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Valorization of coal mine effluents — Challenges and economic opportunities

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ABSTRACT

Coal-mine effluent treatment has the potential to both reduce the environmental impact of the effluent and provide economic opportunities by recovering valuable minerals and clean water. In this study, we modeled a novel treatment process, which includes nanofiltration (NF), two-step crystallization, reverse osmosis (RO), electrodialysis (ED), multi-effect distillation (MED), and a NaCl crystallizer, and performed a techno-economic analysis of its full-scale implementation, using a circular economy approach. We estimated the thermal and electrical energy consumption to be 745.5 kWh_{th}/ton_{NaCl} and 565.1 kWh_{el}/ton_{NaCl} (or 13.6 kWh_{th} and 10.3 kWh_{el} per m^3 of feed effluent), respectively. The levelized cost of the NaCl salt that accounts for the revenue from the plant's co-products ($Mg(OH)_2$, $CaSO_4$ and, pure water) was estimated to be 203 USD/ton_{NaCl}. The economic viability of the treatment chain can be improved by using renewable electricity sources, reducing the total expenditure on NF and RO, and integrating alternate technologies into the treatment plant.

1. Introduction

As the European Union (EU) aims to achieve net zero emissions of greenhouse gases (GHG) by 2050 with measures such as rapid phasing out of coal from the power and heating sector [1], coal remains an important component of the energy mix for several member states, including Poland [2]. The country is a dominant producer of hard coal in the EU, having produced 61.6 million tons or 92% of the EU's hard coal production in 2019 [2]. However, the mining sector exerts tremendous pressure on the water resources, accounting for 13.2% of the industrial water usage [3], and has serious human health and environmental consequences, including water pollution [4]. Through a mine-water discharge of approximately 350,000 m^3/day, the Polish coal mines discharge around 4000 ton/day of chlorides and sulfates to rivers [5], making the water unsuitable for drinking or even bathing [6]. Water protection is one of the priorities of the EU [7,8], and the pretreatment of coal-mine effluents to recover useable water and remove impurities from them before discharging into water bodies is essential. In addition to preventing the pollution of water bodies, the recovery of marketable products such as water, NaCl salt, and minerals such as calcium and magnesium, where the latter is identified as a critical raw material by the European Commission [9], can help incentivize the additional treatment of the effluents.

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Ericsson et al. [10] studied a zero-discharge treatment plant to treat the wastewater from two adjacent coal mines in Poland – Debiensko and Budryk. The treatment plant involved pretreatment and reverse osmosis (RO), and consisted of a thermal plant for brine concentration and crystallization of sodium chloride (NaCl). The paper summarized the initial results of the plant operation and noted a combined energy consumption of $\sim$318 kWh/ton$_{NaCl}$ by the brine concentrator and crystallizer. Turek et al. [5] proposed pre-concentrating the effluent from the Debiensko Polish coal mine using an electrodialysis – electrodialysis reversal (ED-EDR) two-step process instead of a brine concentrator before evaporating the salt. They estimated a reduction in the energy consumption from 970 kWh/ton$_{NaCl}$ to 500 kWh/ton$_{NaCl}$. Turek et al. [11] proposed the use of nanofiltration (NF) as a pretreatment to remove Ca^{+2} and SO_4^{-2} ions to enable the use of low-energy vapor compression (VC) thermal desalination and estimated an energy consumption of 450 kWh/ton$_{NaCl}$. The current study is developed within the framework of an EU-funded project called ZERO BRINE [12], which aims at "closing the loop" by recovering minerals and water from industrial wastewater using a circular economy approach. Within this project, a pilot-scale plant to treat the effluent from the Bolesław Śmiały coal mine in the Upper Silesian Coal Basin (USCB) was developed and demonstrated [13]. The pilot plant, which consists of technologies such as NF, crystallizers, RO, ED, and MED, recovers demineralized water, $Mg(OH)_2$, $CaSO_4$, and NaCl salts. Under this project, Mitko et al. [14] investigated two cases for the treatment of the effluent from the Bolesław Śmiały coal mine in Łaziska Górne, Poland: i) direct use of ED to concentrate the effluent, ii) pre-concentration of the effluent in an NF-RO system and then feeding it to the ED. They observed that pre-concentration in the NF-RO system was necessary to obtain saturated brine from the ED. Within the same project, Micari et al. [15] performed a techno-economic analysis of treating the effluent from a Polish coal mine, wherein they analyzed five treatment chains to extract ions in the form of marketable minerals, such as magnesium hydroxide ($Mg(OH)_2$), calcium sulfate ($CaSO_4$), calcium carbonate ($CaCO_3$), and NaCl. They considered several combinations of technologies in the pretreatment phase (NF, crystallization) and concentration phase (RO + membrane distillation (MD), multi-effect distillation (MED) and salt crystallization). For a brine having total dissolved solids (TDS) of 23 kg/m^3 and a magnesium content of 0.28 kg/m^3, the treatment plant with the lowest levelized cost of NaCl salt (LSC) ($\sim$90 USD/ton$_{NaCl}$) consisted of two NFs, a three-step crystallizer to recover $Mg(OH)_2$, $CaSO_4$, and $CaCO_3$, a MED, and a NaCl crystallizer. This study builds on and extends these studies [14,15] by including additional processes such as ED, using real brine data from the Bolesław Śmiały coal mine, and adopting a new modeling approach to simulate the implemented process flow in the pilot-scale plant to determine the plant economics corresponding to its full-scale implementation.

2. Methodology

In this section, the implemented pilot plant and the assumptions used in its modeling are discussed in detail. The techno-economic parameters of the modeled technologies are discussed with a focus on ED. Lastly, the method used for the economic and sensitivity analyses is explained.

2.1. Description of the modeled treatment plant

After initial treatment in the ultrafiltration and decarbonization units (excluded from this analysis) of the pilot plant, the coal-mine effluent containing ions of Na^+, Cl^-, Ca^{2+}, Mg^{2+}, and SO_4^{-2} was sent to the proposed treatment plant, which consisted of two phases (see Fig. 1): pretreatment and concentration [15]. The first phase is used to separate bivalent ions (i.e., Ca^{2+}, Mg^{2+}, and SO_4^{-2}) from the effluent using a nanofiltration (NF) unit and to recover two co-products, $Mg(OH)_2$ and $CaSO_4$, using an alkaline solution such as dolime in a crystallization unit. The effluent is then sent to the second phase, where the monovalent ions, Na^+ and Cl^-, are concentrated to obtain another co-product, demineralized water, and the main product, NaCl salt. Based on the choice of the main product, we obtained the corresponding levelized cost from the simulation as the levelized cost (in this case, the levelized cost of NaCl salt (LSC)).

Fig. 2 shows the treatment plant modeled in this study in greater detail. The modeled plant differs slightly from the implemented pilot plant (discussed later in this section). The pretreatment phase included two nanofiltration units and a two-stage crystallization process. In the pretreatment part, the effluent is passed through the first nanofiltration unit (NF1), which mainly separates bivalent ions, such as Ca^{2+}, Mg^{2+}, and SO_4^{-2}. The retentate from NF1, rich in these bivalent ions, is sent to a two-step crystallization train, wherein we first extracted the Mg^{2+} ions in the form of $Mg(OH)_2$ using a 10 wt % dolime solution ($(Ca(OH)_2.Mg(OH)_2)$) followed by

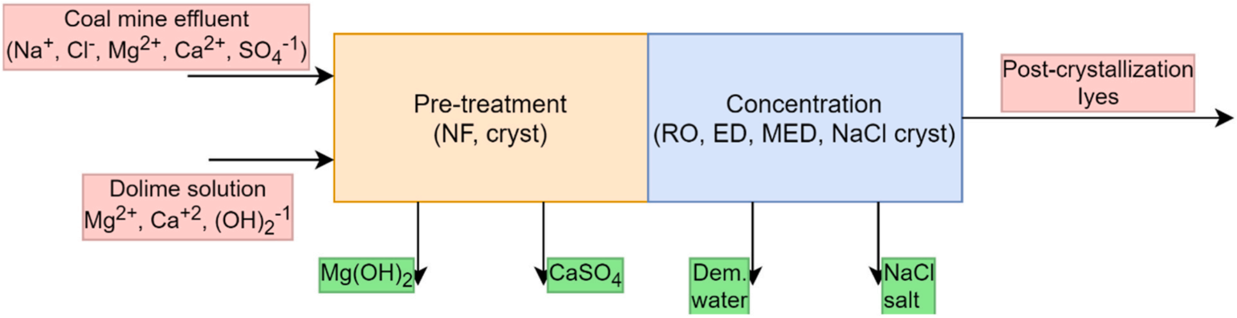

Fig. 1. Schematic overview of the inlets and outlets of the pretreatment and concentration phases of the proposed treatment plant.

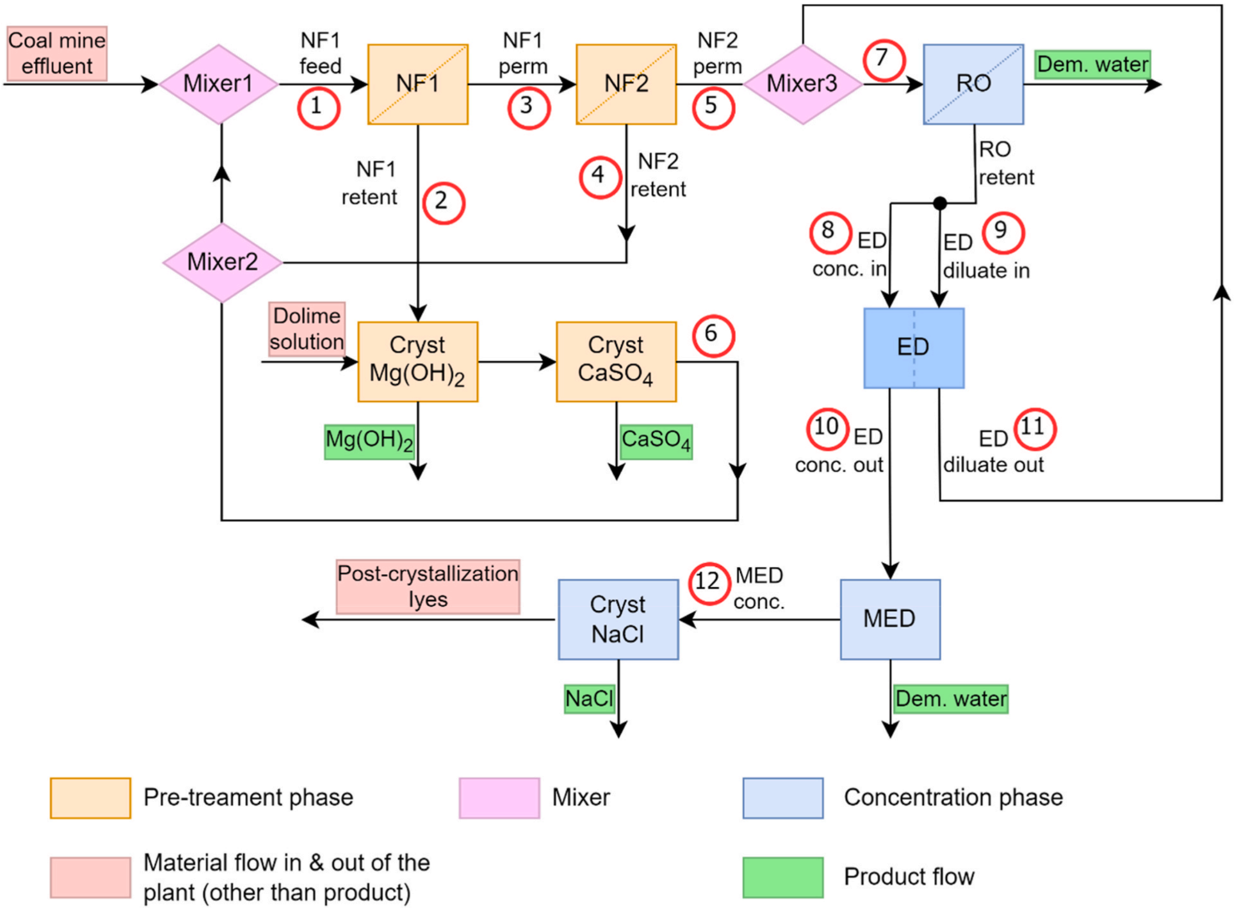

Fig. 2. Schematic of the proposed treatment plant to treat coal-mine effluent.

the precipitation of Ca^{2+} in the form of $CaSO_4$ in the second step. The precipitation of $CaSO_4$ occurs due to the supersaturation of the effluent solution. The availability of additional Mg^{2+} and Ca^{2+} ions from the dolime suspension increases the yields of $Mg(OH)_2$ and $CaSO_4$. The crystallizer effluent is recycled by mixing with the coal-mine effluent in Mixer1 before it enters NF1. The permeate from NF1, rich in monovalent ions, such as Na^+ and Cl^-, was fed to another nanofiltration unit, NF2, to remove the remaining bivalent ions. The retentate from NF2 is also recycled and mixed with the crystallizer effluent in Mixer2.

In the concentration phase, the permeate from NF2 is fed to an RO unit, which concentrates the effluent to a specified retentate concentration (expressed in ppm of NaCl) while providing demineralized water in the form of RO permeate. Based on a user-specified ratio ($Dil - con - ratio_i$), the RO retentate is then split into concentrate and diluate streams for the ED unit. Based on the chosen current density i and the desired salinity of the ED concentrate outlet (expressed in ppm of NaCl), the concentration of the concentrate stream increases along the length of the ED stack, whereas that of the diluate stream decreases. The diluate outlet from the ED is recirculated and mixed with the feed of RO in Mixer3, whilst the concentrate outlet is sent to the MED unit for further concentration. In the MED unit, pure water is obtained as a distillate whereas the concentrate is sent to a NaCl crystallizer to obtain NaCl salt crystals.

It should be noted that the recycling of streams to the preceding components through Mixer3, made it difficult to simulate and design the RO and ED units. This is because Mixer3 requires the flow rates and ionic compositions of both the NF2 permeate and the ED diluate outlet to size the successive technologies in the treatment chain (RO, ED, MED, and NaCl crystallizer). However, until the NF2 permeate passes through the RO and ED units, the flow rate at the ED diluate outlet could not be obtained. To solve this issue, we modeled dummy RO and ED units, which treat only the NF2 permeate. The diluate outlet from the dummy ED is then mixed with the permeate of NF2 in Mixer3 and fed to the actual RO and ED models, which are considered for the techno-economic analysis. It is noteworthy that although a similar design issue exists for NF1, wherein the streams are recycled using two mixers Mixer1 and Mixer2, we addressed it by directly using the mass flow rate and ionic composition (available from the pilot plant measurements) at the NF1 inlet.

Secondly, the ED system implemented in the pilot plant is a cascade system comprising two ED units. The ED unit that we modeled has three salinities as an input parameter: at the concentrate inlet ($S_{c,i}$), the concentrate outlet ($S_{c,o}$) and the diluate inlet ($S_{d,i}$). In this case, it is not possible to model each ED unit independently because, in a cascade operation, one unit depends on the output of the other. Therefore, we modeled the ED as a single unit instead of a cascade system. Lastly, the crystallization train at the pilot plant first precipitates $CaSO_4$, followed by the addition of a dolime solution to recover $Mg(OH)_2$. In our crystallizer model, this sequence is reversed, and we assume that a 100% recovery of minerals is possible. This could result in a deviation in the amount of minerals recovered (elaborated in the Results and Discussion section).

The primary effluent characteristics of the modeled treatment plant are summarized in Table 1. The ion concentration of the effluent at the feed of NF1 and the required salinities of the RO retentate, ED concentrate outlet, and MED concentrate were chosen using on-site measurements at the pilot plant. To simulate a full-scale plant, we increased the coal-mine effluent flow rate used in the pilot plant by 100 times (50 from 0.5 m^3/h), comparable to the volumes produced at coal mines [15].

2.2. *Techno-economic modeling of the technologies*

The techno-economic modeling of all technologies of the proposed treatment plant, except ED, has been done and described in previous works under the ZB project: MED and NF models [16,17], RO [18], and crystallizers [15]. The relevant models have been implemented on Python and integrated on a common simulation platform called RCE (Remote Component environment) [19] to simulate the treatment plant. The input parameters selected for these models are listed in Table 2. Some of these parameters were the same as those considered in the pilot plant, such as the feed pressure and percentage recovery in the NFs (recovery is the ratio of the flow rate in the permeate to that in the feed stream). We assumed that apart from the mass flow rate, other parameters in a full-scale plant would remain unchanged.

In this study, an ED model was developed based on the work of Nayar et al. [20] which itself is adapted from the model of McGovern et al. [21]. An ED system consists of several pairs (N_{cp}) of anion-exchange membranes (AEM) and cation exchange membranes (CEM), wherein each pair of AEM and CEM constitutes one cell pair [20]. These cell pairs together with the cathode, anode, concentrate, and diluate channels, make up the ED stack. In this study, the ED unit used for concentrating the brine was modeled as a single-stage design, implying that the change in the concentrate salinity from the inlet ($S_{c,i}$) to the outlet ($S_{c,o}$) occurs across a single ED cell pair [20]. To simulate salt and water transportation across the stack for the calculation of the cell-pair area A_{cp}, the length of the stack required discretization into small computational cells. $A_{cp,tot}$ represents the total effective cell-pair area and was calculated by multiplying A_{cp} with N_{cp}. It should be noted that in the referred literature a finite-difference method was used to perform the species balance on each channel [20,22]. In our work, we did not perform this analysis and neglected the water flux in the calculation of A_{cp}. This resulted in a lower $A_{cp,tot}$ compared to that computed by Nayar et al. [20] for an industrial stack (self-calculation: 20.5 m^2, Nayar et al.: 22.7 m^2, and reference industrial stack: 24 m^2). To improve the accuracy of the economic and energy consumption calculations, we introduced an area-correction factor, which increases the $A_{cp,tot}$ by 17% (in this example from 20.5 m^2 to ~24 m^2). Based on the new cell-pair area $A_{cp,tot\ (corrected)}$, we recalculated N_{cp}, which was then used for the calculation of the cell-pair voltage V_{cp}. This method of calculating the electricity consumption was validated by comparing our model results with those of Nayar et al. [20] for a standalone ED system having the following parameters: a concentrate inlet flow rate of 50 m^3/h, a $dil-con-ratio_i$ of 80, an ED current density of 300 A/m^2, a concentrate and diluate inlet salinity ($S_{c,i}$ and $S_{d,i}$, respectively) of 35 g_{salt}/kg_{sol} (each), a concentrate outlet salinity ($S_{c,o}$) of 200 g_{salt}/kg_{sol}, and a corresponding annual salt production of 96,836 ton_{NaCl}/year (based on Nayar et al.'s method). For the same conditions, our model estimated a specific energy consumption of 210 kWh_{el}/ton_{NaCl} against that of 219 kWh_{el}/ton_{NaCl} estimated by Nayar et al. (calculated to be within 4.1%). Thus, the calculation of cell-pair area and electricity consumption using our ED model was validated. The main technical parameters of ED used in this study are listed in Table 2. Tanaka et al. [23] stated that a current density of 300 A/m^2 was optimal for a low energy consumption of ED stacks using seawater and desalination feeds. However, we selected a current density of 400 A/m^2, based on the current densities tested at the pilot plant (400 and 650 A/m^2) [14], and subsequently performed a sensitivity analysis. The economic model of ED was based on Nayar et al. [20], wherein the specific capital cost of the ED plant was assumed to be 600 $USD/m^2_{membrane}$. The membrane cost and lifetime were taken as 222 $USD/m^2_{membrane}$ and 7 years, respectively. We assumed a plant life of 30 years.

For the economic analysis of other technologies, the capital cost of NF was based on the Verberne cost model [16], whereas those of the crystallizers [16] and MED [17] were estimated using the module costing technique [25]. The capital cost estimation for RO [18] was based on the literature. These costs were updated to January 2018 using a CEPCI index of 576.7 and then annualized using technology-specific depreciation periods, an interest rate of 6%, and a plant availability of 94%. The annualized total expenditure (TOTEX) was calculated using the summation of capital and operational expenditures (CAPEX and OPEX, respectively). The annual revenues were computed based on the amount of water and minerals recovered through their respective technologies. The

Table 1
Effluent characteristics used in the modeling of the treatment plant.

Parameter	Value
Ion concentration at NF1 feed [mol/m^3]	Na: 611.89, Cl: 625.24, Mg: 6.06, Ca: 19.39, SO_4: 19.19
TDS at NF1 feed [kg/m^3]	39.0
Plant rejection at NF1 and NF2 [-]	Na: 0.22, Cl: 0.235, Mg: 0.956, Ca: 0.903, SO_4: 0.973
Mass flow rate of coal-mine effluent [m^3/h]	50
Mass flow rate at NF1 feed [m^3/h]	69.5
Salinity of RO retentate [ppm of NaCl]	58,561
Salinity of ED conc. outlet [ppm of NaCl]	163,121
Salinity of MED conc. [ppm of NaCl]	241,670

Table 2
Main technical parameters of the technology models.

Nanofiltration (NF1 and NF2)	
Feed pressure P_{feed} [bar]	48 and 38 resp. (pilot plant data)
Recovery [%]	75 (pilot plant data)
Reverse osmosis (RO)	
Number of elements per vessel (first stage)	8
Number of elements per vessel (second stage)	6
Electrodialysis (ED)	
Current density i [A/m^2]	400
Effective cell-pair area of each membrane pair A_{cp} [m^2]	0.395 [20]
Length, width and height of stack [m]	0.84L x 0.47W × 0.005H [24]
Electrode potential V_{el} [V]	2.1 [20,21]
Area correction factor [%]	17
$Dil-con-ratio_i$ [$-$]	5.3 (pilot plant data)
Multi-effect desalination (MED)	
Steam temperature T_s [°C]	100
Number of effects [$-$]	2

technology-wise annualized expenditures and revenues were then expressed as a levelized cost of the main product, NaCl salt ($LSC_{effective}$), as follows [15].

$$LSC_{effective,\ tech}\left[\frac{USD}{ton_{NaCl}}\right] = \frac{CAPEX_{tech} + OPEX_{tech} - Revenue_{water\ or\ CaSO_4 or\ Mg(OH)_2}\left[\frac{USD}{year}\right]}{NaCl\ crystals\ produced\ \left[\frac{ton_{NaCl}}{year}\right]}$$

If each component of the $LSC_{effective,\ tech}$ was expressed independently, it would be referred to as LSC_{capex}, LSC_{opex} and $LSC_{revenue}$, respectively. Adding the $LSC_{effective,\ tech}$ for each technology in the treatment chain gave $LSC_{cumulative}$, which is the minimum price at which NaCl salt needs to be sold for the treatment plant to reach a break-even. Table 3 lists the economic assumptions. The heating cost was based on a low-temperature heat source available at an industrial site [17]. The cost parameters for each technology could be found in the literature [15–18,20].

To account for the variations in the levelized cost of the main product (NaCl salt) or LSC due to input energy cost and by-product price variations, we performed a sensitivity analysis by independently varying the values of the chosen sensitivity parameters by up to 50% on either side (in steps of 25%) as compared to those of the base case. The following parameters were selected for the sensitivity analysis: the costs of electricity and heating, and selling prices of water, $Mg(OH)_2$, and $CaSO_4$. An additional parameter, current density i across the ED stack, was considered.

3. Results and discussion

We first assess the performance of the simulated treatment process and then discuss the results (energy consumption of the treatment plant, amount of minerals and water recovered, economic analysis discussing the technology-wise annual revenues and expenditures, and levelized cost of the main product and its contributors) in this section. The results of the sensitivity analysis are presented and validated by comparing them with the pilot plant measurements.

3.1. Performance of the integrated process

In this sub-section, the performance of individual technology models and their integration in the simulation platform RCE are assessed. We first examine the effectiveness of ion separation in the modeled technologies. Table 4 below shows the ion concentrations of key streams. As seen, the two NFs were able to reject ions like Mg, Ca and SO_4 to the retentate (streams 2 and 4) which could be recovered in the two-step crystallizer. Due to the requirement of the tools for RO, ED and MED technologies, the ion concentration is indicated in terms of NaCl salinity. As the effluent moves through these tools, the ion concentration increases from 25,108 ppm (before RO) to 241,671 ppm (after MED) and these salinities match those from the pilot plant (cf. Table 1).

Table 3
General economic assumptions [15].

Parameter	Value
Capacity factor of the plant [$-$]	0.94
Interest rate [%]	6%
Electricity cost [USD/kWh$_{el}$]	0.1035
Heat cost [USD/kWh$_{th}$]	0.01
Dolime [USD/ton]	60
$Mg(OH)_2$ [USD/ton]	1200
$CaSO_4$ [USD/ton]	40
Pure water price [USD/m^3]	1

Table 4
Flow characteristics (ion concentration, salinity and flow rate) of streams from the RCE simulations (the stream numbers correspond with those depicted in Fig. 2).

Stream	1	2	3	4	5	6	7	8	9	10	11	12
Ion concentration [mol/m^3]												
C_{Na}	611.9	1017.3	474.9	842.3	366.9	938.0	–	–	–	–	–	–
C_{Cl}	625.9	1062.1	477.6	853.5	367.2	968.6	–	–	–	–	–	–
C_{Mg}	6.1	24.7	0.25	1.1	0.01	0	–	–	–	–	–	–
C_{Ca}	19.4	75.8	1.9	8.1	0.19	20.4	–	–	–	–	–	–
C_{SO_4}	19.2	78.1	0.8	3.6	0.03	5.1	–	–	–	–	–	–
NaCl concentration [ppm]												
C_{NaCl}	–	–	–	–	–	–	25,108	58,561	58,561	163,122	38,541	241,671
Flow rate [m^3/h] (in bracket the scaled values from pilot plant data)												
$\dot{Q}$	69.5 (same)	16.5 (17.4)	54.0 (52.1)	11.7 (13.0)	43.0 (39.1)	28.5 (–)	55.5 (62.9)	3.7 (4.5)	19.4 (–)	3.8 (–)	19.4 (23.8)	2.2 (2.8)

Next, the flow rates of key streams are compared with that of the pilot plant (adjusted to full-scale). It can be seen that although recovery ratio (feed-to-permeate conversion ratio) is NF tool's input parameter and was set to 75% for both NFs, the simulated NFs estimate a slightly higher recovery of ~78% and ~80%, respectively. In contrast, despite the resulting higher permeate flow rates from both NFs, the simulated RO feed flowrate (stream 7) was lower than that observed in the pilot plant. This could be due to the higher recovery from the simulated RO (57%).

Generally, the ion concentrations and flow rates of the modeled treatment plant are comparable with the pilot plant data, aside from the above-mentioned deviations.

3.2. *Recovery of minerals and energy consumption*

The modeled treatment chain recovers demineralized water (from RO and MED) and three minerals ($CaSO_4$, $Mg(OH)_2$, and NaCl) at the expense of dolime. Fig. 3 shows the hourly flow rates of these substances in the treatment chain. As seen, NaCl and $Mg(OH)_2$ represent the largest and smallest amounts of mineral recovery, respectively, with NaCl accounting for over 80% of the recovered minerals. Interestingly, the two-step crystallizer consumed ~4% more raw material (dolime) by weight than the amount of minerals ($CaSO_4$ and $Mg(OH)_2$) it recovered. With an effluent flow rate of 50 m^3/h, the treatment chain can recover ~34 m^3/h of demineralized water. Although water is recovered in both RO and MED, the RO unit recovers the bulk of it (~96%) (not shown in the figure).

Four technologies (NF, two-step crystallization, RO, and ED) consume only electricity, while two technologies (MED and NaCl crystallization) consume both heat and electricity. Table 5 lists the electric and thermal power required for each technology. As observed, except for the two-step crystallizer and MED, other technologies required much high electric power. NF2 required ~38% less power than that required by NF1, explained by the reduced feed and ion concentrations (cf. Table 4). The heating power of MED is 2.5 times more than that of the NaCl crystallizer.

Fig. 4 shows the specific electricity and heat consumption of these technologies corresponding to the annual production of NaCl crystals and the effluent feed rate. The MED dominated in terms of heat (~72% of the total heat) as well as total energy consumption (~41% of the total energy). This was followed by the heat consumption of the NaCl crystallizer (210.8 kWh_{th}/ton_{NaCl} or 28% of the total heat and ~16% of the total energy). NF consumed the highest amount of electricity in the treatment plant (~37% of the total electricity and 16% of the total energy). The electricity consumptions of RO and ED were comparable, whereas those of MED and the

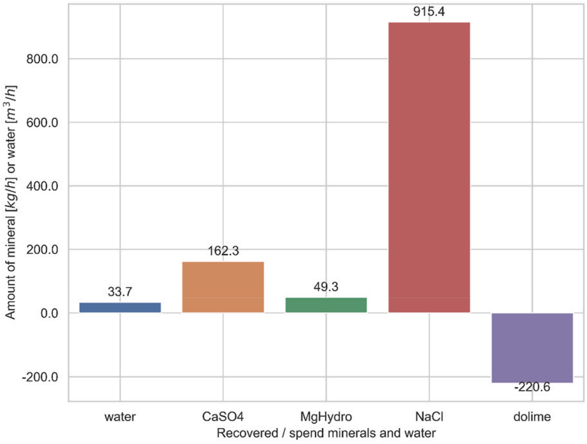

Fig. 3. Expenditure on and recovery of minerals and water in the treatment chain. Except for water [m^3/h], the units of measurement are kg/h.

Table 5
Electric and thermal power required by the technologies in the proposed treatment plant.

Technology	NF1	NF2	Crystallizer (two-step)	RO	ED	MED	NaCl crystallizer
Electric power [kW_{el}]	119.3	74.0	4.6	114.9	103.8	2.5	98.0
Thermal power [kW_{th}]	–	–	–	–	–	489.4	192.9

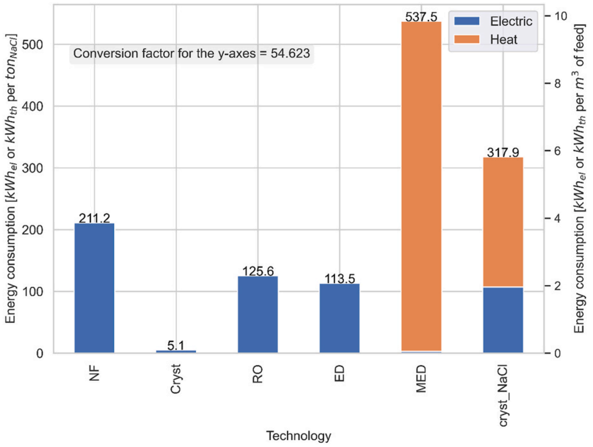

Fig. 4. Electric and thermal consumptions of different technologies in the treatment plant. On the left Y-axis, this is represented corresponding to the NaCl recovery, whereas that on the right Y-axis corresponding to the effluent feed rate.

two-step crystallizer were negligible. Overall, the treatment chain has a thermal and electrical energy requirement of 745.5 kWh_{th}/ton_{NaCl} and 565.1 kWh_{el}/ton_{NaCl} (or 13.6 kWh_{th} and 10.3 kWh_{el} per m^3 of feed coal mine effluent), respectively.

3.3. *Economic analysis*

Fig. 5 compares the annual expenditures (CAPEX and OPEX) for each technology within the treatment plant and the generated revenues (for a detailed breakdown of the annual expenditures, refer to Table A- 1 in the appendix). The depicted water revenue is the combined value of that of RO and MED. The CAPEX and OPEX of NF1, NF2, and RO are comparable and higher than those of any other technology. When combined, the NF units account for 55% and 46% of the overall CAPEX and OPEX, respectively. The CAPEX and OPEX of the NF system were dominated by the costs of the energy supply system (88%) and quality control and installation (44%), respectively (cf. Table A- 1). The CAPEX and OPEX of RO are driven by structural (53%) and maintenance (47%) costs, respectively. Structural costs include the costs of membranes, pressure vessels, and piping and instrumentation. The combined TOTEX of the NF and

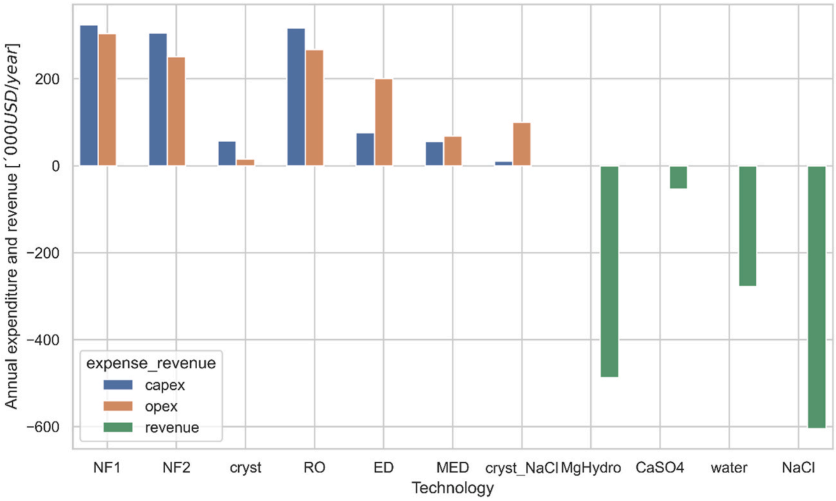

Fig. 5. Technology-wise expenditure and revenue of the treatment plant.

RO units accounted for more than 75% of that of the entire treatment plant. Among all the technologies, only ED, MED, and NaCl crystallizer had an OPEX higher than their CAPEX. In the cases of ED and NaCl crystallizer, electricity consumption was the biggest contributor to their OPEX (44% and 84%, respectively).

In the case of either crystallizer, it is observed that the revenue generated by the recovered minerals substantially outweighs their TOTEX. The revenue from $Mg(OH)_2$ accounts for more than 90% of the total revenue generated by the two-step crystallizer despite accounting for only ~23% by weight of its mineral recovery (cf. Fig. 3). The revenues generated by the two-step crystallizer, RO, and NaCl crystallizer account for 742%, 46%, and 549% of their annual expenses, respectively. The total annual revenue (1.42 million USD/year) accounts for ~60% of the total annual expenditure.

The bar and line plots in Fig. 6 depict the technology-wise distribution of the effective levelized cost of salt ($LSC_{effective}$) and the $LSC_{cumulative}$ along the treatment chain, respectively. It can be seen that only the two-step crystallization unit had a negative $LSC_{effective}$ (62 USD/ton$_{NaCl}$), mainly due to the large revenues generated from $Mg(OH)_2$ (cf. Fig. 5). Although the RO unit had LSC_{capex} and LSC_{opex} comparable to those of the NF1 and NF2 units, its $LSC_{effective}$ is reduced considerably due to its water revenue. The NF units were the biggest cost contributors with a combined $LSC_{effective}$ of ~157 USD/ton$_{NaCl}$, accounting for over 77% of the total $LSC_{effective}$ of 203 USD/ton$_{NaCl}$. The LSC contributions of the MED and NaCl crystallizer were relatively small.

3.4. Sensitivity analysis

Using the LSC (203 USD/ton$_{NaCl}$) calculated in the preceding subsection as the base case, a sensitivity analysis was performed. The results are shown in Fig. 7. As expected, increasing the price of the by-products ($Mg(OH)_2$, $CaSO_4$, and water) of the treatment chain reduced the LSC (quadrant IV of the graph) and vice versa. It was observed that an increase in the price of $Mg(OH)_2$ had the greatest impact on the reduction of LSC (~171 USD/ton$_{NaCl}$), whereas that of $CaSO_4$ had the least (~200 USD/ton$_{NaCl}$). This effect is due to the revenue from $Mg(OH)_2$ being considerably higher than that of $CaSO_4$ (cf. Fig. 5). Increasing the price of water by 50% resulted in an LSC of ~185 USD/ton$_{NaCl}$. In contrast, reducing the price of $Mg(OH)_2$, $CaSO_4$, and water increased the LSC to 236, 207, and 222 USD/ton$_{NaCl}$, respectively. Despite the variation in the current density of ED exhibiting a similar behavior, it is likely to be a non-linear one, because increasing the current density by 50% to 600 A/m^2 results in a marginal reduction in LSC (202 USD/ton$_{NaCl}$) compared to the increase in LSC (221 USD/ton$_{NaCl}$) upon reducing the current density to 200 A/m^2.

On the other hand, as expected, decreasing the energy cost (electricity and heat) decreased the LSC (quadrant III of the graph). Although thermal energy dominated the energy consumption of the entire treatment plant (see subsection 3.2), the impact on LSC upon decreasing the electricity cost is considerably higher impact (up to ~174 USD/ton$_{NaCl}$) compared to that (up to ~200 USD/ton$_{NaCl}$) upon reducing the heat cost. Similarly, increasing the electricity and heat costs increases the LSC to up to 233 and 207 USD/ton$_{NaCl}$ respectively.

3.5. Discussion and analysis

We validated the material recovery and electricity consumption values calculated using the RCE model of the treatment plant by comparing them with the pilot plant measurements. This is followed by a discussion of potential opportunities to reduce the LSC.

Table 6 compares the recovery amounts of minerals and water. It can be seen that the model estimates a considerably larger recovery of $CaSO_4$ compared to the site measurement. This could be due to a combination of factors such as the relatively small scale of the pilot plant as well as model limitations (assumption of complete mineral recovery in the two-step crystallizer, precipitation of

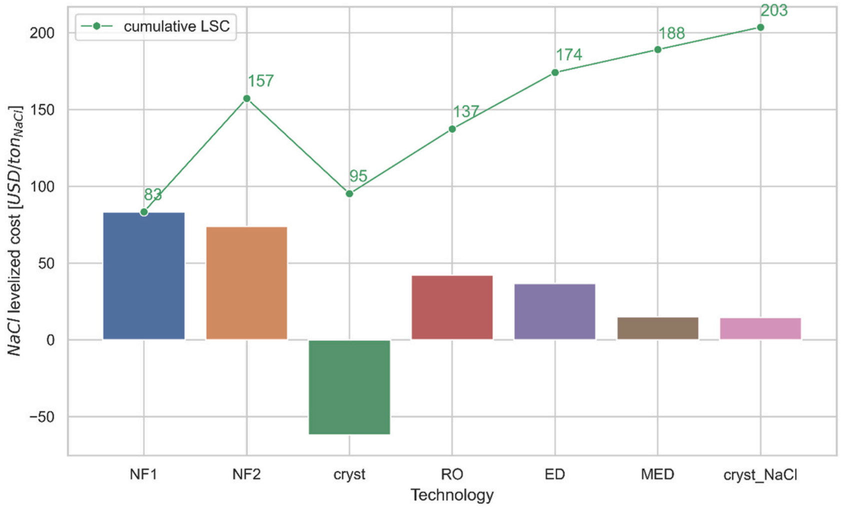

Fig. 6. Technology-wise distribution of effective levelized cost of salt ($LSC_{effective}$) as a bar plot. $LSC_{cumulative}$ plotted as a line.

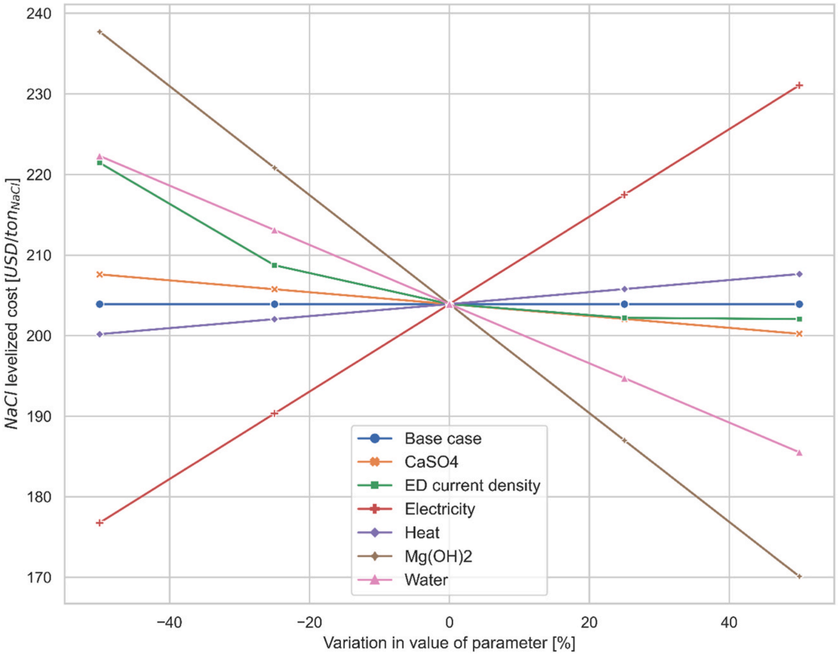

Fig. 7. Sensitivity of LSC according to chosen sensitivity parameters. The parameters are varied by - 50% and +50% as compared to the "base case" of 203 USD/ton$_{NaCl}$.

Table 6
Comparison of amount of recovered minerals/water at site with the model results.

Recovered mineral/water	Model	Site measurement adjusted to full-scale model [26]
$CaSO_4$ [kg/h]	162.3	33.69
$Mg(OH)_2$ [kg/h]	49.3	45.49
Water [m^3/h]	33.7	38.8
NaCl [kg/h]	915.4	825.61

$CaSO_4$ after the addition of dolime solution, that is, in the second step instead of the first as is the case in the pilot plant). However, this should not affect plant economics considerably, given the low share of $CaSO_4$ in revenue generation (cf. Fig. 5). Moreover, the model's estimation of $Mg(OH)_2$, water, and NaCl recovery is comparable to the site measurements.

Table 7 presents a comparison of the energy consumption of the technologies within the pilot plant with the model results. As observed, the energy consumption of the modeled NF and RO technologies closely matches that of the pilot plant. However, the estimated electricity consumption of the modeled ED, which is a single-unit and single-stage system, is less than half of that of the pilot plant's two-unit cascade system, indicating its high energy efficiency. In the case of MED, the pilot plant value is based on the literature for a fully electric and modern MED and thus could not be directly compared with the modeled MED, which primarily used thermal energy in the form of low-temperature steam at 100 °C.

Thus, apart from a few deviations, the RCE model of the treatment chain was validated against the pilot plant with comparable estimations in terms of mineral/water recovery and energy consumption.

Based on the discussed model assumptions and techno-economic parameters of the technology models, we estimated a salt cost of 203 USD/ton$_{NaCl}$. This was 35%–153% higher than the typical market value of high-purity (99.5%) NaCl salt, ranging between 80 and 150 USD/ton$_{NaCl}$ [15]. The NF and RO units were the biggest cost factors, accounting for over 75% of the plant's annual expenditures (cf. subsection 3.3). This limits the potential economic benefits of modifying the parameters and technologies downstream of the treatment plant. Since the CAPEX and OPEX of the NF unit were fairly distributed (cf. Fig. 5), it could be beneficial to investigate their main contributors: the costs of the energy system (88% of CAPEX), and quality control and installation (44% of OPEX). Based on the results of our sensitivity analysis, a reduction in the electricity cost and an increase in the market prices of $Mg(OH)_2$ and water will have the maximum impact on LSC reduction. Procuring electricity from a renewable source, especially onshore wind (>1 MW), whose auctioned price in Poland between 2016 and 2019 is as low as ~37% of our assumed electricity price [28], can considerably reduce the LSC. As shown by our sensitivity analysis, a 50% reduction in electricity price brought the difference between the LSC and market price to as low as 14%. The possibility of additional cost reductions could be investigated by using different technology combinations downstream (RO + ED + NaCl crystallization, ED + MED + NaCl crystallization, etc.).

Table 7

Comparison of technology-wise energy consumption at site with the model results (all values in kWh_{el} per m^3 of feed effluent, unless otherwise specified).

Technology	Model	Pilot plant [27]
Nanofiltration 1	2.39	2.251
Nanofiltration 2	1.48	1.681
Reverse osmosis	2.3	2.112
Electrodialysis	2.08	4.654
MED[1]	0.05 kWh_{el} and 9.8 kWh_{th} per m^3 of effluent	0.495 kWh_{el} per m^3 of effluent

[1] The site value for MED is based on literature for a “modern, high-end” MED (12 kWh_{el} per ton of evaporated water)

4. Limitations and areas of future research

Although every attempt was made to model the pilot plant as closely and robustly as possible, addressing the following points can improve the accuracy of the results.

- **Modeling ultrafiltration and decarbonization:** Techno-economic modeling of these two technologies can provide a more accurate estimation of the LSC.
- **Water transport calculation across the ED membrane**: To simplify the calculation of the cell-pair area A_{cp}, we neglected water transportation across the membrane. Although we accounted for the resulting underestimation of the cell-pair area by a correction factor, its value was based on a comparison with an industrial ED operating at a current density of 250 A/m^2, whereas the ED unit in our analysis operated at 400 A/m^2. Using a finite-difference method to model salt and water transportation (as used by Nayar et al. [20]) could estimate the value of A_{cp} more reliably.
- **Two-unit cascade system of ED**: Modeling two ED units working in a cascade system, as in the pilot plant, may help make the replication of the pilot plant in the model more accurate. The modeling of such a system could be possible by performing an optimization instead of a simulation, as used in this study.
- **Modeling transport of non-NaCl ions in ED and RO:** The current models for RO and ED only characterized the transportation of Na^+ and Cl^- ions. Although the presence of other ions at the RO and ED inlets was negligible in this study, characterizing their transport will increase the versatility of the models for a broader range of applications
- **Reducing recovery from the crystallizers:** Since a 100% recovery of minerals is practically not possible, the crystallizer model should ideally include percentage recovery as a parameter, based on experimental results.

5. Conclusion

Coal-mine effluents pose a serious threat to the local environment if left untreated. In this study, we modeled a treatment chain that was tested in an experimental pilot plant. The following technologies were included in our model: NF, two-stage crystallization, RO, ED, MED, and NaCl crystallizer. We found that the recovery of minerals, especially $Mg(OH)_2$ and demineralized water, offered considerable cost benefits. The annual expenditure of the treatment plant was dominated by the NF and RO units, accounting for over 75% of it. The modeled treatment plant was able to replicate the pilot plant’s mineral/water recovery and energy consumption with a fair degree of accuracy. Based on the parameter assumptions, we estimated the levelized cost of salt (LSC) to be 203 USD/ton_{NaCl}, considerably higher than the market value, which range between 80 and 150 USD/ton_{NaCl}. Further investigation into the cost parameters of the highest cost-contributing technologies (NF and RO), use of renewable electricity, and modification in the treatment plant by adding or subtracting a technology could potentially make the treatment of coal-mine effluents economically attractive.

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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Nomenclature

Acronyms

AEM	anion exchange membrane

CEM	cation exchange membrane
CAPEX	annual capital expenditure [USD/year]
ED	electrodialysis
LSC	levelized cost of salt [USD/ton$_{NaCl}$]
MED	multi-effect distillation
NF	nanofiltration
OPEX	annual operational expenditure [USD/year]
RCE	remote component environment
RO	reverse osmosis
TDS	total dissolved solids
TOTEX	total annual expenditure [USD/year]
TSS	total suspended solids
VC	vapor compression
ZB	Zero Brine

Roman symbols

A	area [m^2]
i	current density [A/m^2]
S	salinity [kg$_{salt}$/kg$_{sol}$]
T	temperature [°C]
V	voltage [V]

Subscripts

c	concentrate
cp	cell-pair
d	distillate
el	electrode or electrical
i	inlet
o	outlet
s	steam
th	thermal
tot	total

Appendix

Table A1
Break-up of CAPEX and OPEX for all technologies in the proposed treatment plant (all values in '000 USD/year)

CAPEX component	Value	OPEX component	Value
Nanofiltration (NF1 and NF2)			
Civil	227.1	Energy	164.8
Mechanical (pumps, filters & piping system)	450.8	Chemical	220.5
Electro (energy supply system)	5429.3	Maintenance	122.8
Membrane	323.2	Quality control & daily operation	245.6
Two-step crystallizer			
Crystallizer	12.0	Dolime solution	11.8
Filter	45.2	Electricity	4.0
Reverse Osmosis (RO)			
Structure	169.1	Personnel	19.3
Intake pretreatment	37.2	Maintenance	125.5
Pump	20.7	Electricity	97.9
Site	22.7	Membrane replacement	3.2
Indirect cost	67.4	Chemicals	21.4
Electrodialysis (ED)			
ED plant CAPEX	76.1	Electricity	88.5
-	–	Membrane	76.9
-	–	Maintenance	14.9
-	–	Chemicals	3.6
–	–	Labour	16.7
Multi-effect distillation (MED)			
Evaporator	26.1	Personnel	2.9
Pre-heater	12.9	Maintenance	22.0

(*continued on next page*)

Table A1 (*continued*)

CAPEX component	Value	OPEX component	Value
Condenser	15.6	Electricity	2.1
Flash-box	1.2	Heat	40.3
-	–	Chemicals	0.48
NaCl Crystallizer			
Plant CAPEX	10.6	Electricity	83.5
-	–	Heat	15.9
-	–	Brine disposal	0.003

References

[1] Commission, E., The European Green Deal: Communication from the Commission, 2019, p. 24. Brussels.
[2] EURACOAL, *EURACOAL Annual Report 2019*. 2020, European Association for Coal and Lignite: Brussels.
[3] P. Tsakalides, et al., Smart Water Grids: A Cyber-Physical Systems Approach, CRC Press, 2018.
[4] R.B. Finkelman, A. Wolfe, M.S. Hendryx, The future environmental and health impacts of coal, Energy Geoscience 2 (2) (2021) 99–112.
[5] M. Turek, P. Dydo, R. Klimek, Salt production from coal-mine brine in ED–evaporation–crystallization system, Desalination 184 (1–3) (2005) 439–446.
[6] R.K. Tiwary, Environmental impact of coal mining on water regime ands its management, Water Air Soil Pollut. 132 (2001) 185–199.
[7] EU, E.P.a.t.C.o.t., Directive 2000/60/EC: Establishing a Framework for Community Action in the Field of Water Policy, E.P.a.t.C.o.t. EU, 2000.
[8] Commission, E. *WFD*: Timetable for implementation. Available from: https://ec.europa.eu/environment/water/water-framework/info/timetable_en.htm.
[9] Commission, E., Critical Raw Materials Resilience: Charting a Path towards Greater Security and Sustainability, 2020, p. 23.
[10] B. Ericsson, B. Hallmans, Treatment of saline wastewater for zero discharge at the Debiensko coal mines in Poland, Desalination 105 (1996) 115–123.
[11] M. Turek, P. Dydo, R. Klimek, Salt production from coal-mine brine in NF — evaporation — crystallization system, Desalination 221 (1–3) (2008) 238–243.
[12] (EU), E.U. Zero Brine project. EU Horizon 2020 Innovation Action (IA) 2021; Available from: https://zerobrine.eu/.
[13] D. Xevgenos, et al., Redesigning the Coal Mine Wastewater Treatment to Enable Transition to Circular Economy Practices: the ZERO BRINE and Dębieńsko Case Studies, 2021, pp. 1–22.
[14] K. Mitko, et al., Electrodialysis of Coal Mine Water, 25, Water Resources and Industry, 2021.
[15] M. Micari, et al., Techno-economic analysis of integrated processes for the treatment and valorisation of neutral coal mine effluents, J. Clean. Prod. 270 (2020), 122472.
[16] M. Micari, et al., Combined membrane and thermal desalination processes for the treatment of ion exchange resins spent brine, Appl. Energy 254 (2019), 113699.
[17] M. Micari, et al., Techno-economic assessment of multi-effect distillation process for the treatment and recycling of ion exchange resin spent brines, Desalination 456 (2019) 38–52.
[18] M. Micari, et al., Towards the implementation of circular economy in the water softening industry: a technical, economic and environmental analysis, J. Clean. Prod. 255 (2020), 120291.
[19] (DLR), D.Z.f.L.-u.R.e.V. RCE, Available from: https://rcenvironment.de/, 2021.
[20] K.G. Nayar, et al., Cost and energy requirements of hybrid RO and ED brine concentration systems for salt production, Desalination 456 (2019) 97–120.
[21] R.K. McGovern, et al., On the cost of electrodialysis for the desalination of high salinity feeds, Appl. Energy 136 (2014) 649–661.
[22] K.M. Chehayeb, et al., Optimal design and operation of electrodialysis for brackish-water desalination and for high-salinity brine concentration, Desalination 420 (2017) 167–182.
[23] Y. Tanaka, et al., Ion-exchange membrane electrodialytic salt production using brine discharged from a reverse osmosis seawater desalination plant, J. Membr. Sci. 222 (1–2) (2003) 71–86.
[24] A. Chemicals, Selemion ion exchange membranes, in: A. Chemicals (Ed.), Model: DW Type 3E, 2013. Japan.
[25] R. Turton, et al., Analysis, Synthesis and Design of Chemical Processes, Pearson Education, 2008.
[26] K. Mitko, in: K. Panteleaki (Ed.), Email Communication about Resource Recovery in Polish Pilot Plant, 2021.
[27] K. Mitko, in: N.D. Pawar (Ed.), Email Communication about Energy Consumption in Polish Pilot Plant, 2021.
[28] J. Paska, et al., Electricity generation from renewable energy sources in Poland as a part of commitment to the polish and EU energy policy, Energies 13 (16) (2020).