

Supplemental Information

Engineering a Tandem Leaching System for Highly-Selective Recycling of Valuable Metals from Spent Li-ion Batteries

Linlin Chen,^a Yanhong Chao,^{*b} Xiaowei Li,^a Guolang Zhou,^a Qingqiang Lu,^a Mingqing Hua,^a Hongping Li,^a Xiaoguang Ni,^c Peiwen Wu,^{*a} Wenshuai Zhu^{*a}

*Correspondence: chaoyh@ujs.edu.cn

**Correspondence: wupeiw@ujs.edu.cn

***Correspondence: zhuws@ujs.edu.cn

Contents:

1. Supplementary Figures

Fig. S1 FT-IR spectra of acetic acid, ChCl, and DES-a.

Fig. S2 FT-IR spectra of propionic acid, ChCl, and DES-p.

Fig. S3 FT-IR spectra of n-butyric acid, ChCl, and DES-b.

Fig. S4 ¹H-NMR of DES-f.

Fig. S5 ¹H-NMR of formic acid.

Fig. S6 ¹H-NMR of ChCl.

Fig. S7 ¹H-NMR of obtained at 25 °C and maintained for 90 min.

Fig. S8 ¹H-NMR of acetic acid.

Fig. S9 ¹H-NMR of DES-a.

Fig. S10 ¹H-NMR of propionic acid.

Fig. S11 ¹H-NMR of DES-p.

Fig. S12 ¹H-NMR of n-butyric acid.

Fig. S13 ¹H-NMR of DES-b.

Fig. S14 XRD patterns of pristine LCO.

Fig. S15 XRD patterns of LCO residue being leached by formic acid at 90°C for 4 and 12 h.

Fig. S16 Co leaching efficiencies over LCO and the residue by DES-f at different temperatures.

Fig. S17 SEM of (a) original LCO; (b) residues obtained after formic acid leaching.

Fig. S18 XRD pattern of recycled Li₂CO₃ from formic acid extractant. Inset is the optical photograph of recycled Li₂CO₃.

Fig. S19 XRD pattern of recycled CoCO₃ from DES-f extractant. Inset is the optical photograph of recycled CoCO₃.

Fig. S20 FT-IR spectra of DES-f and the recycled DES-f after leaching process at 70 °C.

Fig. S21 FT-IR spectra of DES-a and the recycled DES-a after leaching process at 70 °C.

Fig. S22 FT-IR spectra of DES-p and the recycled DES-p after leaching process at 70 °C.

Fig. S23 FT-IR spectra of DES-b and the recycled DES-b after leaching process at 70 °C.

Fig. S24 ¹H-NMR spectra of DES-f that being treated at 70 °C for 2, 4, 8, and 12 h.

Fig. S25 Recyclability of the DES-f.

1. Supplementary Figures

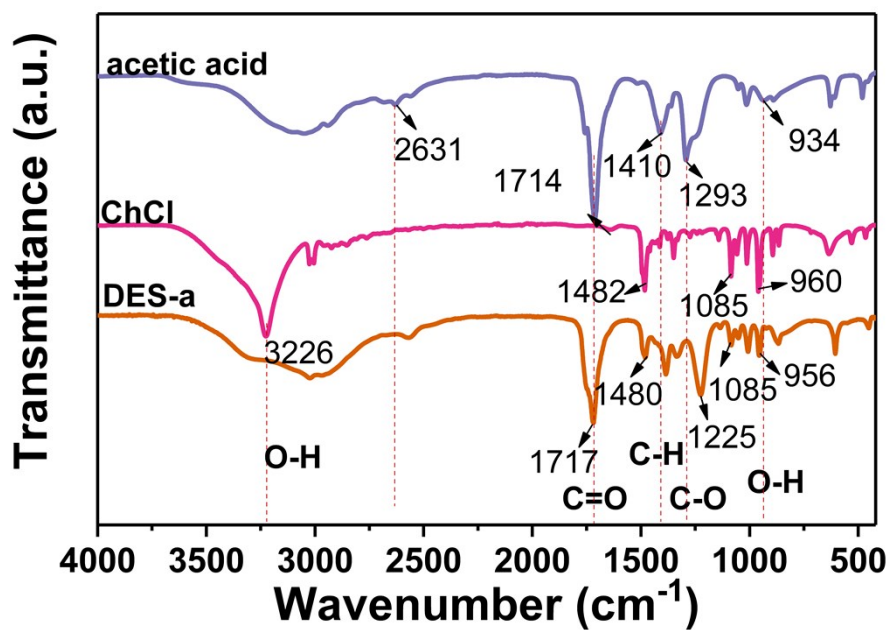


Fig. S1 FT-IR spectra of acetic acid, ChCl, and DES-a.

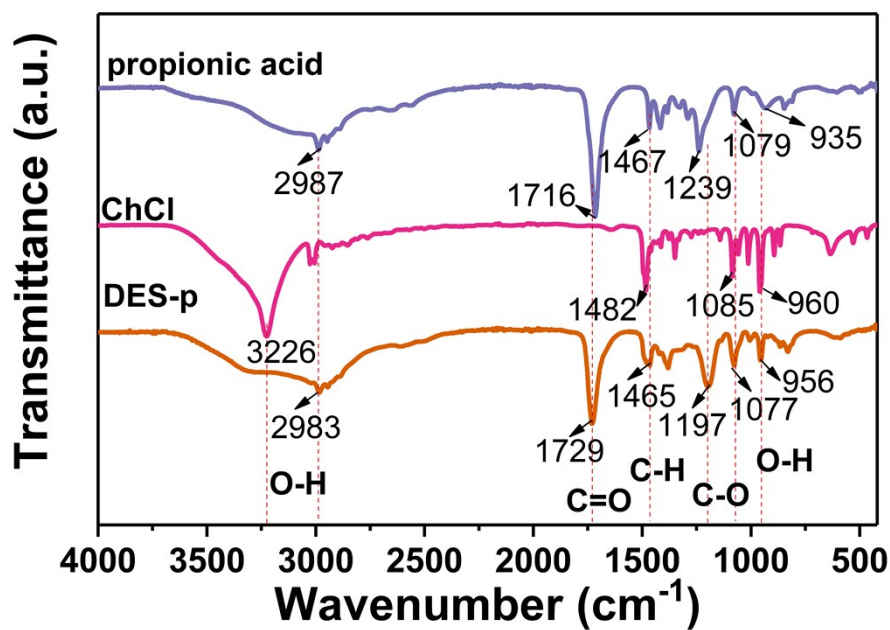


Fig. S2 FT-IR spectra of propionic acid, ChCl, and DES-p.

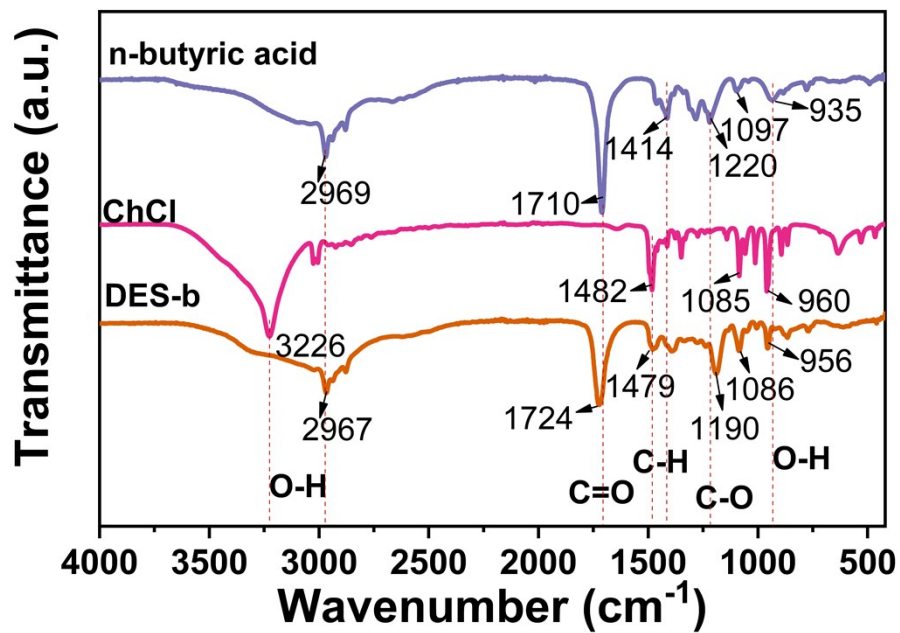


Fig. S3 FT-IR spectra of n-butyric acid, ChCl, and DES-b.

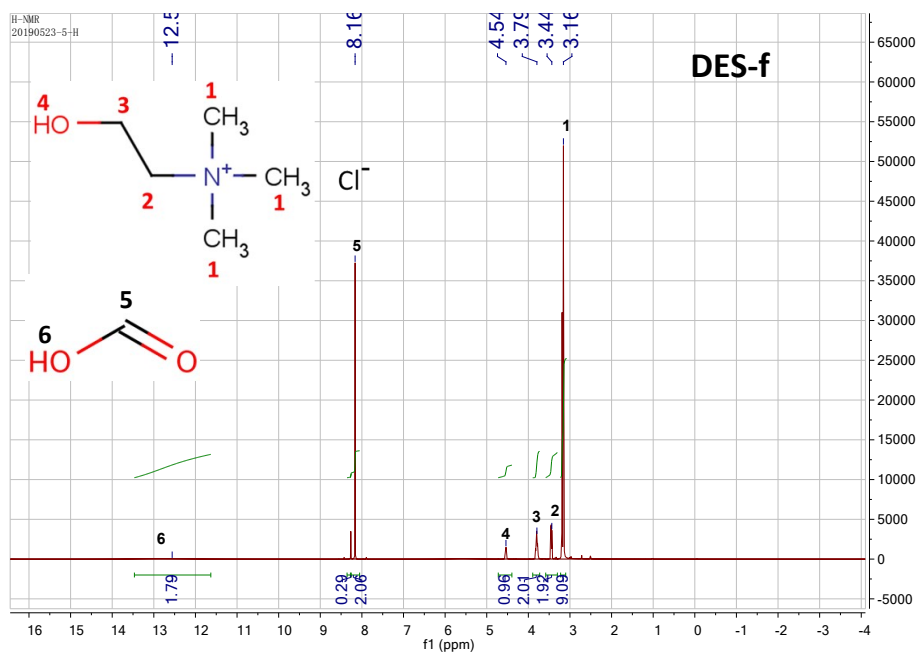


Fig. S4 $^1\text{H-NMR}$ of DES-f

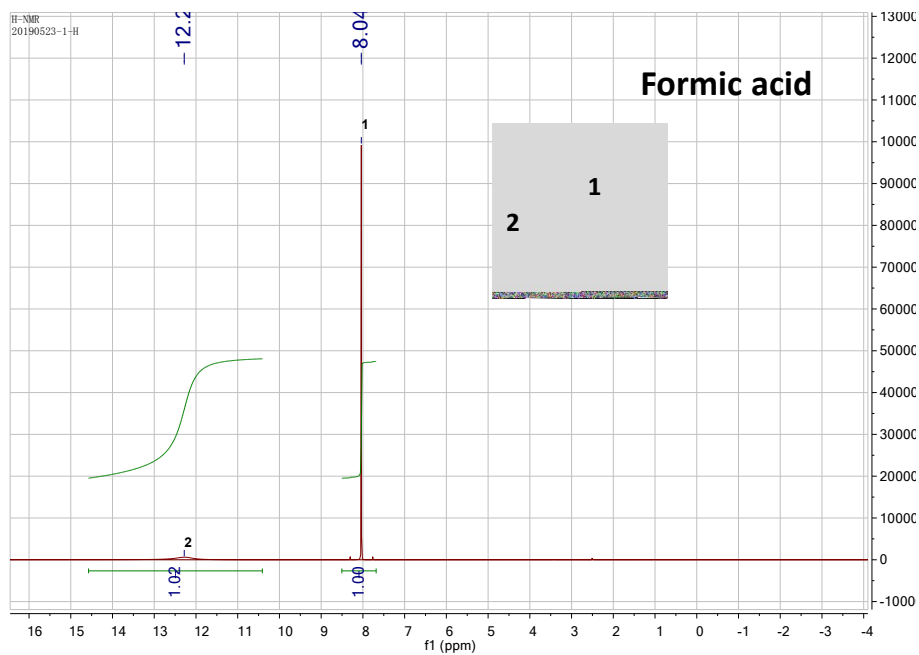


Fig. S5 ¹H-NMR of formic acid

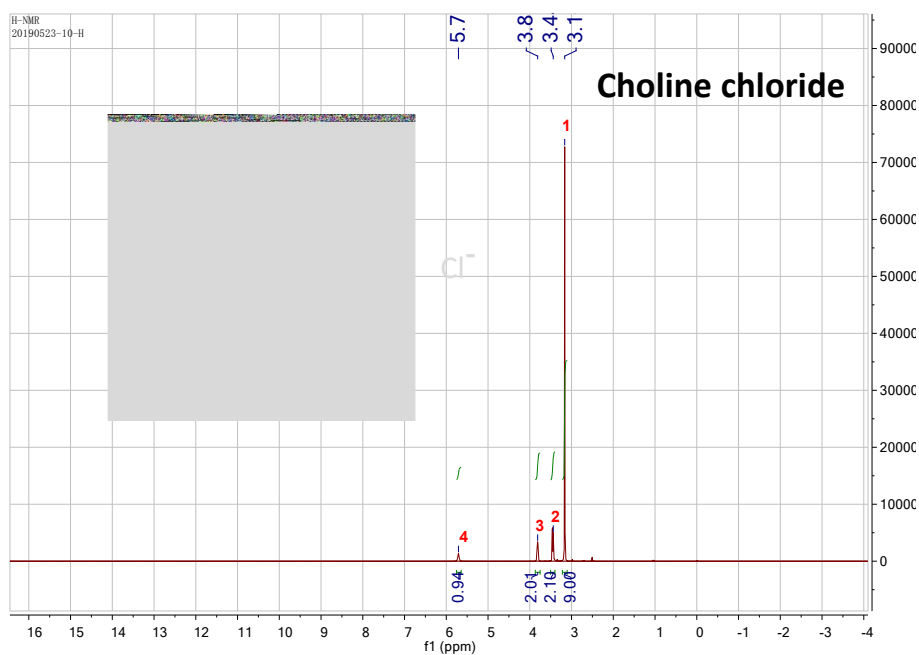


Fig. S6 ¹H-NMR of ChCl

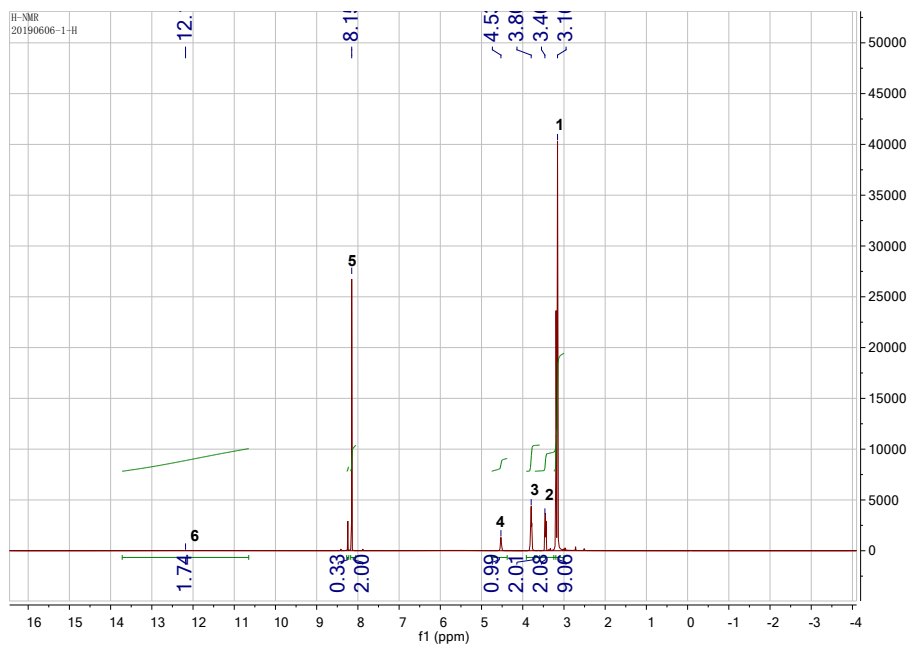


Fig. S7 $^1\text{H-NMR}$ of DES-f obtained at 25°C and maintained for 90 min

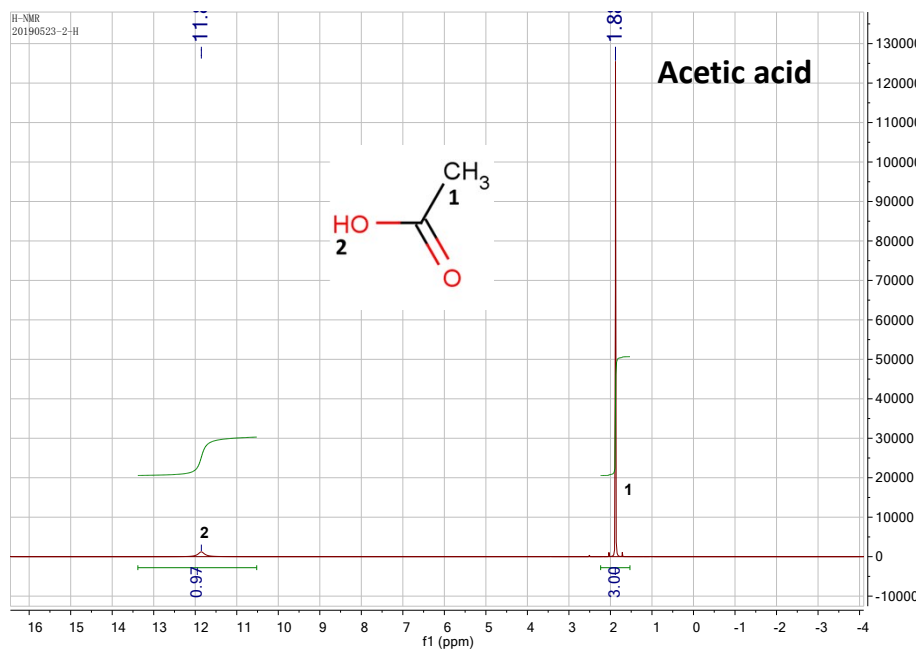


Fig. S8 $^1\text{H-NMR}$ of acetic acid

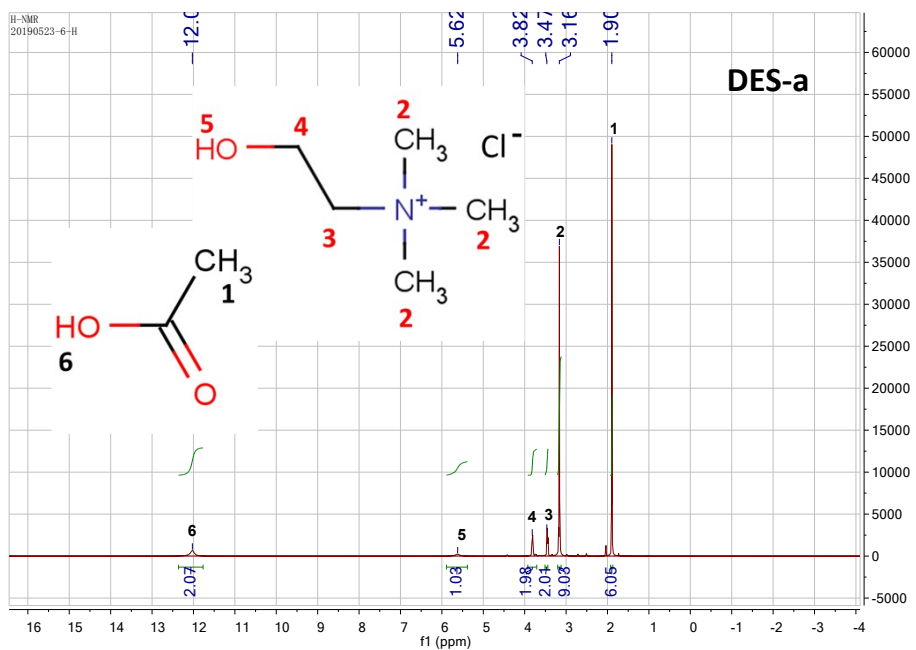


Fig. S9 ¹H-NMR of DES-a

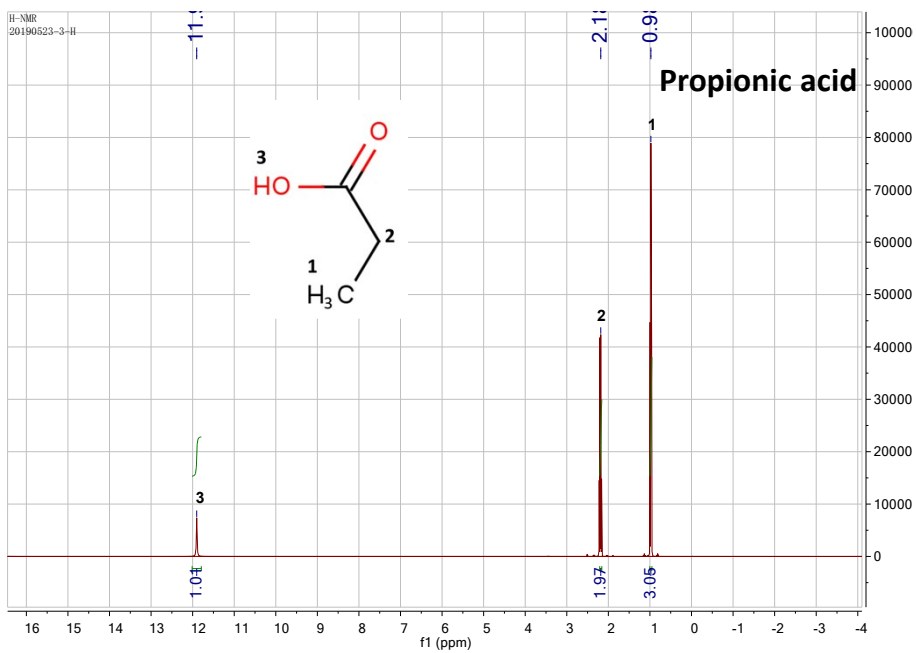


Fig. S10 ¹H-NMR of propionic acid

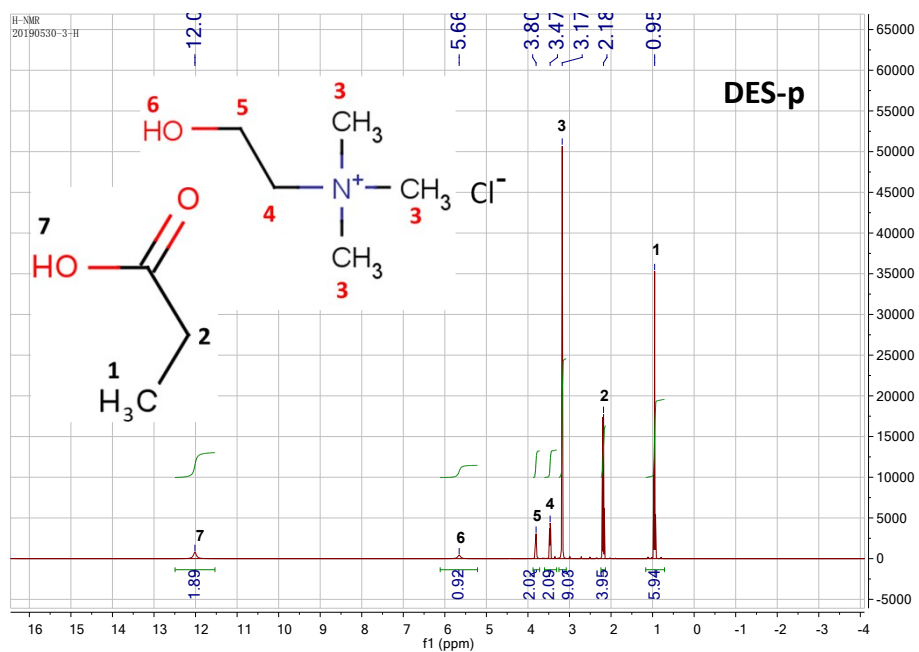


Fig. S11 ¹H-NMR of DES-p

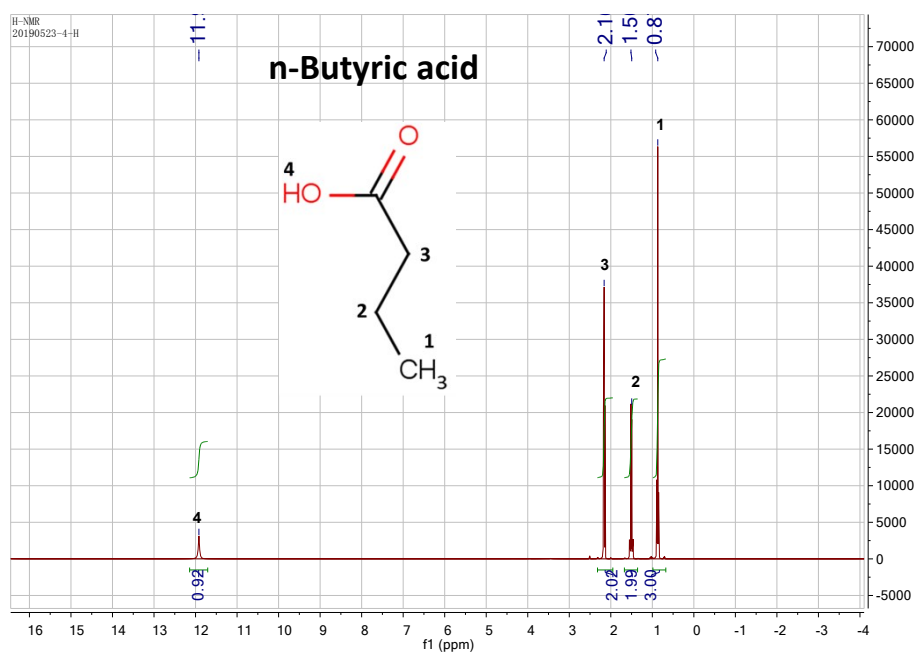


Fig. S12 ¹H-NMR of n-butylric acid

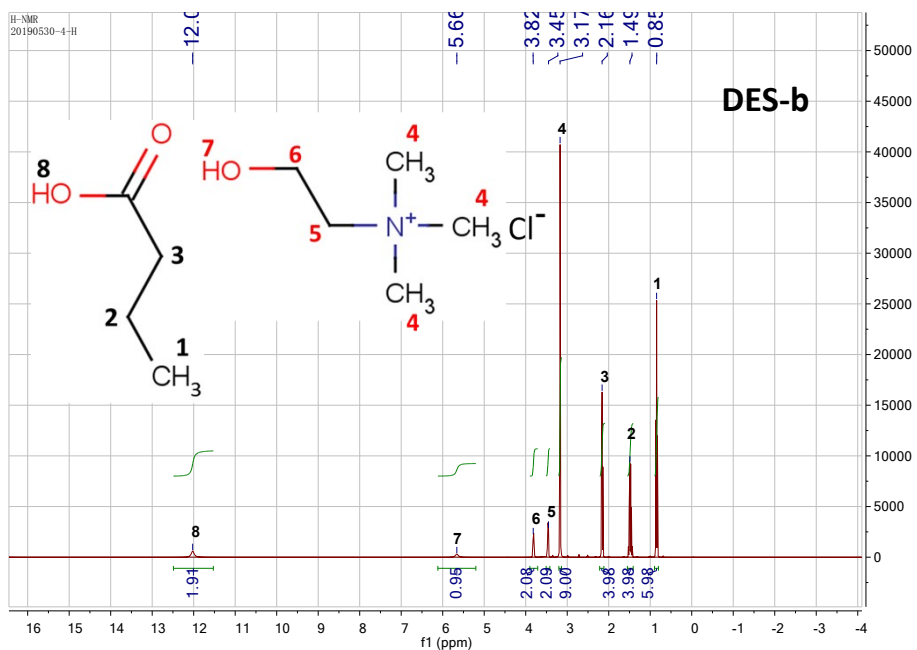


Fig. S13 ¹H-NMR of DES-b

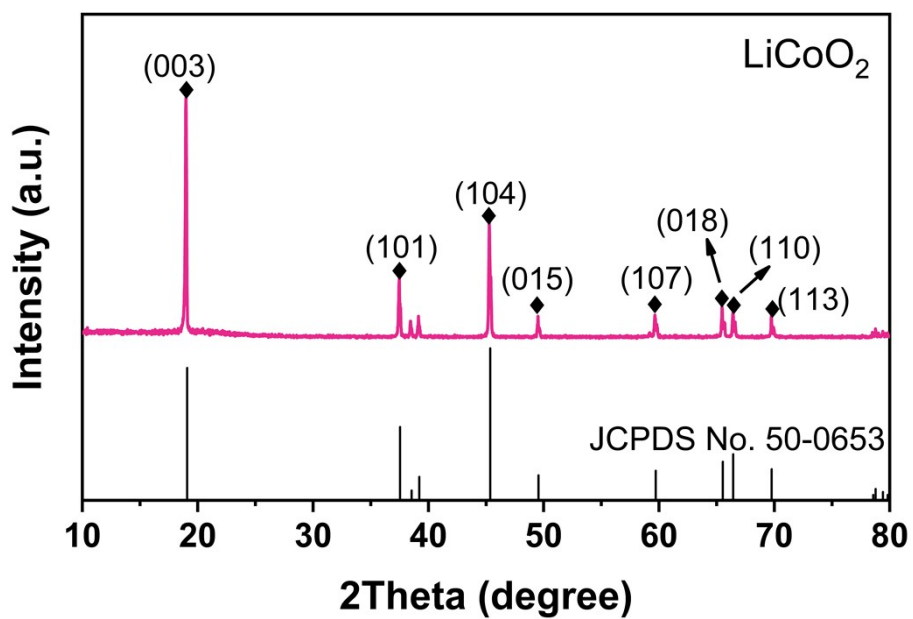


Fig. S14 XRD patterns of pristine LCO.

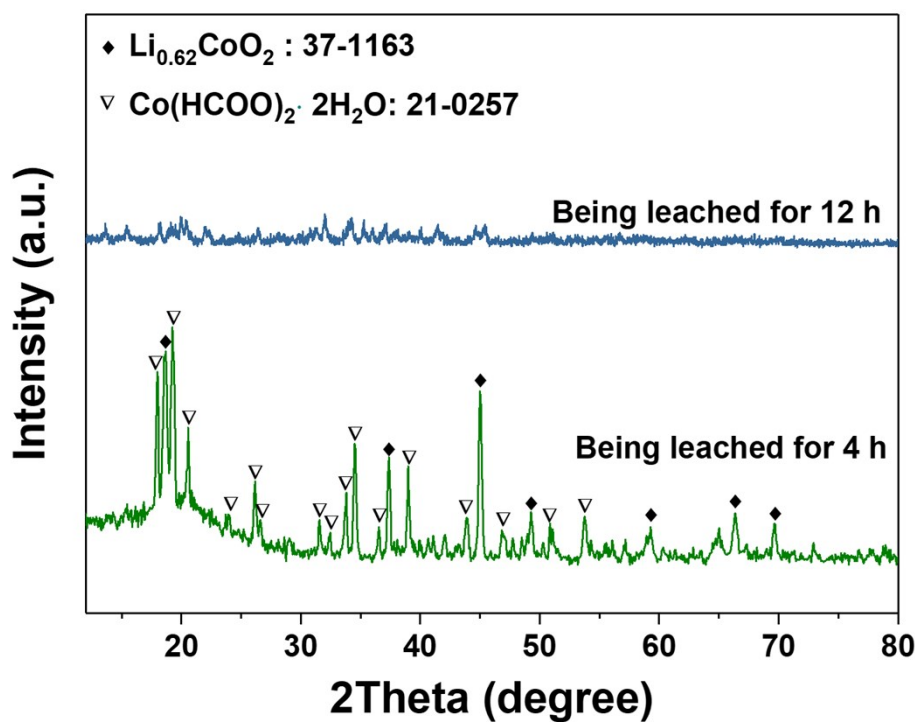


Fig. S15 XRD patterns of LCO residue being leached by formic acid at 90°C for 4 and 12 h.

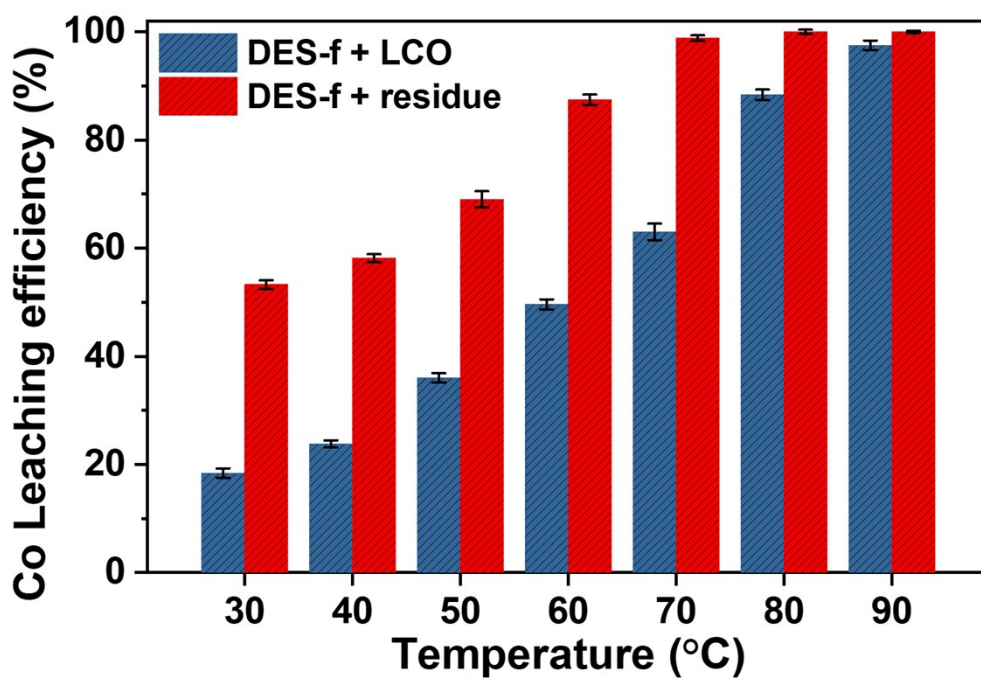


Fig. S16 Co leaching efficiencies over LCO and the residue by DES-f at different temperatures.

Leaching conditions: $m(\text{cathode}) = 0.1 \text{ g}$; $m(\text{DES}) = 5 \text{ g}$; $t = 12 \text{ h}$.

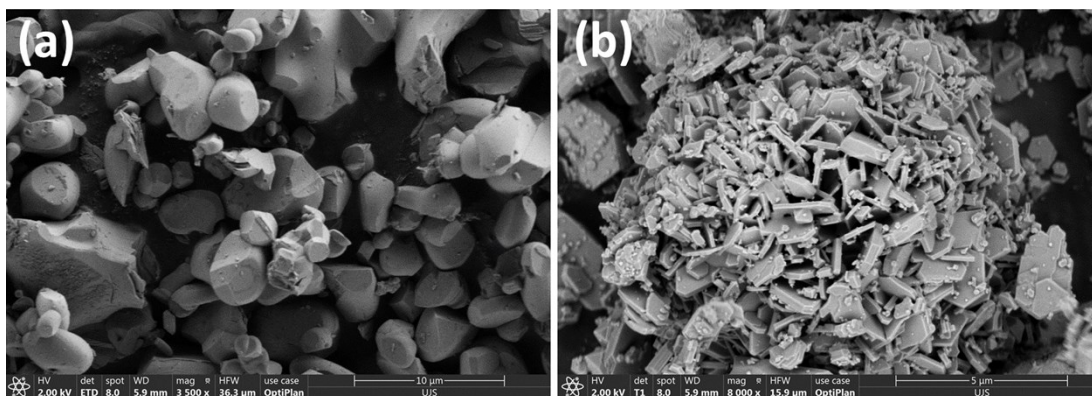


Fig. S17 SEM of (a) original LCO; (b) residues obtained after formic acid leaching.

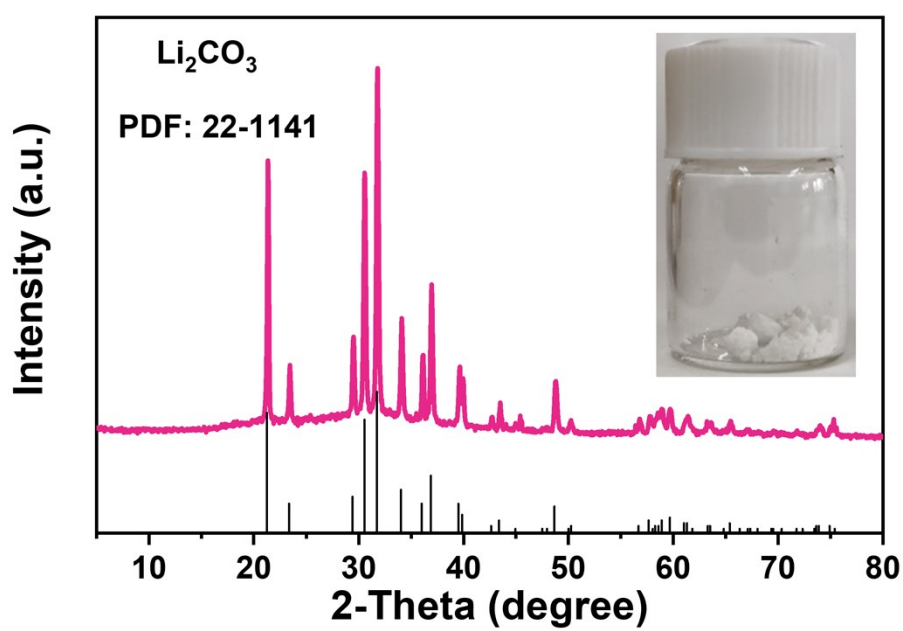


Figure S18 XRD pattern of recycled Li_2CO_3 from formic acid extractant. Inset is the optical photograph of recycled Li_2CO_3 .

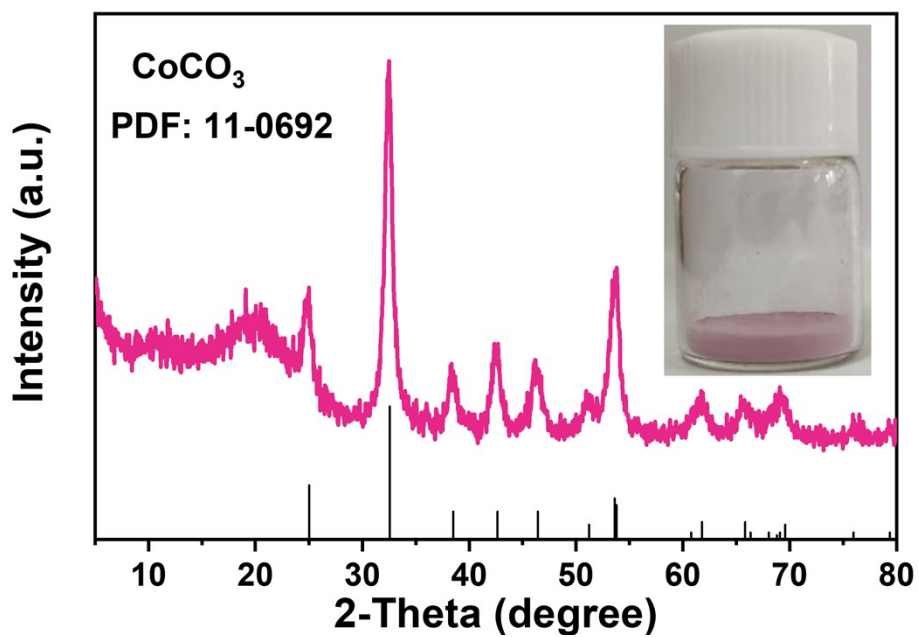


Fig. S19 XRD pattern of recycled CoCO_3 from DES-f extractant. Inset is the optical photograph of recycled CoCO_3 .

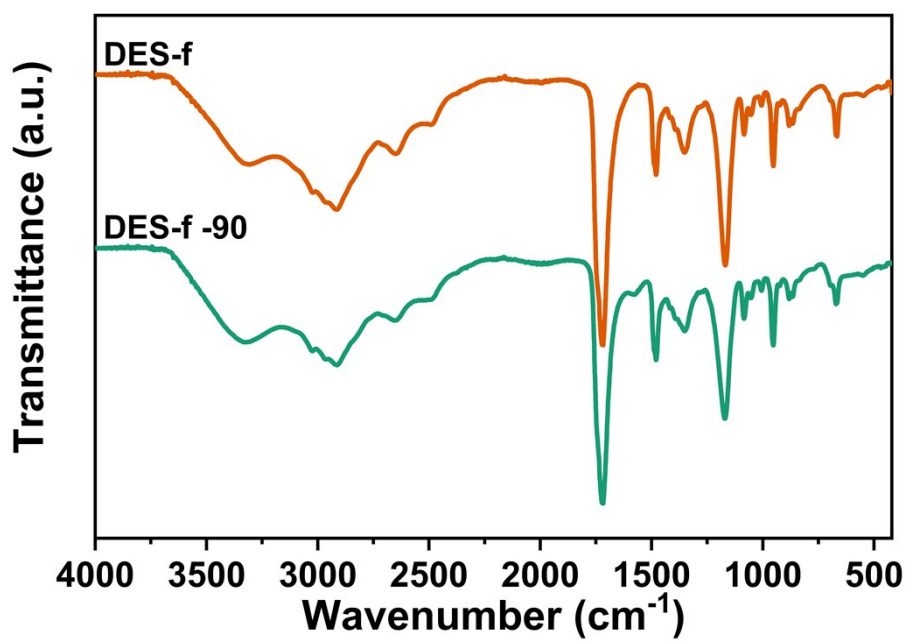


Fig. S20 FT-IR spectra of DES-f and the recycled DES-f after leaching process at 70°C .

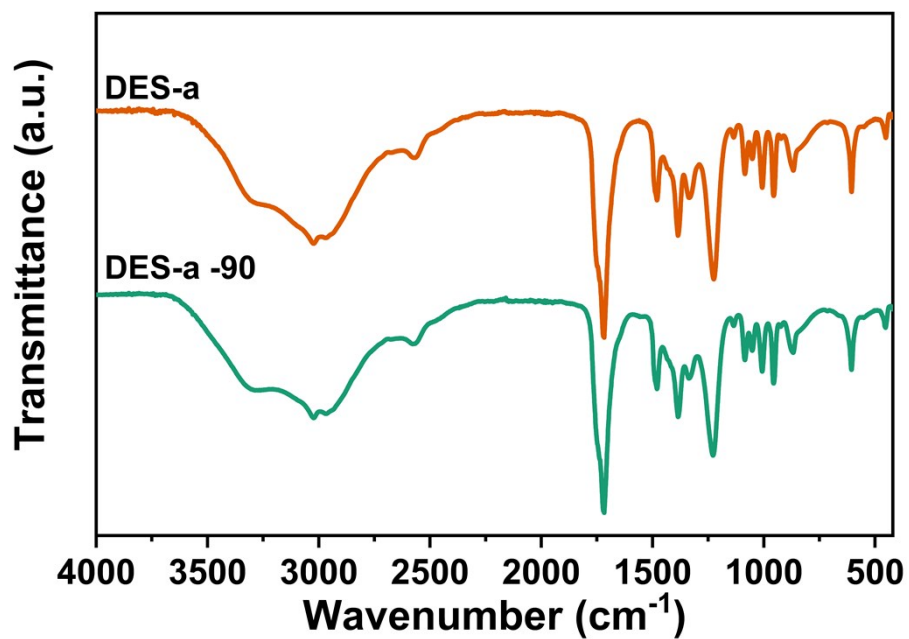


Fig. S21 FT-IR spectra of DES-a and the recycled DES-a after leaching process at 70 °C.

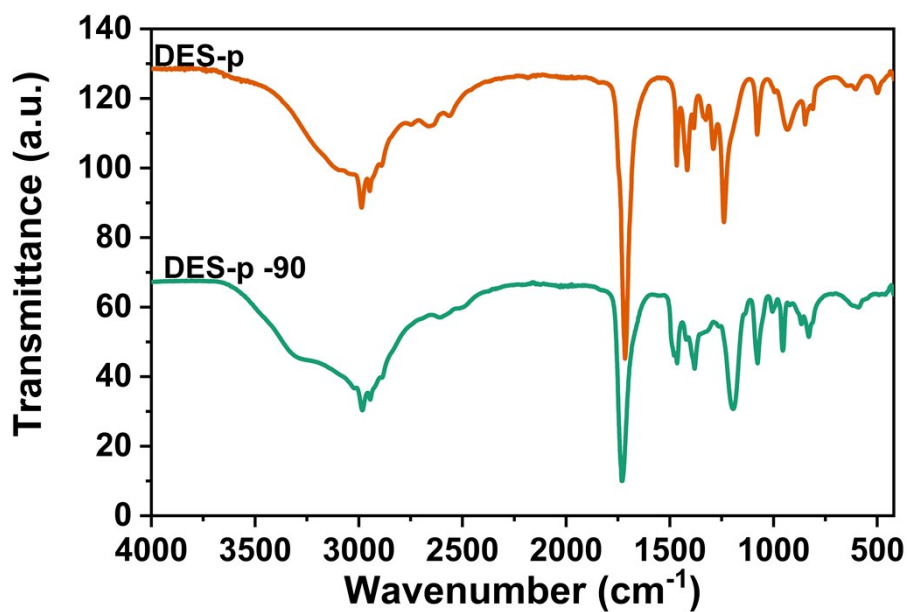


Fig. S22 FT-IR spectra of DES-p and the recycled DES-p after leaching process at 70 °C.

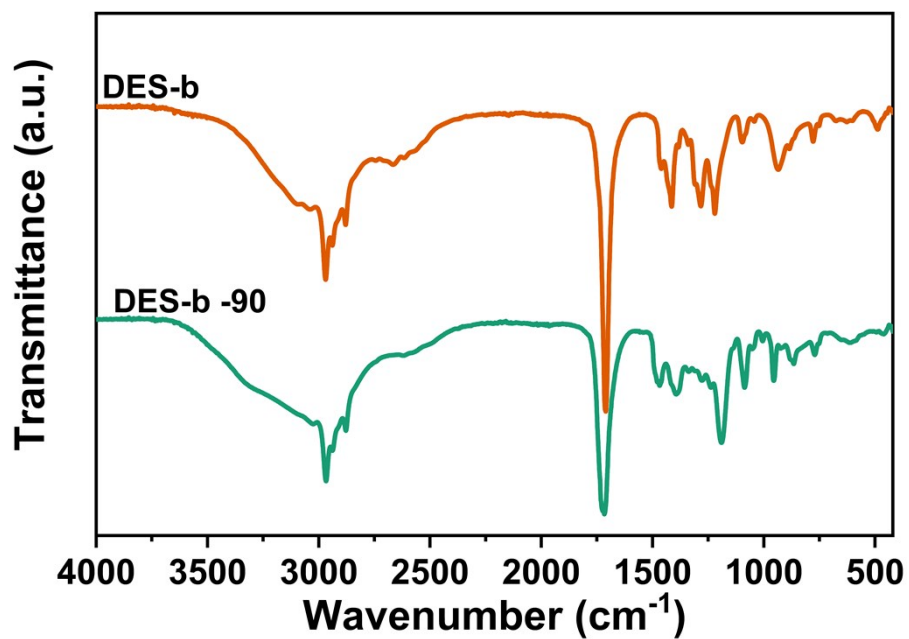


Fig. S23 FT-IR spectra of DES-b and the recycled DES-b after leaching process at 70 °C.

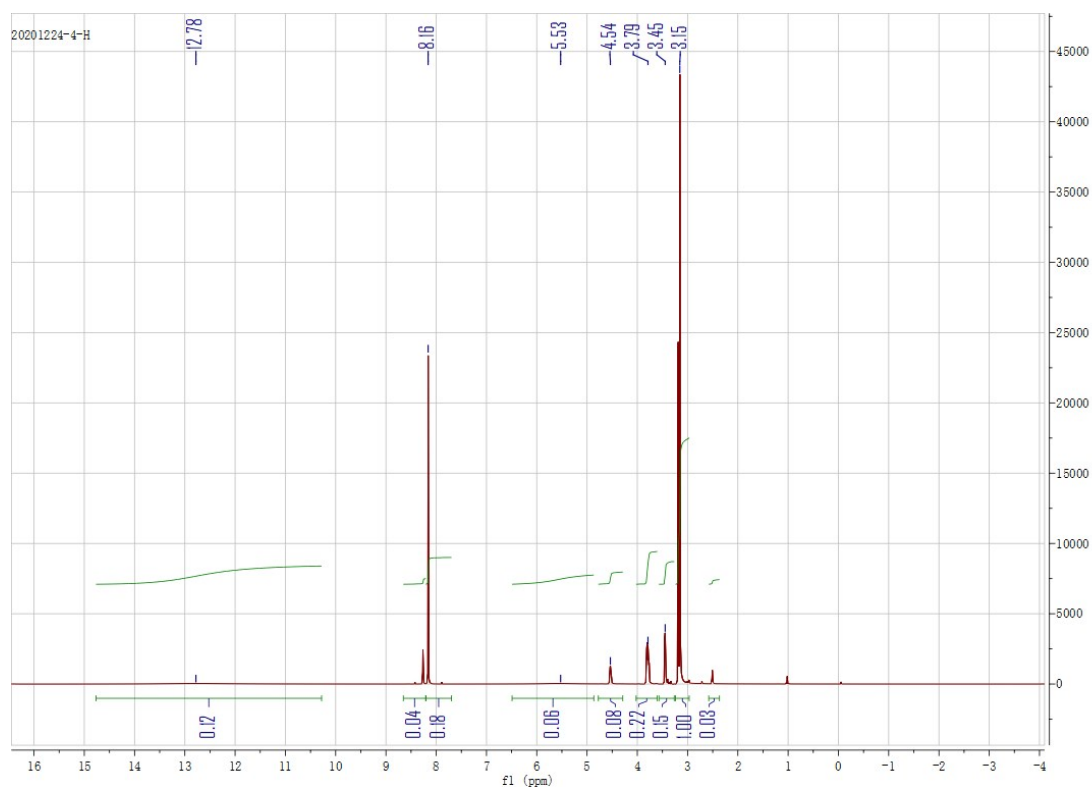


Fig. S24 $^1\text{H-NMR}$ spectra of DES-f that being treated at 70 °C for 2 h.

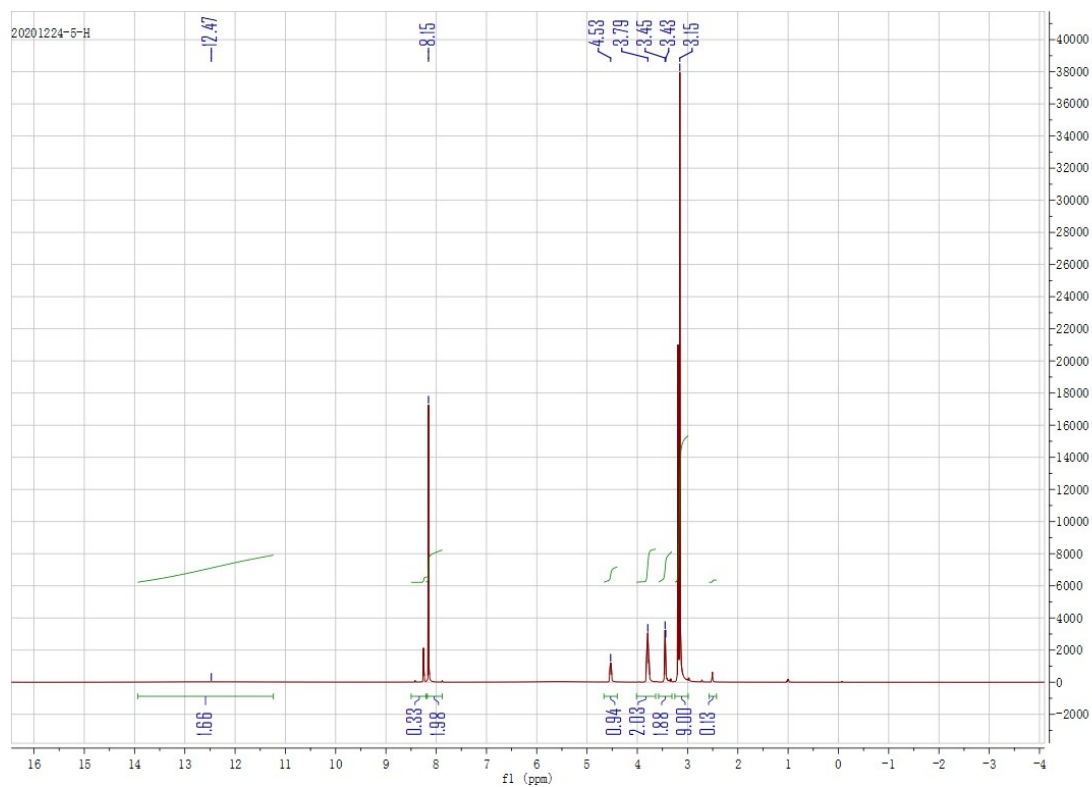


Fig. S25 $^1\text{H-NMR}$ spectra of DES-f that being treated at 70 °C for 4 h.

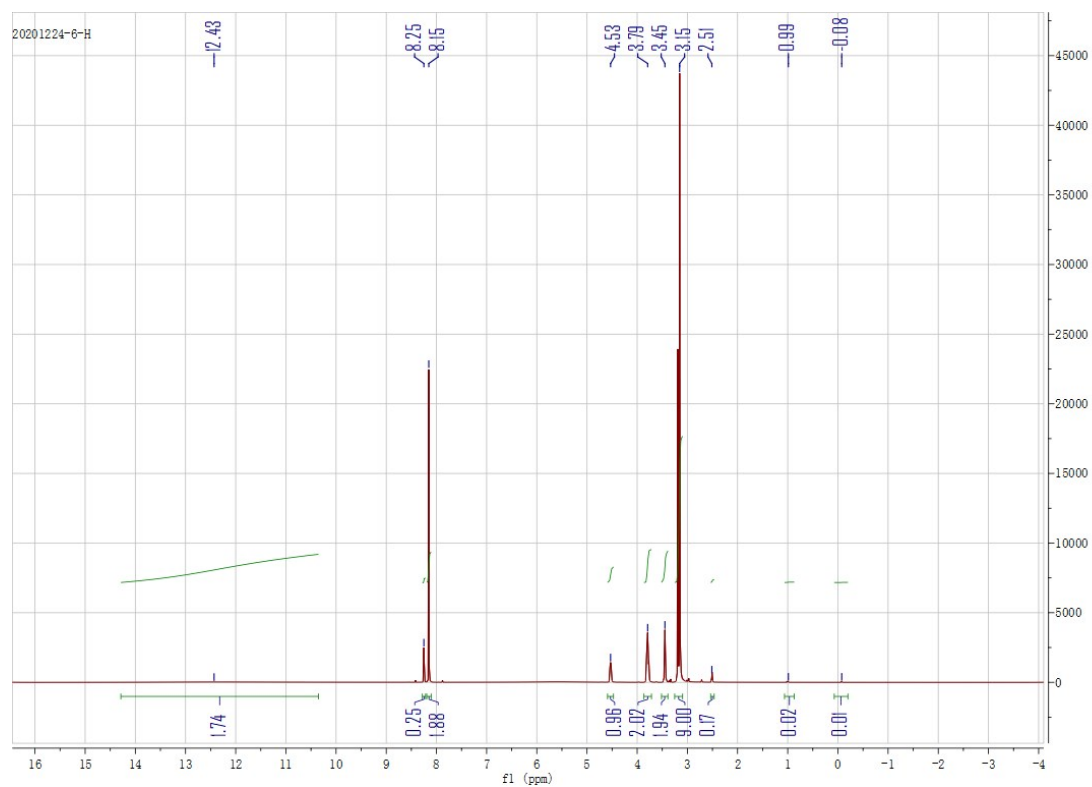


Fig. 26 $^1\text{H-NMR}$ spectra of DES-f that being treated at 70 °C for 8 h.

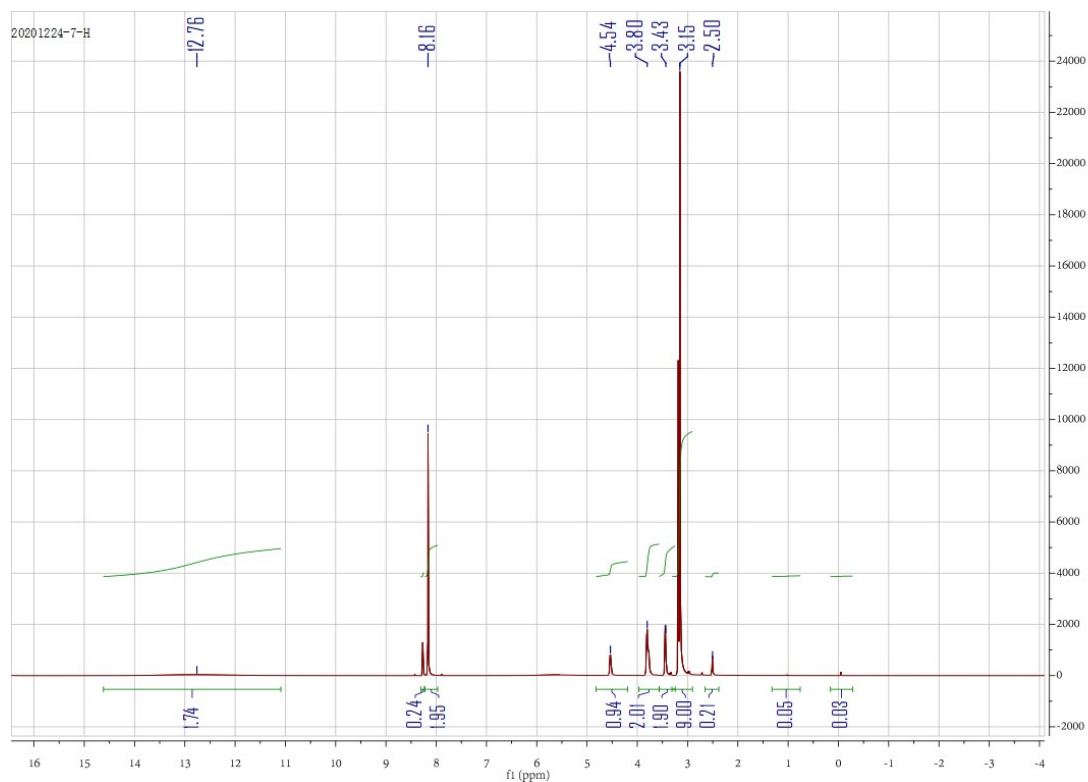


Fig. 27 $^1\text{H-NMR}$ spectra of DES-f that being treated at 70 °C for 12 h.

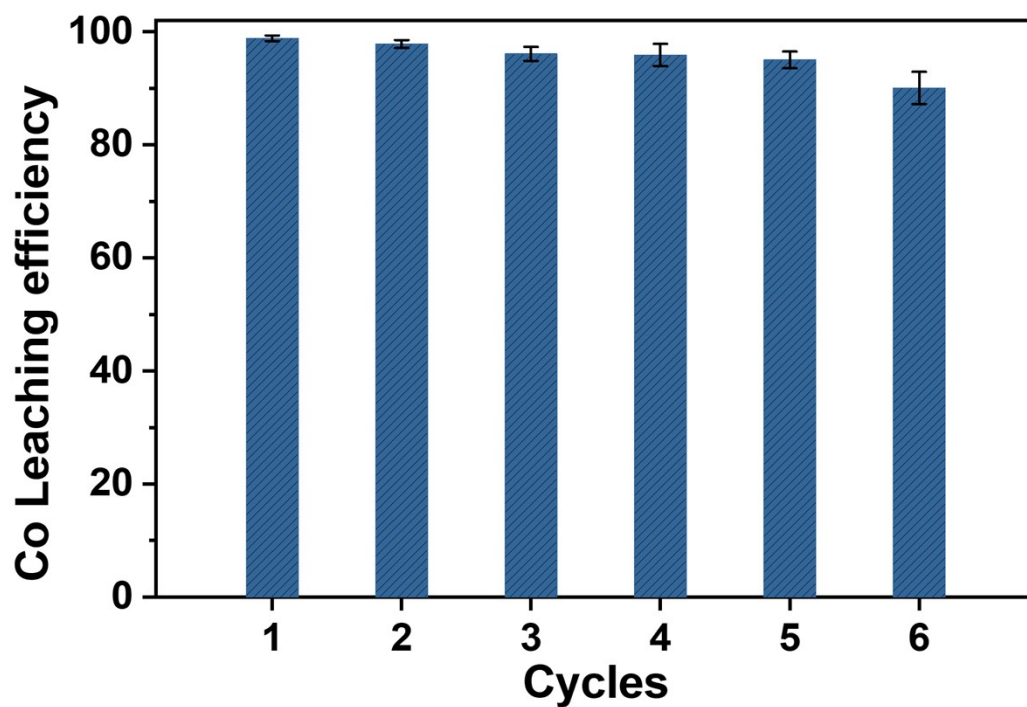


Fig. S28 Recyclability of the DES-f.

Leaching conditions: $m(\text{cathode}) = 0.1 \text{ g}$; $m(\text{DES}) = 5 \text{ g}$; $t = 12 \text{ h}$.