

## Electrodialysis and electro dialysis reversal for the concentration of Li containing solutions

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### ABSTRACT

In this paper, the application of electro dialysis reversal (EDR) in the recovery and concentration of lithium chloride and lithium carbonate solutions is described. Both salts are raw materials that can be used for the production of lithium ion batteries. EDR is characterized by a periodical change of electric polarity and hydraulic flows. This allows the minimization of scaling effects and concentration polarization on the membrane surface during the treatment of lithium containing brines or solutions stemming from lithium ion battery recycling. Lab scale investigations at room temperature were conducted during which the concentrated solution was kept in the cycle throughout the respective investigations, and the diluted solution was exchanged, when a minimum conductivity of 0.08 mS/cm was reached. At the same time, the polarity was changed and the hydraulic flows were reversed. A comparison between EDR and conventional ED at the same operating conditions showed similar current efficiencies of up to 70 % during the concentration of lithium chloride. A maximum lithium chloride concentration in the concentrate solution of about 25 g/l was achieved. The investigations showed the general applicability of EDR for the concentration of lithium chloride and lithium carbonate, which has a restricted solubility in water. Generally, EDR could play a major role in lithium ion battery recycling.

**Keywords:** Circular economy, ED, EDR, ion exchange membranes, Li recovery

## 1 INTRODUCTION

Lithium has become one of the most important metals in the world. The projected demand for lithium will more than triple from 685 tonnes in 2023 to 2114 tonnes in 2030 [1].

Today, more than 80% of the lithium mined worldwide is consumed in battery production [2].

Next to the demand of the battery industry with an annual growth >10%, the demand of lithium based lubricants is growing at about 3.5% annually, and ceramics and glass industry demand by 3-5% annually [3].

Despite proven lithium sources in Finland [4], Poland, Portugal and Germany [5], an independent supply of lithium to Europe seems to be impossible in view of the increasing demand.

The limited availability of lithium reserves in Europe therefore necessitates the use of alternative sources,

especially secondary raw materials, to ensure a sustainable supply chain and reduce dependence on lithium-rich regions.

Electronic waste (spent lithium-ion batteries LIBs) and production water from the oil and gas industry offer compelling opportunities to diversify sources of lithium recovery and mitigate the environmental impacts associated with primary lithium production, s. [6].

The composition of LIBs currently used in eMobility is depicted in table 1.

**Table 1. Typical composition of lithium-ion battery cathodes**

Li	Co	Ni	Cu	Graphite
wt.% [7], [8], [9]				
1.2-2.0	22.7-17.3	1.2-14.8	17.3-16.6	12.5

Since the production of lithium containing products in industry is mainly based on lithium hydroxide (LiOH) or lithium carbonate (Li<sub>2</sub>CO<sub>3</sub>), recycling technologies must be aimed at the production of these compounds.

Methods for lithium recovery from LIBs investigated to date include mechanical pretreatment and a combination of pyrometallurgical, hydrometallurgical, bio-hydrometallurgical and electrochemical processes [10].

Mechanical pre-treatment is used to separate Li rich cathode materials from e.g. pantographs and binders. The aim is to reduce contamination and to facilitate the subsequent Li recovery [11]. In most cases, the first treatment step is the comminution of the battery cases: Wang et al. [12] describe different shredding technologies that may be applied. Zhang [13] used the combination of a shear crusher followed by an impact crusher. Liu et al. [14] investigated lab scale cryogenic grinding with liquid nitrogen.

The received mixture of particles requires separation. To this end froth flotation was investigated by Liu [14], while Yu et al. describe the application of grinding flotation [15]. Other alternatives are e.g. gravity separation [16] and magnetic separation [17].

An overview of the advantages, disadvantages and future challenges of the different approaches for Li recovery from LIB is depicted in table 2.

**Table 2. Advantages, disadvantages and future challenges of the different approaches for Li recovery from LIB**

Approach	Advantages after [9], [18]	Disadvantages
Hydrometallurgy	High recovery rate	Generation of additional wastewater
	High product purity	Application of additional agents
	Low energy demand	Post-treatment of the aqueous phase required, e.g. precipitation
	High selectivity	
	Well known technology in mineral processing	

Pyrometallurgy	Simple processes with high capacity	High energy demand
	High efficiency	Low recovery efficiency
		Waste gas formation (including CO <sub>2</sub> )
		High costs of waste air treatment
Direct physical recycling	Short process chain	High demands on the process and the equipment
	Low energy demand	High demands on the spent LIB characteristics
	Environmentally friendly	
	High recovery rate	Incomplete recovery

Due to the limitations of the conventional Li recovery processes, the application of electro dialysis (ED) is suggested:

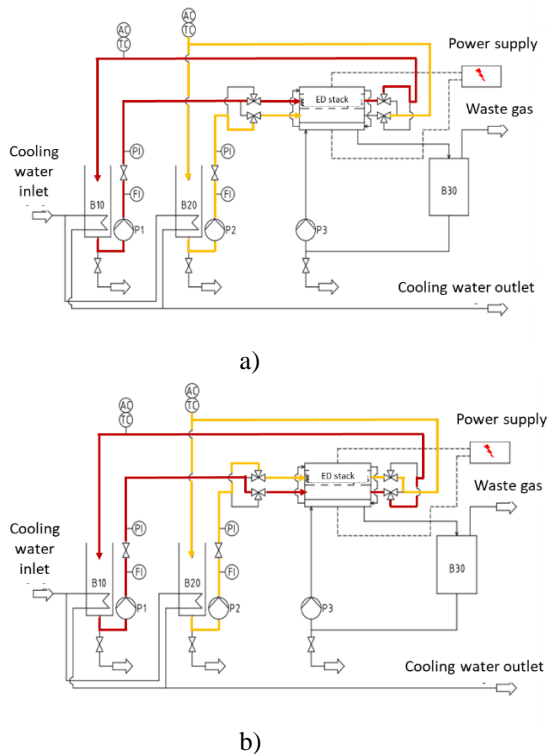
A recent study conducted by Gmar et al. revealed that - due to its monovalent-selective characteristics - a Neosepta® exchange membrane had higher selectivity for Li<sup>+</sup> over divalent cations, while AGC's CSO membrane showed similar selectivity for cobalt<sup>2+</sup> and nickel<sup>2+</sup>. The transport rate of Li<sup>+</sup> through the Neosepta membrane was higher compared to AGC's CSO membrane, and the flux of divalent cations was significantly lower [19]. Also other authors describe the transport behavior of ions in membranes using lithium-cobalt based solutions: Villen-Guzman et al. [20] stress the importance of different operating parameters such as pH, current density, and membrane type. Iizuka [21] describes the transport behavior of bipolar membranes, while Afifah et al. [22] investigated the influence of monovalent selective membranes.

A recent research paper describes the behavior of PC Cell's PCSK and PCSA ion exchange membranes during concentrating of lithium-bearing saline solutions using conventional ED [23].

However, ED performance can be affected by deposits and contaminations of the membranes [24]. E.g., Gmar et al. observed precipitation of solid particles within ion exchange membranes, especially with rising ratio between divalent metal ions and Li<sup>+</sup> ions [19].

The reported membrane scaling makes electro dialysis reversal (EDR) - a variant of conventional ED - an attractive option for the concentration of lithium containing solutions, such as those derived from battery recycling. EDR refers to the simultaneous reversal of the electric field and the simultaneous change of concentrated and diluted solution in the membrane stack. This way,

depleted scalants are removed periodically from the membrane surface. Figure 1 depicts the flow chart of an EDR plant in conventional mode (a) and reversal mode (b).



**Figure 1. Flow chart of EDR; a) prior to reversal of the hydraulic streams, b) after reversal of the hydraulic streams**

A disadvantage of this procedure is that during the switch from conventional mode to reversal mode, mixed solutions of diluate and concentrate are generated (off-spec). These could be removed using additional valves. Furthermore, the influence of the mixing on the total performance can be reduced by minimizing the volume of the respective pipes connected to the membrane stack.

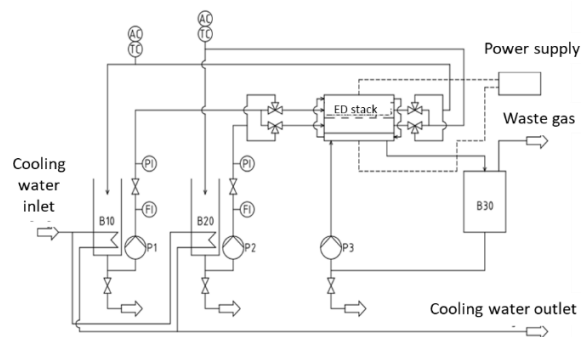
## 2 MATERIALS AND METHODS

### 2.1 Lab scale ED plant

The general setup of the applied ED plant as well as some aspects on the experimental background were described before elsewhere [23]. The investigations were conducted using a lab-scale test plant type BED1-3 provided by PCCell GmbH, Heusweiler, Germany that was converted to reverse operation. This includes the installation of three-way valves to change the direction of flow inside the membrane stack when the electric polarity of the electrodes is changed. The valves are operated manually, whereas the change of polarity is initiated through the control systems of the test plant.

The test plant comprises three tanks for the process media (diluate, concentrate, and electrode rinse solution),

a membrane stack, a DC supply, centrifugal pumps and downstream filters, PVDF tubes for the transport of the media, conductivity meters, and manual valves for the reversal of the hydraulic streams (s. Figure 2).



**Figure 2. Flow chart of the lab scale system BED1-3 after conversion to EDR mode**

To reduce mixing of diluate and concentrate after polarization reversal and reversal of the hydraulic streams, a minimum length of the tubes that connect the valves with the membrane stack, was chosen.

To keep the temperatures of diluate and concentrate solutions at  $22 \pm 0.5$  °C, the respective tanks are equipped with cooling spirals connected to a thermostat.

### 2.2 Membrane stack

An ED module type ED 64004 from PCCell GmbH, Heusweiler, Germany with following characteristics was used, see also [23]:

- Effective membrane area ( $A_{\text{eff}}$ ) = 64 cm<sup>2</sup>;
- 10 cell pairs, consisting of 9 PcSK membranes (strongly acidic non-monovalent cation exchange membrane), 10 PcSA membranes (strongly acidic anion exchange membrane), and 2 PcMPE membranes adjacent to the electrode chambers;
- Spacer thickness ( $d_{\text{spacer}}$ ) = 0.45 mm.

### 2.3 Measurement equipment

In the diluate and concentrate cycle, JUMO CTI-500 conductivity and temperature transmitters are included, each having tolerances of  $\leq 0.5\%$ .

For the preparation of the electrode rinse solution and the respective Li containing feed solutions, a multiparameter meter type HI 9829 from Hanna Instruments was used with an accuracy of  $\pm 1\%$ .

A scale type PM4800 DeltaRange from Mettler (Max. 4.100 g/800 g; d = 0.1 g/0.01 g) was used for weighing the lithium salts.

For the preparation of the aqueous lithium salt solutions, a volumetric flask with a volume of 5.000 mL ( $\pm 1.2$  mL) was used.

## 2.4 Chemicals

For the preparation of synthetic solutions, following chemicals were used:

- Lithium chloride ( $\geq 98.5\%$ , pure), Carl Roth GmbH + Co. KG;
- Lithium carbonate, ( $\geq 99\%$  pure), Carl Roth GmbH + Co. KG;
- Electrode rinse: Sodium sulfate ( $\geq 99\%$ , anhydrous), Carl Roth GmbH + Co. KG;
- Demineralized water 9  $\mu\text{S}/\text{cm}$  (mixed bed ion exchanger, TH Köln).

## 2.5 Operation mode

Concentrate and diluate tank were initially filled with approx. 920 ml ( $\pm 50$  ml) of the same synthetic solution. During operation, the diluted solution was exchanged, when the minimum conductivity reached 0.08 mS/cm. The concentrated solution remained in the concentrate cycle during each measurement series. Polarity reversal and the reversal of the hydraulic streams was done each time the diluate solution was exchanged.

Material flows of diluate and concentrate were set at 25 l/h to 35 l/h.

The selected voltage of 20 V and 30 V, respectively, was based on previous experiments with conventional ED, and the selected maximum current was 0.51 A [21].

## 2.6 Correlation of conductivity and concentration

The measured conductivity of the synthetic LiCl and  $\text{LiCO}_3$  solutions was converted to molar concentrations based on calibration functions obtained in preliminary examinations using the multiparameter meter type HI 9829.

## 2.7 Plant performance

The description of the plant performance is based on the following equations.

The electric charge  $C$  refers to the current  $I$  that is applied for the ionic transport within the membrane stack for a given period  $\Delta t$ . It is calculated according to equation (1), s. [24].

$$C = \int_0^t I(t) dt = \bar{I} * \Delta t [As] \quad (1)$$

$\Delta t$  period [s]  
 $\bar{I}$  average current [A] during the period  $\Delta t$ .

Current efficiency (CE)  $\eta$  is calculated through Faraday's law as the actual transport of ions divided by the theoretical one, s. equation (2). Undesired by-effects, such as chemical reactions at the electrodes, heating of the solutions, or leakage current, reduce the current efficiency [24].

$$\eta = \frac{z * F * n}{N * \sum_{t=0}^t (I_t * \Delta t)} * 100\% [\%] \quad (2)$$

$z$  charge of the respective ion [-]

$n$  amount of transported ions [moles]  
 $F$  Faraday's constant [96,485 C/mol]  
 $N$  cell pair number [-]  
 $t$  considered period [s].

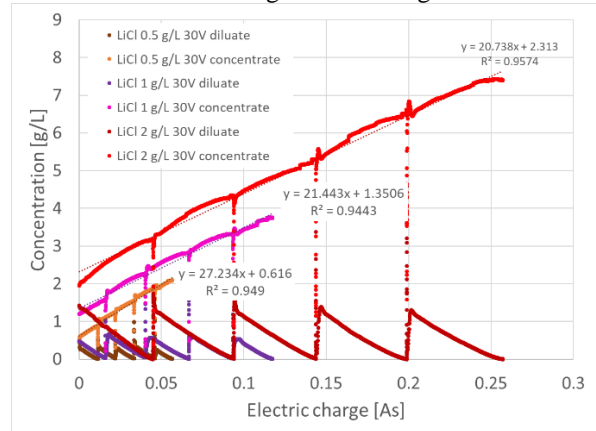
## 3 INVESTIGATIONS OF EDR BASED ON SYNTHETIC LI SALT SOLUTIONS

In a first series, EDR performance of was estimated using different initial concentrations of both, LiCl and  $\text{Li}_2\text{CO}_3$  at a maximum voltage of 30 V and a temperature of ca. 22 °C. During each set of investigations, the depleted diluate solution was exchanged 5-6 times.

In a second series, the performance behavior of LiCl during long-term trials was investigated. The initial concentration of diluate solution and concentrate solution  $c_0$  was 3 g/L and the maximum voltage was 30 V, while the temperature was kept at ca. 22 °C.

### 3.1 Influence of the initial LiCl concentration $c_0$ on the concentrate concentration

Figure 3 depicts the concentrations in diluate and concentrate with increasing electric charge.



**Figure 3. Concentrations of LiCl solutions during EDR @ 22 °C and 30 V as a function of the electric charge. Initial concentrations  $c_0$ : 0.5 g/L; 1 g/L; 2 g/L**

Linear regression was used to calculate the slope of the concentrate curve progressions. In all cases, the correlation coefficient  $R^2 > 94\%$ . Deviations from the linear functions may be explained by fluctuations of the temperatures within the membrane stack ( $\pm 0.5$  °C) and fluctuations of the volume flow that may have an influence on the ion concentrations in the streaming boundary layer next to the membranes.

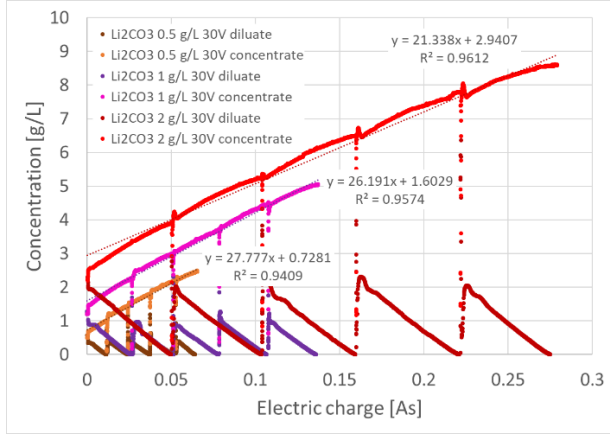
Furthermore, the current efficiencies of the successive 4 batches were calculated, s. table 3.

**Table 3. Current efficiency (CE) @ 22 °C and 30 V with varying initial concentration of LiCl**

	Feed $c_0 = 0.5 \text{ g/L}$	Feed $c_0 = 1 \text{ g/L}$	Feed $c_0 = 2 \text{ g/L}$
CE 1 <sup>st</sup> batch	90.3	59.3	62.4
CE 2 <sup>nd</sup> batch	79.9	55.3	62.8
CE 3 <sup>rd</sup> batch	57.9	49.3	56.3
CE 4 <sup>th</sup> batch	62.8	42,7	57.2
CE total (1-4)	72.7	51.6	59.7

### 3.2 Influence of the initial $\text{Li}_2\text{CO}_3$ concentration $c_0$ on the concentrate concentration

Figure 4 depicts the concentrations in diluate and concentrate as a function of the electric charge.



**Figure 4. Concentrations of  $\text{Li}_2\text{CO}_3$  solutions during EDR @ 22 °C and 30V. Initial concentrations  $c_0$ : 0.5 g/L; 1 g/L; 2 g/L as a function of the electric charge**

Linear regression was used to calculate the slope of the concentrate curve progressions. In all cases, the correlation coefficient  $R^2 > 94\%$ . Also here, deviations from the linear functions may be explained by temperature fluctuations and fluctuations of the volume flow.

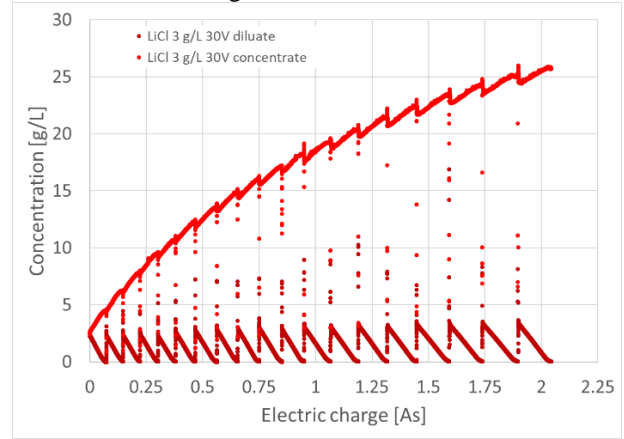
Furthermore, the current efficiencies of the successive 4 batches were calculated, s. table 4.

**Table 4: Current efficiency (CE) at 22 °C and 30 V with varying initial concentration of  $\text{Li}_2\text{CO}_3$**

	Feed $c_0 = 0.5 \text{ g/L}$	Feed $c_0 = 1 \text{ g/L}$	Feed $c_0 = 2 \text{ g/L}$
CE 1 <sup>st</sup> batch	60.2	51.8	44.8
CE 2 <sup>nd</sup> batch	38.2	41.4	33.2
CE 3 <sup>rd</sup> batch	33.8	37.0	33.4
CE 4 <sup>th</sup> batch	32.5	38.2	32.9
CE total (1-4)	41.2	42.1	36.1

### 3.3 Longterm investigation of EDR using a synthetic LiCl solution

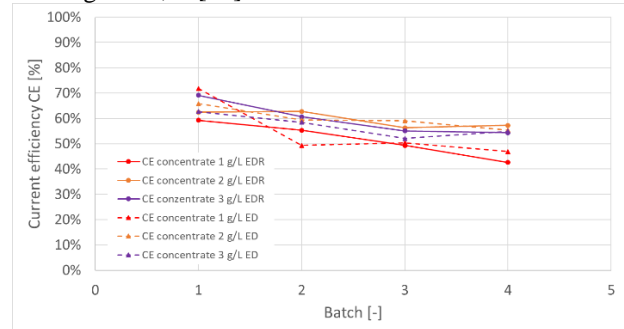
Figure 5 depicts the concentrations in diluate and concentrate as a function of the electric charge during the longterm investigations. During this period, the diluate solution was exchanged 19 times.



**Figure 5. Concentration of a synthetic LiCl solution ( $c_0=3 \text{ g/L}$ ) during EDR as a function of the electric charge  $C$  during longterm investigations (13.5 h)**

### 3.4 Comparison of EDR and ED using a synthetic LiCl solution

Figure 6 depicts the comparison of the current densities of EDR and conventional ED at the same experimental conditions during 4 diluate batches. The values of the ED experiments were achieved during previous investigations, s. [21].



**Figure 6. Comparison of the current efficiencies of ED and EDR using synthetic LiCl solutions (initial concentration  $c_0 = 1, 2$  and  $3 \text{ g/L}$ )**

## 4 DISCUSSION

### 4.1 Influence of the initial LiCl and $\text{Li}_2\text{CO}_3$ concentration $c_0$ on the concentrate concentration

Basically, the linear approach for the concentration over the electric charge is in line with Faraday's law. From figures 3 and 4 it becomes apparent that with increasing initial concentration, the slope of the linear approach becomes flatter. This can be explained as follows: At higher salt concentration, the conductivity of the solution

risers. This leads to a higher current and accordingly to a higher transport rate of (solvated) ions. As a result, the actual concentration of Li ions is growing slower. To a certain extent with increasing electrical charge also back diffusion due to the rising concentration gradient between concentrate and diluate solution can be expected, an effect that also leads to a slower growth of the ion concentration in the concentrate.

Accordingly, two trends are visible from table 3 and 4: Current efficiency decreases with increasing initial concentration  $c_0$  of the solutions and with progressive batch number, as the ionic water transport leads to a comparably lower salt concentration in the concentrate solutions.

The decreasing current efficiency in subsequent batches can be attributed to the rising concentration difference between diluate and concentrate solution that may lead to a back diffusion of ions to the diluate. Hence, back diffusion works against the applied current.

#### **4.2 Longterm investigation of EDR using a synthetic LiCl solution**

During the long-term investigations of EDR, a maximum concentrate concentration >25 g/L LiCl was achieved, when the initial concentration was 3 g/L. The resulting concentrate concentration as a function of the electric charge cannot be approximated any longer by a linear function. Instead, the measured values tend to reach a steady state. The flattening of the concentrate concentration with increasing electric charge can be attributed to ionic water transport from the diluate into the concentrate solution. This is in line with the findings of previous investigations done in the authors' lab [21].

#### **4.3 Comparison of EDR and ED using a synthetic LiCl solution**

In both set-ups – conventional ED and EDR - the current efficiency is decreasing with the number of batches; the curve progression shows comparable gradients at any given initial concentration, although the measured values are fluctuating. Differences between ED and EDR performance can be explained by slightly differing streaming velocities in the stack that lead to differences in concentration polarization at the membrane surface.

## **5 CONCLUSIONS AND FUTURE RESEARCH**

Generally, electrodialysis reversal (EDR) offers advantages over conventional electrodialysis (ED), e.g. the reduction of scaling effects and concentration polarization, when solutions with a high scaling potential should be treated. However, an influence on the plant performance can be expected due to fluctuations of operation parameters and backmixing, when polarity is reversed and material flow directions are changed. The

investigations showed that current efficiency of EDR becomes comparable to conventional ED, when this influence is minimized.

Investigations on the concentration of lithium carbonate  $\text{Li}_2\text{CO}_3$  that has a lower solubility compared to lithium chloride  $\text{LiCl}$ , could be carried out successfully. Even, when the concentration was above saturation limit (13.3 g/L at 20 °C), no scaling and no precipitation was detected. Precipitation occurred only after several days in the resting solution. The studies with  $\text{Li}_2\text{CO}_3$  show that EDR is suitable for this purpose. Further investigations to prevent  $\text{Li}_2\text{CO}_3$  precipitation involve pH adjustment during the concentration process.

In addition, further investigations should be carried out with regard to the actual saturation limit, since in the case of crystalline precipitation of  $\text{Li}_2\text{CO}_3$ , a filter system can be installed downstream or combined with precipitation in order to obtain the  $\text{Li}_2\text{CO}_3$  directly in crystalline form for further processing, without the need for energy-intensive drying processes.

In addition, further investigations should be carried out with regard to the actual saturation limit, since in the case of crystalline precipitation of  $\text{Li}_2\text{CO}_3$ , a filter system can be installed downstream or EDR can be combined with a precipitation step in order to obtain solid  $\text{Li}_2\text{CO}_3$  directly.

However, higher current efficiency was achieved during the concentration of  $\text{LiCl}$  compared to  $\text{Li}_2\text{CO}_3$  solutions.

In long-term tests with  $\text{LiCl}$  solution, a maximum concentration of 825% compared to the initial value could be achieved.

Possibilities for improving the performance of the existing laboratory EDR system include the optimization of the streaming velocity in the stack by using e.g. larger pumps. Additionally, the applied voltage and current should be adapted to the respective medium. This should be done based on the limiting current density which needs to be investigated.

An important next step are EDR investigations using real, complex industrial feed solutions, such as brines and lithium containing solutions from battery recycling. Especially, two general problems need to be solved: the separation of sodium ions, which may be present at high concentrations and the removal of divalent cations, such as cobalt<sup>2+</sup> and nickel<sup>2+</sup>.

Another challenge will be the scale up the process. Here, the challenges are related to the implementation of an appropriate pre-treatment, the overall plant design (reducing the so-called off-spec solution that is generated when the hydraulic streams are changed), the stack design (material and structure of the electrodes, reduced leakage current), and membrane fouling during long-term operation.

## 6 DECLARATIONS

**Author declaration:** The authors declare that the work submitted has not been published before and is not under consideration for publication elsewhere. All authors have read and agreed to the published version of the manuscript

**Author contributions:** Conceptualization, C.M. and F.R.; practical investigation, C.M.; methodology, C.M. and F.R.; supervision, F.R.; visualization, C.M. and F.R.; writing of the original draft, C.M. and F.R.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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