Supplementary Information

Pilot-scale demonstration of an electrochemical system

for lithium recovery from the desalination concentrate

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Experimental details

The electrochemical monopolar-type stack cell based on 14 pairs of λ -MnO₂ and Ag electrodes was used in the primary recovery process. Rectangular graphite plates with 200 mm length, 100 mm width, and 2 mm thickness were used as current collectors. λ -MnO₂ electrodes were prepared by the acid treatment of LiMn₂O₄ composite electrodes in 0.5 M of HCl solution for 1 hour. LiMn₂O₄ composite electrode was fabricated by the following steps. First, 80 wt % LiMn₂O₄ (TOB New Energy, China) as the active material, 10 wt % Super P (Timcal, Switzerland) as the conductive material, and 10 wt % polyvinylidene fluoride (PVDF, Sigma-Aldrich) as the binder were mixed with the 1-methyl-2-pyrrolidone (NMP, Sigma-Aldrich) solution and the slurry was made. Then, the slurry was casted on the current collector with a thickness of 300 mm, followed by the solvent evaporation at the temperature of 120 °C in the vacuum oven. Ag electrodes were prepared as the following steps. 80 wt % silver powder (Ag, Sigma-Aldrich) as the active material, 10 wt % Super P (Timcal, Switzerland) as the conductive material, and 10 wt % polytetrafluoroethylene (PTFE, Sigma-Aldrich) as the binder were mixed with the ethanol (99%, Sigma-Aldrich) and the slurry was made. The slurry was rollpressed with a thickness of 300 mm and dried at the temperature of 120 °C in the vacuum oven. After the solvent evaporation, Ag electrodes were attached onto the current collector with the carbon paint (TED PELLA INC., USA). The electrochemical cell used in the secondary recovery process was comprised of single electrode pair of the λ -MnO₂ and Ag electrode. As a current collector, circular graphite sheets with 100 mm diameter and 0.2 mm thickness were used. The electrodes used in this process were fabricated in a similar way to the previous one.

Physicochemical characterization and operation of the electrochemical cell were conducted by the battery cycler (WBCS3000; WonATech, Korea) and the automated program (CIMON, Korea). The cation concentrations of the intermediate product and the final product were measured by the ion chromatography (IC-1100; Thermo-fisher, United states). A pH meter (Lab 860, SI Analytics GmbH) and a conductivity meter (F-74BW, Horiba) were used to characterize the desalination concentrate. TDS and element composition of the desalination concentrate were calculated from the chloride concentration measured by the Ion chromatography (DX-120, Thermo Fisher Scientific Inc.) and composition of standard seawater¹.



Fig. S1 Overview and specification of the pilot-scale electrochemical lithium recovery system.



Fig. S2 (a) The electrode stack used in the primary recovery process and (b) the electrodes comprising the stack. (c) The electrochemical cell used in the secondary recovery process and (d) the electrode comprising the cell

| Measured properties | | |
|---------------------|---------------|-------|
| рН | 8.0 | |
| Conductivity (S/m) | 6.53 | |
| TDS (mg/L) | 43895 | |
| Composition | | |
| Compound . | Concentration | |
| | mg/L | mM |
| Chlorine | 25246 | 712 |
| Sodium | 14463 | 629 |
| Magnesium | 1732 | 71 |
| Sulfur | 1215 | 38 |
| Calcium | 553 | 14 |
| Potassium | 536 | 14 |
| Bromine | 90 | 1 |
| Carbon | 38 | 3 |
| Strontium | 11 | 0.121 |
| Boron | 6 | 0.559 |
| Silicium | 3 | 0.096 |
| Fluorine | 2 | 0.092 |
| Lithium | 0.24 | 0.035 |
| Nitrogen | 0.20 | 0.014 |
| Rubidium | 0.16 | 0.002 |
| Phosphorous | 0.08 | 0.003 |
| Iodine | 0.08 | 0.001 |

Table S1 Characterization of the desalination concentrate



Fig. S3 Cyclic voltammogram of λ -MnO₂ electrode (solution: 1 M LiCl, scan rate: 1 mV/s)

Reference

1 S. A. Gerlach, *Marine Pollution*, Springer Berlin Heidelberg, Berlin, Heidelberg, 1st edn., 1981, ch. 1, pp. 1-5.