Supplementary Information

A novel method for screening deep eutectic solvent to recycle cathode of Li-ion batteries

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Deep Eutectic Solvent Recyclability and Selectivity

The electrodeposition technique is a common method to recycle metals from the loaded DES apart from chemical precipitation. The three-electrode system (in section: **Analysis of DES and leachate**) at cathodic potential of -0.9 V and at 100˚C for 10 hours was employed to recycle metal from the DES sample that digested $LiCoO_2$ (LCO) at 140˚C. The loaded DES after electrodeposition became from blue to yellow (**Fig. S8b** and **S8c**). The similar yellow ChCl : urea DES was obtained from colorless ChCl : urea DES after cyclic voltammetry.[1] The yellow ChCl : urea DES was extracted metals from LCO powder and its color became bluish-green containing cobalt of 0.459 g L^{-1} as similar as original DES at 140˚C (**Fig. S8d**). That indicated DES can be renewable in the process of LIBs recycle.[2]

Dismantled cathode materials of spent LIBs (LCO) were sieved by a 100 mesh sieve (LCO-1) and 120 mesh sieve (LCO-2). The extraction results were shown as **Fig. S9**. The extraction efficiency of Li and Co from spent LCO was close to that from LCO powder. The extraction of Al was weak by using ChCl : urea DES from spent LCO. That indicates that the DES could also be used to recycle spent LIBS.

Three kinds of $LiNi_xCo_yMn_zO_2$ (NCM) powder were employed, where ratios of x:y:z were 811, 532 and 111. The extraction results were shown as **Fig. S10**. The extraction efficiency of Li, Ni, Co and Mn from NCM powder was about 90%. That indicated ChCl : urea DES could also be used to recycle NCM powder.

Figures and Tables:

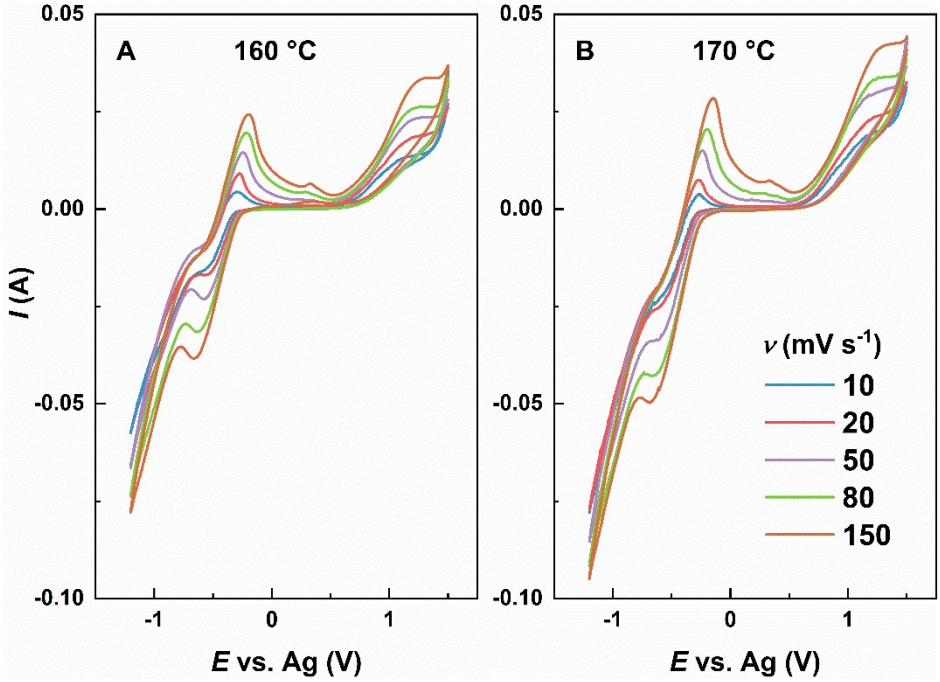

Fig. S1 Cyclic voltammetry behavior of ChCl : urea DESs at various scan rates and 160 °C and 170 °C.

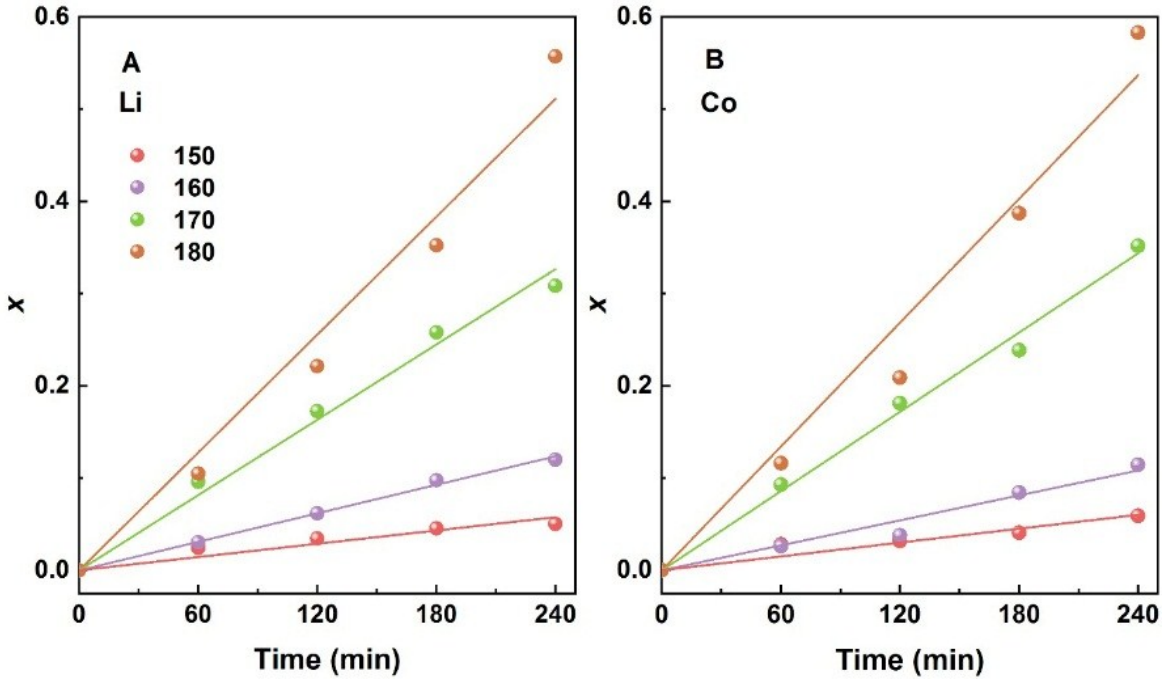

Fig. S2 Solution diffusion control model for kinetics of Li (A) and Co (B) extraction.

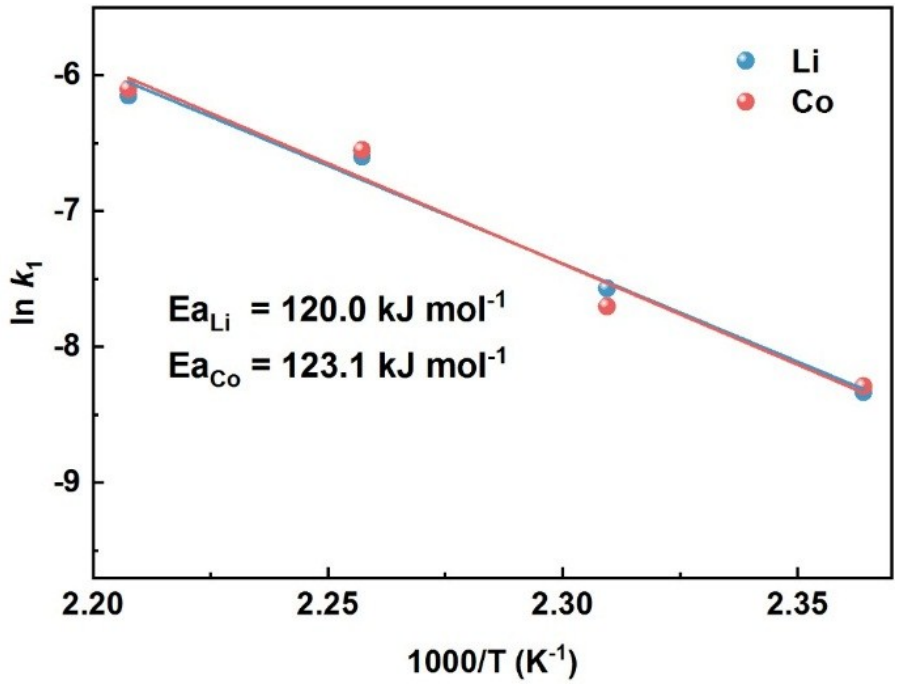

Fig. S3 Arrhenius plot for Li and Co extraction in series of temperatures (423 – 453 K).

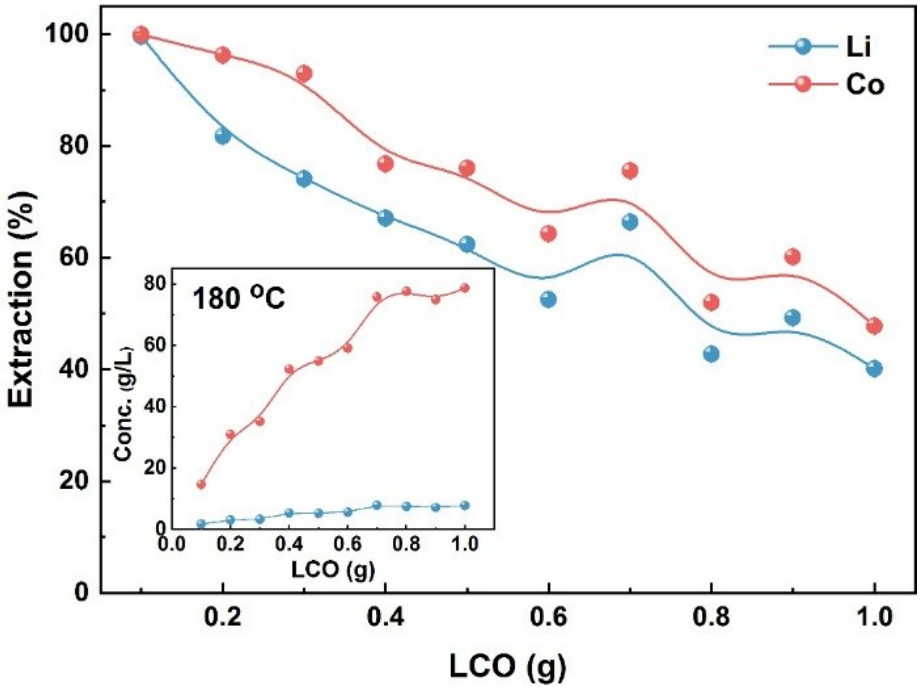

Fig. S4 Extraction and concentation of Li and Co by using 5 g ChCl : urea DES under various mass of LCO and 180 °C.

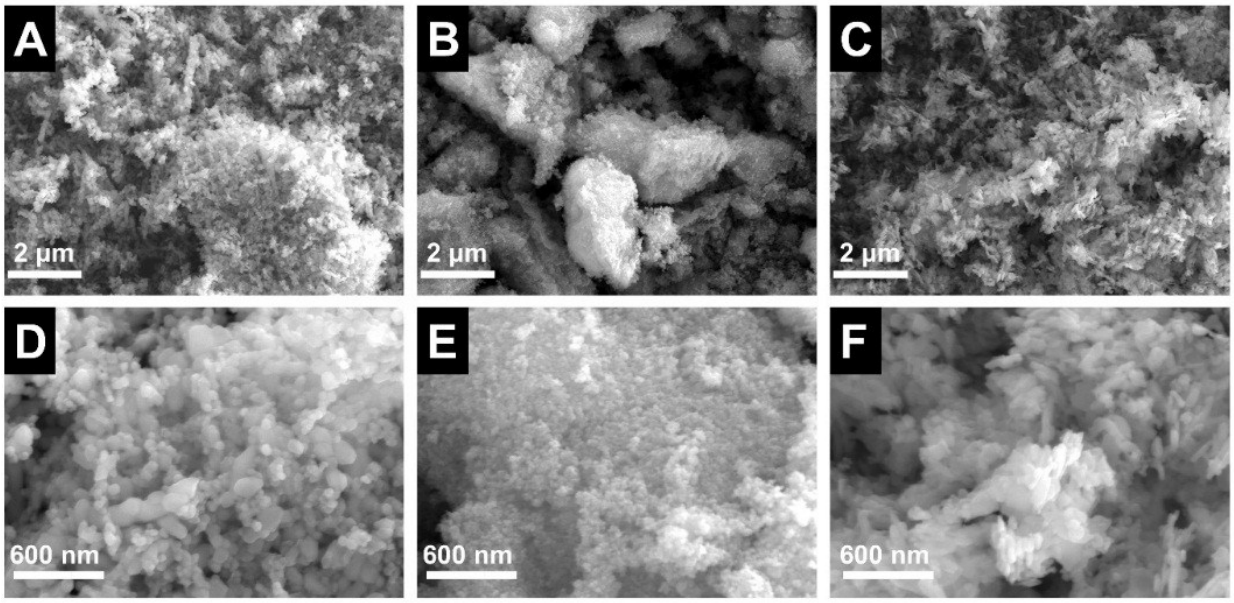

Fig. S5 FESEM images of the calcined powder after deionized water dilution.

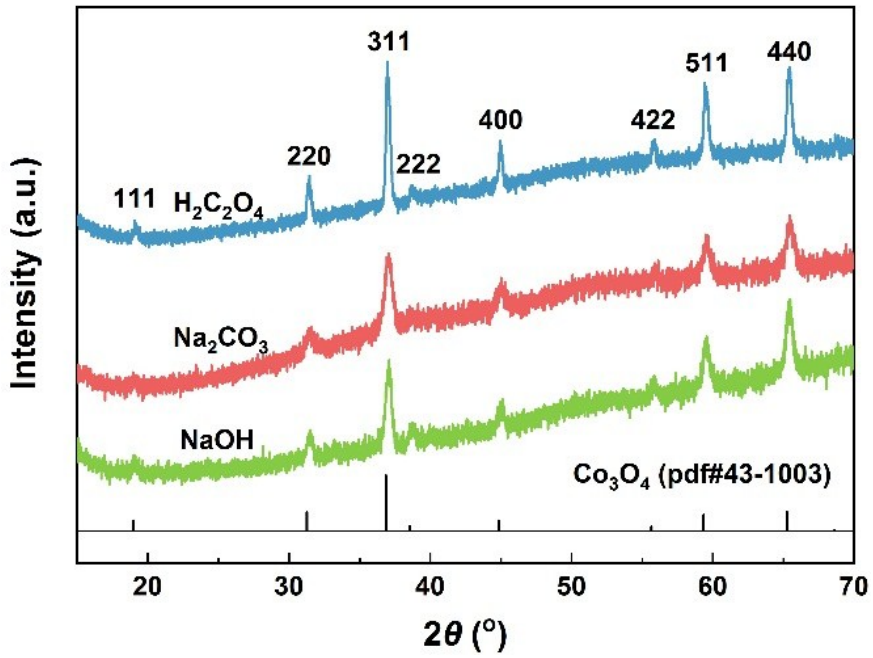

Fig. S6 XRD spectra of the calcined powder after deionized water dilution.

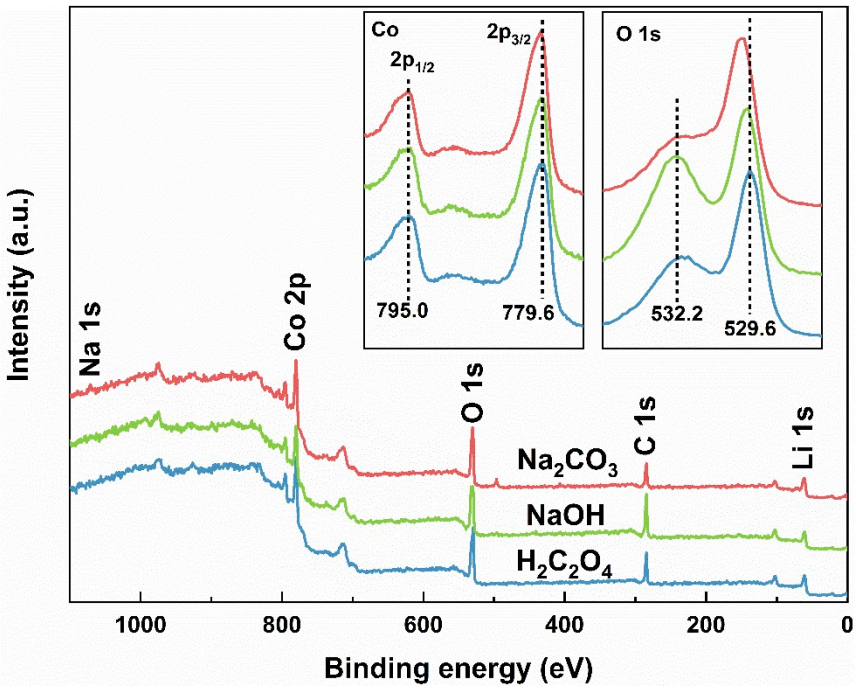

Fig. S7 XPS spectra of the calcined powder after deionized water dilution.

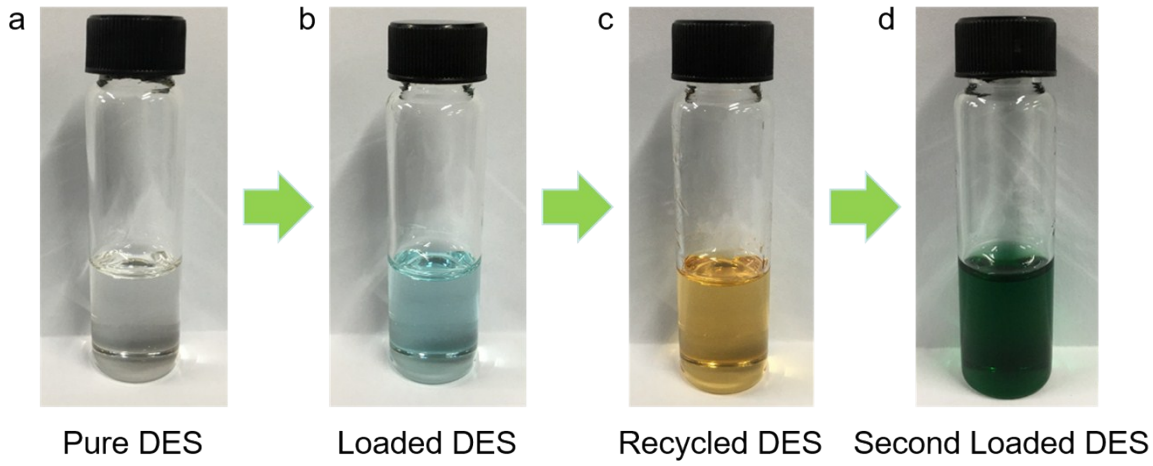

Fig. S8 Recyclability of ChCl : urea DES.

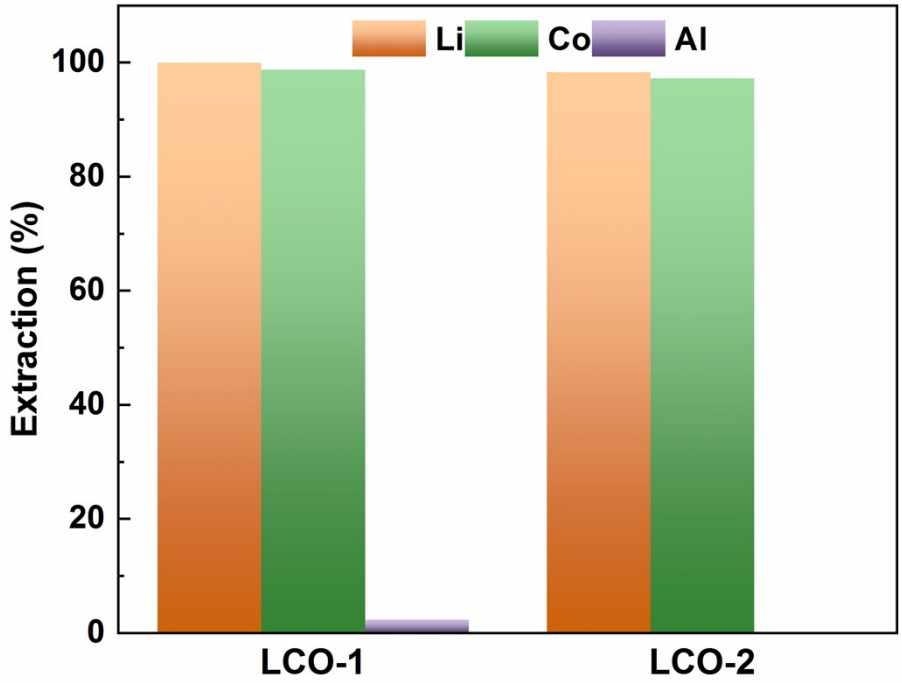

Fig. S9 Extraction of Li, Co and Al by using ChCl : urea DES at 180 °C and 15 h

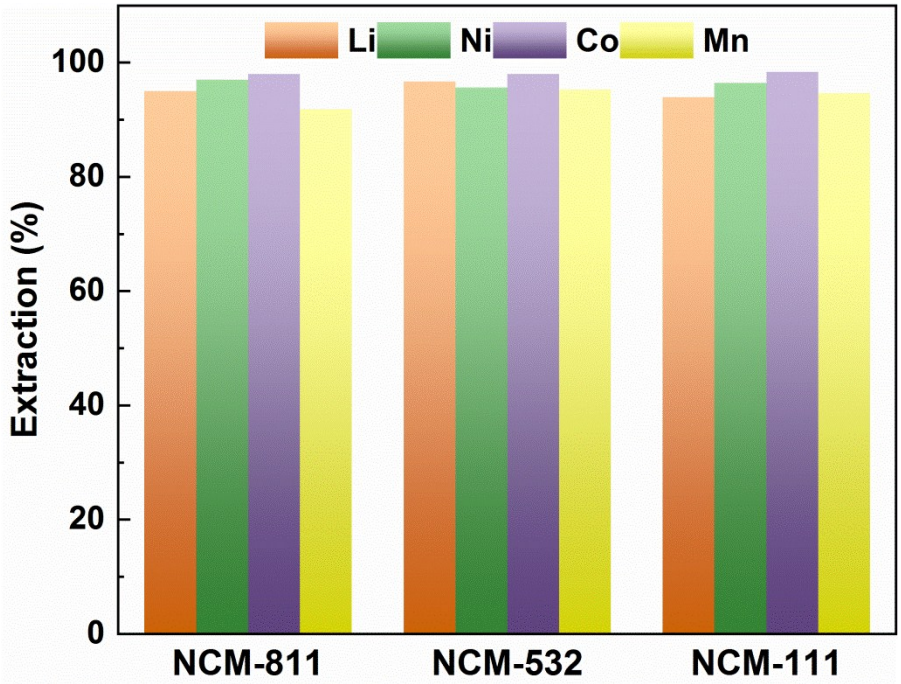

Fig. S10 Extraction of Li, Co, Ni, and Mn by using ChCl : urea DES at 180 °C and 24 h.

Table S1 FT-IR spectra comparison of DES in the present and absent of Co(II) cations

Wavenumber (cm^{-1})	DES 1	DES 2	Intensity
3347	ν_{as} (amide N–H)	ν_{as} (amide N–H)	Broad, strong
3205	ν_{s} (amide N–H)	ν_{s} (amide N–H)	Broad, strong
2208	-		Sharp, weak
2164	-		Sharp, weak
1668	ν (amide C＝O)	ν (amide C＝O)	Sharp, strong
1622	δ (amide N–H)	δ (amide N–H)	Sharp, strong
1450	ν (amide C–N)	ν (amide C–N)	Sharp, medium
1161	ν_{as} (CN)	ν_{as} (CN)	Weak
1085	ρ (CH2)	ρ (CH2)	Weak
956	ν_{as} (CCO)	ν_{as} (CCO)	Sharp, strong

1, 2 denote DES in the absent and present of Co^{2+}, respectively;

ν = stretching, δ = bending, ρ = rocking, as = asymmetric, s = symmetric

References

1. D. Y. Yue, Y. Z. Jia, Y. Yao, J. H. Sun and Y. Jing, *Electrochimica Acta*, 2012, **65**, 30-36.
2. M. K. Tran, M. T. F. Rodrigues, K. Kato, G. Babu and P. M. Ajayan, *Nature Energy*, 2019, **4**, 339-345.