelled a pilot ED plant for SWRO brine concentrations. Based on the Nernst-Planck equation and mass balance equations, they have developed a model describing the plant performance with respect to the transport of sodium chloride, however calcium and magnesium transport was neglected. Although the desired chlor-alkali feed concentration of 300 g/L as NaCl was not reached in the experiments because current density was not high enough, the model predicted that such concentrations should be reached in the investigated plant at ca. 600–700 A/m² after 25–28 h of operation (inlet concentration ca. 65 g/L as NaCl).

It's also worth to note that the results predict it should be possible to cross the sodium chloride solubility (ca. 192 g/L as Cl[−] at room temperature). Although this may be hard to achieve practically, given it requires simultaneously very high current density and very low linear flow velocity in the concentrate compartment, it indicates a possibility of electrochemically-driven membrane crystallization of sodium chloride to occur. This is a subject of a separate, ongoing study.

3.2. Cost analysis

The cost analysis was performed for the second case, ED working on the model solution resembling retentate from NF-RO system working on the “Bolesław Śmiały” coal mine water. In order to estimate costs of electrodialytic brine production, following model has been assumed:

- 1) calculate the membrane length required for obtaining 180 g dm^{−3} as Cl[−] given the concentrate linear flow velocity and current density by rearranging Eqns. (9) and (10):

$$L = (C_{Cl^-} - a_8 i) \frac{u_{cf}}{a_7} = (180 - 0.087i) \frac{u_{cf}}{0.264} \quad (11)$$

- 2) correlate current density, i [A·m^{−2}], and voltage drop on the electrodialyzer, U [V], using data presented in Table 3 ($R^2 = 0.957$):

$$U = 0.0151i + 2.7257 \quad (12)$$

In the industrial-scale electrodialyzer the voltage drop on the electrodes and in the anolyte/catholyte compartments is negligible compared to the voltage drop on the membrane stack itself; however, in the case of bench-scale studies, when small number of membrane cells is used (4 in this experiments), the voltage drop on the electrodes and in the catholyte/anolyte compartment is disproportionally large compared to the voltage drop on the membrane stack. To mitigate this issue and better translate the collected laboratory data into large-scale electrodialysis, it was assumed that the intercept of Eqn. (11) is equal to the voltage drop outside the membrane stack – meaning the voltage drop of electrode reaction and the voltage drop in the 0.36 mm channel filled with 0.5 M sodium sulfate solution would be 1.36 V per cathode and anode each, which is a reasonable value. The correlation between the current density and voltage was thus assumed as:

$$U = 0.0151i \quad (13)$$

- 2) calculate the DC energy costs required for production of 1 m³ of 180 g dm^{−3} as Cl[−] concentrate, C_{DC} [kWh/m³] in the electrodialyzer equipped with 4 pairs of membranes of effective length L , flow channel width h of 0.11 m and intermembrane distance s of 0.36 mm, working at current density i and concentrate linear flow velocity u_{cf} :

$$C_{DC} = \frac{ULhiC_{current}}{\eta_r 4u_{cf}hs} \quad (14)$$

where η_r is the AC/DC rectifier efficiency, assumed as 95%, and $C_{current}$ is the DC energy costs, assumed as 0.06 €/kWh.

- 3) calculate the equipment costs, C_{cap} [€·m^{−3}], assuming membrane life-time t_{life} of 85 000 h, membrane costs C_m of 100 €·m^{−2}, and membrane costs being 30% of total equipment costs [18]:

$$C_{cap} = \frac{9C_m Lh}{0.3 \cdot 4u_{cf}hs t_{life}} \quad (15)$$

- 4) calculate the pumping costs, C_{pump} [€·m^{−3}], assuming diluate flow being 10 times higher than the concentrate flows:

$$C_{pump} = \frac{(10 + 1)u_{cf}hs\Delta PC_{current}}{\eta_p 4u_{cf}hs} \quad (16)$$

where η_p is the pump efficiency, assumed as 80%, and the pressure drop, ΔP [bar], is given by the empirical correlation [18]:

$$\Delta P = 0.0862u_{cf}L \quad (17)$$

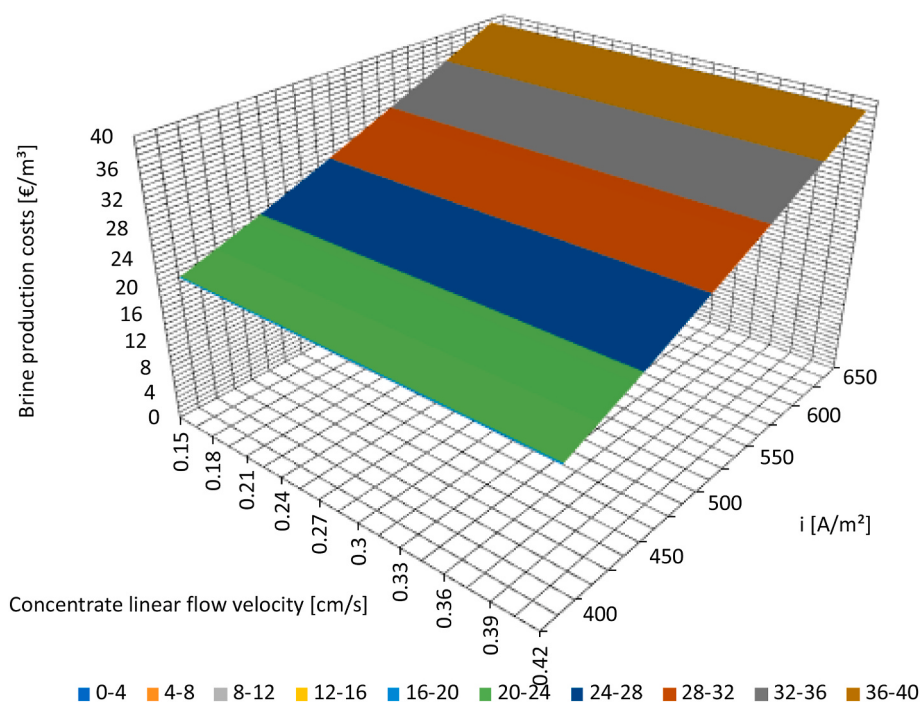


Fig. 3. Estimated costs of saturated brine production by electrodialysis using RO retentate from the NF-RO system working on “Bolesław Śmiały” coal mine water as the feed.

The overall costs, C [€/m³], are given by the equation:

$$C = C_{DC} + C_{cap} + C_{pump} \quad (18)$$

The influence of current density and the concentrate linear flow velocity on the saturated brine costs is presented in Fig. 3. The costs are highly dependent on the applied current density, ranging from ca. 20 €/m³ to 40 €/m³, but the concentrate linear flow velocity has almost negligible effect – this is because the majority (81–92%) of the costs are the DC energy costs. This suggests the necessity for investigating membranes with lower electric resistance and intermembrane spacers with better mass transfer coefficients, even at the cost of increasing the equipment or pumping costs. Such membrane properties could be achieved by fabricating thinner homogenous membranes, even at the cost of lowering their mechanical strength. Because the linear flow velocities are low, this may be the way to go further with membrane development, at least for the specific use case discussed here. On the other hand, decreasing the membrane thickness may lead to increased osmotic water flux, significant factor if high concentration difference between the diluate and concentrate is desired, and eventually prohibit the electrodialysis from achieving high sodium chloride concentration in the concentrate stream.

4. Conclusions

A semi-empirical mass transfer model, describing the ions and water transport across the ion-exchange membranes, was established and used for the prediction of maximum attainable chloride concentration in the industrial-scale electrodialyzer. It was found that the electrodialysis of the “Bolesław Śmiały” coal mine water can be applied as a method for the production of concentrated solution (>180 g dm⁻³ as Cl⁻), a raw material for evaporated salt production, as long as the raw coal mine water is pre-concentrated in the NF-RO system. The projected costs of saturated brine production range from 20 to 40 €/m³, with the DC energy costs being the major contribution.

Author statement

Krzysztof Mitko: Conceptualization, Methodology, Investigation, Writing - Original Draft, Writing - Review & Editing, Visualization, Project administration, Resources, Data Curation.

Aleksandra Noszczyk: Investigation.

Piotr Dydo: Investigation, Resources.

Marian Turek: Writing - Original Draft.

CRedit authorship contribution statement

Krzysztof Mitko: Conceptualization, Methodology, Investigation, Writing – original draft, Writing – review & editing, Visualization, Project administration, Resources, Data curation. **Aleksandra Noszczyk:** Investigation. **Piotr Dydo:** Investigation, Resources. **Marian Turek:** Writing – original draft.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: We declare possible conflict of interests with one of the Editors – Dr. H. Spanjers works in the same research project by which the presented research was funded, we suggest another Editor should be handling the manuscript.

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