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Achieving reusability of leachate for multi-element recovery of the discarded $LiNi_xCo_yMn_{1-x-y}O_2$ cathode by regulating the co-precipitation coefficient

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ABSTRACT

Conventional hydrometallurgy recycling process for treating wasted lithium-ion batteries (LIBs) typically results in the consumption of large amounts of corrosive leachates. Recent research on reusable leachate is expected to significantly improve the economic and environmental benefits, but is usually limited to specific and unique chemical reactions which could only apply to one type of metal elements. Herein, we report the co-extraction of multiple metal elements can be extracted without adding precipitates by mixed crystal co-precipitation, which enables the reusability of the leachate. We show that an oxalic acid (OA): choline chloride (ChCl): ethylene glycol (EG) type DES leachate system can leach transition metals from wasted $LiNi_xCo_yMn_{1-x-y}O_2$ (NCM) cathode materials with satisfactory efficiency (The time required for complete leaching at 120 °C is 1.5 h). The transition metals were then efficiently extracted (with a recovery efficiency of over 96% for all elements) by directly adding water without precipitants. Noteworthy, the leachate can be efficiently recovered by directly evaporating the added water. The successful realization of reusability of leachate for the synergistic extraction of multiple elements relies on the regulation of the mixed crystal co-precipitation coefficient, which is realized by rationally design the reaction condition (composition of leachate, temperature and time) and induces the extraction of originally soluble manganese element. Our strategy is expected to be generally applicable and highly competent for industrial applications.

With the widespread and large-scale application of LIBs, the quantity of End-of-Life (EoL) LIBs has been increasing rapidly [1–9]. Efficient recovery of valuable elements (particularly transition metals within the cathode materials) holds significant importance in terms of environmental protection, economic benefits, and resource utilization [10–15]. Hydrometallurgical methods for EoL-LIBs recycling have garnered increasing attention compared to pyrometallurgical recycling due to their lower energy consumption and less gas emissions [16–20]. The extraction process for transition metals in EoL-LIBs primarily consists of two steps: the leaching and the precipitation of specific elements using leachate [21]. Conventional hydrometallurgical strategy usually consumes large amounts of highly corrosive leachate, which leads to safety and environmental concerns [22]. It is highly desirable to develop reusable and mild leachate to improve the overall cost and prepare for the massive amounts of EoL-LIBs in the coming years [23–25]. However, realizing reusability is challenging because the fundamental mechanism for extracting specific elements in hydrometallurgical strategy involves the dissolution and the precipitation processes, during which the required elements can be separated from the wasted residues [26–30]. This necessitates the property of the leachate to be changed between two states: the elements can dissolve or precipitate in the leachate. Typical hydrometallurgical strategy relies on strong inorganic acid to dissolve the metal elements, as well as the subsequent addition of precipitants (usually alkaline or carbonate) to neutralize the leachate for precipitation [21,31-34]. As a result, the leachate would irreversibly lose its capability for next-round leaching.

Recently, reusable leachate for recycling lithium cobalt oxide (LCO) has been reported, where the key is to reversibly control the solubility product (K_{sp}) of the cobalt element based on discovering specific chemical reaction pathways [35]. Fundamentally, the controllable switching for the metal elements to change between the soluble and precipitated state can be realized without adding pre-

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cipitants, thus maintaining the composition and property for the leachate and enabling reusability. Despite the successful demonstration of the above strategy for recycling LCO, this method relies on the specific property (controllable formation of specific metal complex species with opposite solubility product) of the cobalt element [30,36,37]. Considering that many LIBs suppliers have been pursuing to decrease the usage of cobalt due to economic consideration, the extension to the simultaneous recycling of multiple elements using reusable leachate is calling for urgent development. $LiNi_xCo_yMn_{1-x-y}O_2$ (NCM) family materials represent the mainstream solution for a variety of high energy density LIBs [38–40]. However, unlike cobalt element, the extraction of other valuable element including nickel and manganese is challenging considering the requirement for achieving reusability of leachate (no precipitants allowed) [41,42]. Co-precipitation, which refers to the phenomenon that a normally soluble substance can be carried down upon the formation of a precipitate. This is expected to provide the possibility for the synergistic extraction of the originally soluble elements. The mechanism for co-precipitation includes mixed crystal co-precipitation, occlusion co-precipitation and adsorption co-precipitation [43]. We propose that, if the extraction of originally soluble elements is proceeded without adding precipitants, but by the formation of the precipitated metal compound, precipitant-free extraction might be achieved.

Herein, we report the synergistic and complete recovery of Mn, Ni, Co elements from spent NCM materials where the reusability of leachate has been successfully achieved by regulating the mixed crystal co-precipitation coefficient of the leachate. An OA:ChCl:EG type deep eutectic solvent (DES) was designed as the leachate. DESs are a diverse range of solvents composed of hydrogen bond donors (HBD) and hydrogen bond acceptors (HBA) with cost-effectiveness, low toxicity, and biodegradability [44–46]. A variety of new chemical reaction pathway has been found in DESs compared with aqueous or organic solvents, which provides potential for developing effective hydrometallurgical recycling strategies [18,27,47-50]. The leachate can effectively dissolve the NCM materials at 90 °C for 4 h with a leaching efficiency of nearly 100%. Noteworthy, in this specific OA:ChCl:EG type DES, the subsequent addition of deionized water (DI-water) could lead to the co-extraction of the Ni, Co and Mn elements with extremely high efficiency (98.6%, 99.5%, and 96.9%). For comparison, without the existence of Co and Ni, pure Mn element could not be precipitated under similar condition. Meanwhile, compared to widely studied ChCl:OA type DES, the EG composition in our designed DES significantly improve the co-precipitation coefficient (0.746 in OA:ChCl:EG *vs.* 0.582 in OA:ChCl).This indicates that regulating the mixed crystal co-precipitation coefficient provides a viable approach for the extraction of multiple transition metal elements including those originally soluble elements without precipitants. The added DI-water can be evaporated to restore the reusability of the leachate. We show that identical leaching processes were observed over five repeated leaching/extraction cycles, which demonstrated the significant potential for repeated utilization of DES leachate. Furthermore, the precipitate obtained through mixed crystal co-precipitation was utilized as a precursor for the synthesis of active materials, demonstrating the establishment of closed-loop the recycling. The successful realization of reusable leachate for multi-elements recycling is expected to provide significant advancement for large-scale hydrometallurgical recycling of EoL-LIBs with cost and environmental effectiveness.

The characterization of the OA:ChCl:EG type DES are shown in Fig. S1 (Supporting information). In the NMR spectra, the hydroxyl group of ChCl shifts from 5.7 ppm to 6.5 ppm (Fig. S1a), and the carboxyl carbon atom of OA (161.35 ppm) undergoes splitting (Fig. S1b). These indicate the formation of hydrogen bonds which produce the shielding effect [22]. Such superstructure of the DESs could produce unique reaction pathways for interacting with various kinds of metal compounds [51]. The leaching process can be described using the shrinking-core model (Formulas S1-S4 in Supporting information). As evidenced with SEM images in Fig. 1a (pristine materials) and Fig. 1b (half-leached materials), the leaching process is accompanied with the collapse of the secondary particles and followed by shrinking-core reactions [52]. Assuming that the elemental ratio of the ionic nickel, cobalt, and manganese dissolved in the solution corresponds to the elemental ratio in the pristine raw materials, and the leaching rate can be calculated by using UV–vis spectra to quantify the intensity of adsorption peak (without separating the adsorption signal from whether cobalt or nickel, and the divalent manganese ions do not show observable UV–vis signals in the detectable range of wavelength). By collecting the concentration data at various reaction conditions (different temperatures and times, Figs. 1d-g and Fig. S2 in Supporting information), the activation energy (E_a) of the reaction can be calculated to be 124.71 kJ/mol, indicating that the leaching reaction can be accelerated by simply increasing the temperature (Fig. 1c).

The recycling process involves the leaching and extraction of metal elements as well as the recovery of the leaching solution (Fig. S3 in Supporting information). The co-extraction of the dissolved metal elements without adding precipitants is the key for maintaining the reusability of the leaching. We show case the mixed crystal co-precipitation strategy can be used for the co-extraction of the nickel, cobalt and manganese ions from the solution. After the leaching process, an appropriate amount of DI-water (twice the mass of DES) is added. With the addition of DI-water, cobalt and nickel ions have been reported to form $MC_2O_4{\cdot}2H_2O$ (M = Ni, Co) and would precipitate out from the solution at a precipitation rate ranging from 79% to 99% [22]. However, pure manganese ions would stay as the soluble state in the OA:ChCl:EG system (Fig. S4 in Supporting information). Herein, we demonstrate that for the leachate containing the mixed metal ions, the formation of the mixed crystal co-precipitation would guide the co-extraction of the metal elements (even for the originally soluble manganese ions). We propose a mixed crystal co-precipitation formation equation as follows (the derivation process is detailed in Supporting information) [43]:

$$\ln(T(\mathrm{Mn})/L(\mathrm{Mn})) = \lambda \ln\left(T\left(\mathrm{Ni\&Co}\right)/L\left(\mathrm{Ni\&Co}\right)\right) \quad (1)$$

where $L(xx)$ is the content of xx in the solution, $T(xx)$ is the total amount of xx, and λ represents mixed crystal co-precipitation coefficient.

Fig. 2a illustrates that the mixed crystal co-precipitation coefficient (λ) determines the precipitation efficiency of manganese. The mixed crystal co-precipitation coefficient is a function of various parameters such as temperature, time, concentration of precipitant, solvent system, etc. Indicates that λ is difficult to be theoretically predicted [53,54]. For the OA: ChCl: EG leachate system, the SEM-EDS (Fig. 2c and Fig. S5 in Supporting information) of the precipitated particle indicates that manganese could precipitate with the existence of nickel and cobalt elements. The precipitation efficiencies of nickel, cobalt, and manganese are 98.6%, 99.5%, and 96.9%, respectively, at 30 °C for 12 h (Fig. 2b), which indicates that the λ can be regulated to close to 1 under the optimized condition. Noteworthy, without EG, the precipitation efficiency of manganese in OA:ChCl leachate system is only 75.8% at 30 °C for 12 h (Fig. 2d), indicating the elegant design of the composition for the leachate is critical for the highly efficient co-extraction. XRD and XPS have been used to characterize the structure of the mixed crystal co-precipitation. XRD pattern (Fig. S6 in Supporting information) of the mixed crystal co-precipitation shows the consistent feature peaks as the standard card with no impurities. XPS spectra (Fig. 2d) curve fitting of the Ni 2p, Co 2p and Mn 2p further

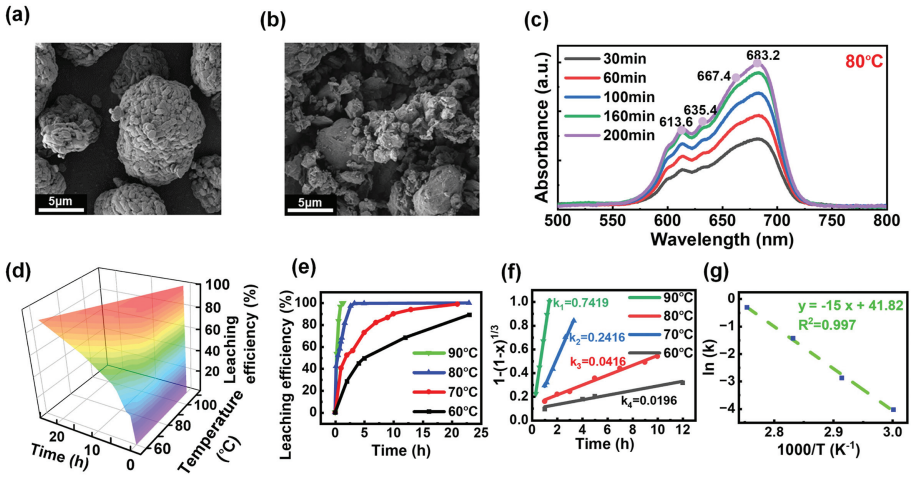

Fig. 1. The leaching kinetics of the NCM materials in the DES leachate: (a, b) The discard $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ (D-NCM111) before and after leaching for 30 min, respectively. (c) UV-visible spectra of the leachates obtained at various extraction times at 80 °C (using DMSO as a diluent for DES). (d) The time-dependent variation of leaching efficiency at different temperatures. (e) The leaching efficiency under different conditions (60–90 °C, 0–23 h). (f, g) The fitting of activation energy for the leaching reaction.

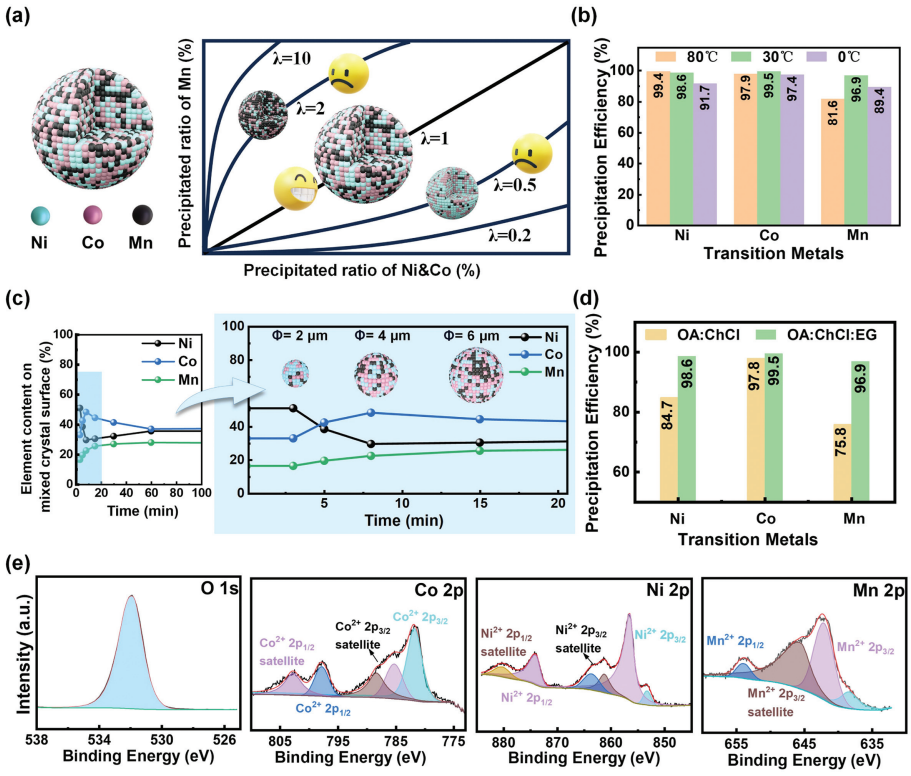

Fig. 2. Characterization of the mixed crystal co-precipitation. (a) The regulation of co-precipitation coefficient is the key for highly efficient precipitation of manganese based on mixed crystal theory. (b) The precipitation efficiency of nickel, cobalt, and manganese elements at different temperatures. (c) The ratio of elements on the surface of the mixed crystal co-precipitation forming process quantified by EDS. (d) Comparison of precipitation efficiency between OA:ChCl:EG and OA:ChCl types DES systems (30 °C). (e) XPS spectra of the precipitate.

confirm the composition of the mixed crystal co-precipitation is $MC_2O_4{\cdot}2H_2O$ (M = Ni, Co, Mn) [55].

Compared to conventional extraction strategy using precipitants, the extraction of metal ions realized by mixed crystal co-precipitation could avoid the precipitants, thus maintaining the composition and structure of the leachate to the maximum (only the OA species is consumed, Eqs. S1 and S2 in Supporting information). This could potentially provide the possibility for the leachate to be restored by directly evaporating the added DI-water. The ^{13}C NMR spectra of the unrepaired DES (the leachate after extracting the metal elements and evaporating the added DI-water content, Fig. 3a) shows that the intensity for the signal at 160 ppm significantly reduced and the peak at 6.5 ppm in the ^{1}H NMR spectra (Fig. S7 in Supporting information) is shifted to 5.6 ppm. The FTIR spectra (Fig. 3b) shows the intensity of the peak at 1741 cm^{-1} ($\nu_{C=O}$ of OA) for the unrepaired DES decreased. Other NMR and FTIR signals are completely consistent with the pristine DES, indicating that the content of OA in the DES is decreased while no impurities are introduced during the entire recovery process. It can be seen from the leaching reaction (Eqs. S1 and S2) that only OA is consumed during the leaching process. This indicates that DES can be repaired by adding an appropriate amount of OA to achieve

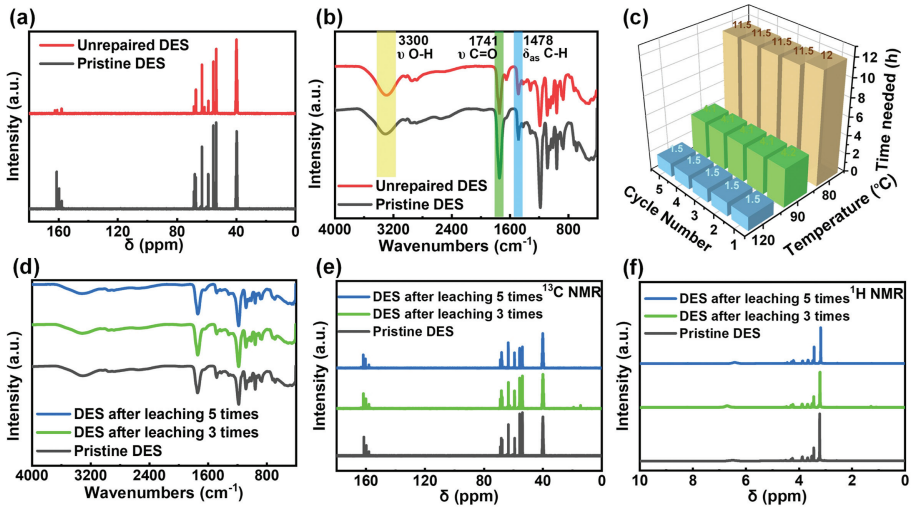

Fig. 3. Reusability of DES: (a) The ^{13}C NMR spectra and (b) FT-IR of pure DES and DES after extracting the metal elements and evaporating the DI-water (unrepaired DES), respectively. (c) The required leaching time for complete leaching under different conditions (80–120 °C, 1–5 cycles). (d) The FT-IR, (e) ^{13}C NMR spectra, and (f) ^{1}H NMR spectra of DES (has repaired) after multiple leaching cycles at 80 °C.

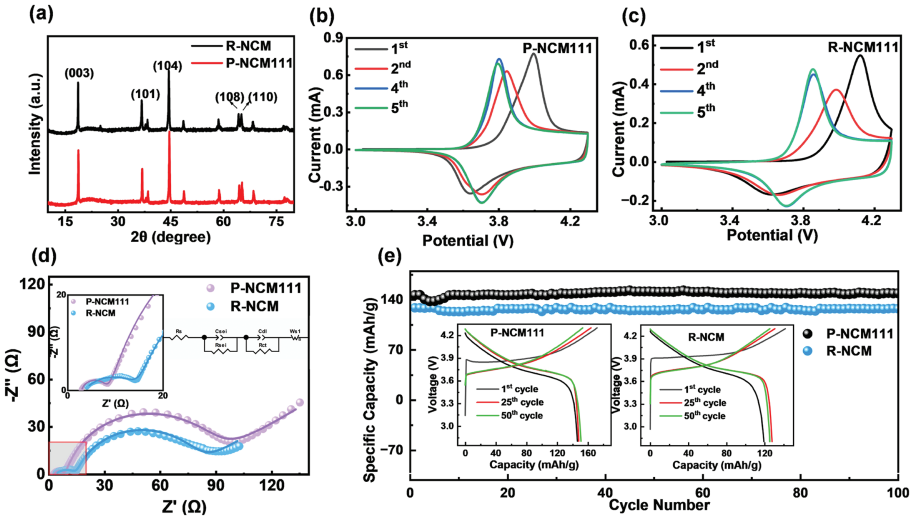

Fig. 4. Properties of regenerated NCM materials: (a) XRD spectrum. (b) Electrochemical impedance. (c, d) CV curves for the regenerated and commercial NCM111 materials (R-NCM & P-NCM111), respectively. (e) Cycle test for the R-NCM and P-NCM111.

good reusability of the leachate, and the amount of OA needed to be added was determining by the sodium hydroxide (NaOH) titration. The digital photographs of pristine DES, unrepaired DES and repaired DES are shown in Fig. S8 (Supporting information). The appearance of the unrepaired DES is completely consistent with pristine DES after adding the OA. For the repeatedly used leachate (5 cycling), the leaching efficiency (Fig. 3c), precipitation efficiency (Fig. S9 in Supporting information), FTIR and NMR spectra (Figs. 3d–f) show no observable changes, indicating ideal capability for the leachate to function repeatedly. After multiple cycles of leaching, the content of lithium ions in the leachate would gradually accumulate, providing a favorable condition to extract lithium with high efficiency. Based on the principle that lithium oxalate has low solubility within ethanol at low temperature, we added ethanol to the lithium-enriched leachate and precipitated lithium oxalate by lowering the temperature to achieve lithium extraction (Fig. S10 in Supporting information). The time required for complete leaching using leachate under different cycles is almost the same (Fig. 3c), indicating that the accumulation of lithium ions in the leachate show no observable influence on the leaching efficiency [56].

The precipitated powder can be used as the raw material to regenerate the NMC materials. TG-DSC was used to study the thermal properties of mixed crystal co-precipitation. The crystal begins to decompose around 300 °C (Fig. S11 in Supporting information) and reacts with LiOH in the range of 500–900 °C (Fig. S12 in Supporting information). Based on this, the obtained mixed crystal co-precipitation was calcined at 400 °C to obtain the precursor (Eq. S3 in Supporting information). Then, an appropriate amount of LiOH (110% the required molar ratio normalized to transition metal elements) was added and ground evenly. The mixture was then heated at 900 °C (Eq. S4 in Supporting information) for 12 h to obtain the regenerated NCM materials (R-NCM). The R-NCM and commercial pristine NCM111 materials (P-NCM111) were characterized by XPS (Fig. S13 in Supporting information) and XRD (Fig. 4a) and show identical signals, indicating the successful closed-loop recycling of the NCM materials (the impurity peak at 25° was caused by the impurity which was difficult to determine the composition). The R-NCM and P-NCM111 were assembled with lithium foil as the counter electrode into a half cell for electrochemical performance testing. The EIS (Fig. 4b) and CV (Figs. 4c and d) spectra of the R-NCM has similar redox pairs and impedance with the commercial

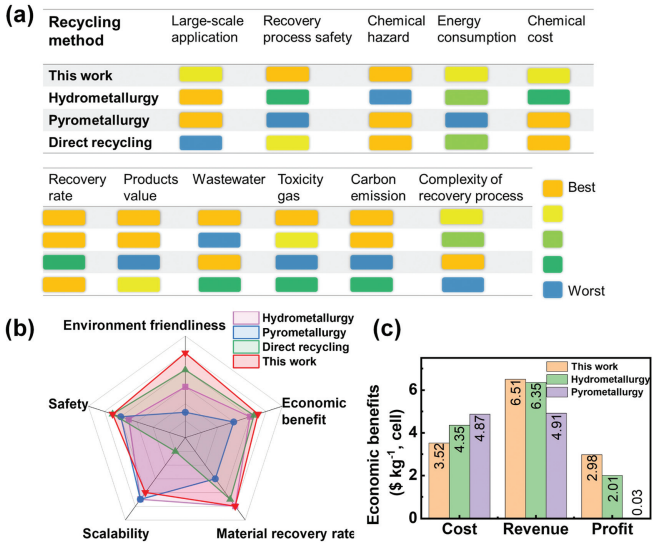

Fig. 5. Multiscale assessment of various existing road maps. (a) Comparison of advantages and disadvantages of different recycling approaches. The figure is made based on the data of Ref. [1,59–63]. (b) Comprehensive comparison of different parameters for various recycling approaches. The figure is made based on the data of Ref. [44,60–63]. (c) Economic comparison of different recycling approaches [64].

NCM111 materials, indicating the electrochemical kinetics of the recycled P-NCM is similar to the P-NCM111. As shown in (Fig. 4e), at the current density of 0.2 C and voltage range of 2.8–4.3 V, the R-NCM shows an initial discharge capacity of 128.8 mAh/g. After 100 cycles, it retains 99.1% of its initial capacity. This indicates that R-NCM has good capacity retention and high commercial value.

Based on the mixed crystal co-precipitation mechanism, we have realized the closed-loop recycling of various metal elements using reusable leachate (Fig. S14 in Supporting information) which is expected to provide various advantages. The process of this recycling strategy is simple, convenient for industrialization, and generates minimum wastewater or harmful gas. We have further performed multiscale comparison with the traditional hydrometallurgy and pyrometallurgy strategies (Figs. 5a and b) (environment friendliness, which is evaluated based on the pollution like wastewater and toxic gas emission; safety, which is evaluated based on the chemical hazard, reaction temperature, danger of manual operation and so on; economic benefit, which is evaluated based on the cost and revenue for the entire recovery process; scalability, which is derived from the potential application of these strategies in industry; and the material recovery rate is evaluate based on the experimental data). This strategy has significant advantages in terms of energy consumption (Fig. S15a in Supporting information), economic benefits (Fig. 5c) and environmental impact (Fig. S15b in Supporting information) [57–63]. The above advantages are expected to provide competent road maps for pushing the frontiers of recycling EoL-LIBs.

In summary, based on mixed crystal co-precipitation, the ternary transition metal elements in EoL-LIBs can be synergistically extracted without adding precipitants, thus realizing reusability for the leachate. We demonstrate that the OA:ChCl:EG type DES is a highly efficient leachate that can completely leach NCM111 at 120 °C for 1.5 h with $RS/L = 20$. The subsequent extraction is achieved by adding DI-water, which controllably change the reaction pathway and leads to the co-precipitation of nickel, cobalt, and manganese with a precipitation efficiency of 98.6%, 99.5%, and 96.9%, respectively. After filtrating out the solids, the leachate can be reused for the next-round leaching process after directly evaporating the DI-water and the addition of small amount of the OA component. The strategy presented here has fundamentally improved the recycling process for extracting multiple metal elements without adding precipitate to achieve reusability of the leachate, which is expected to push the frontiers for the practical recycling of EoL-LIBs with higher economic benefits.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.cclet.2024.109726.

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